Anomalous thermal expansion in Ising-like puckered sheets

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Motivated by efforts to create thin nanoscale metamaterials and understand atomically thin binary monolayers, we study the finite temperature statistical mechanics of arrays of bistable buckled dilations embedded in free-standing two-dimensional crystalline membranes that are allowed to fluctuate in three dimensions. The buckled nodes behave like discrete, but highly compressible, Ising spins, leading to a phase transition at $T_c$ with singularities in the staggered “magnetization,” susceptibility, and specific heat, studied via molecular dynamics simulations. Unlike conventional Ising models, we observe a striking divergence and sign change of the coefficient of thermal expansion near $T_c$ caused by the coupling of flexural phonons to the buckled spin texture. We argue that a phenomenological model coupling Ising degrees of freedom to the flexural phonons in a thin elastic sheet can explain this unusual response.

In recent decades, metamaterials with unique properties, such as auxetic behavior and extreme stretchability, have been realized at the macroscale [1, 2] as well as the nanoscale [3–7]. More recently, there has been growing interest in designing mechanical materials with programmable memory, using multistable buckled materials [1, 8–12] and origami [13–16].

Buckled configurations have also been found (via either first-principles simulations or experiments) in atomically thin materials such as stanene, SnO, PbS, and borophane polymorphs [17–23], as well as in graphene with topological defects or substitutional impurities [24–27]. At the nanoscale, thermal fluctuations can strongly influence any mechanical memories stored in the material as the energy barriers between bistable states become comparable to the temperature. Furthermore, thermal fluctuations also profoundly change the mechanics of atomically thin materials at long wavelengths [3, 28–33]. Yet, few studies exist on the interplay between microstructure (e.g., defects) and thermal fluctuations in these atomically thin materials.

We study here the thermal response and phase transitions of puckered sheets with square arrays of buckled positive and negative dilational defects using molecular dynamics simulations. We find that puckered membranes undergo highly compressible Ising-like phase transitions. We also observe an anomalous thermal expansion, where the typically negative coefficient of thermal expansion briefly becomes positive close to the transition, which we explain with a theoretical model coupling spin and elastic degrees of freedom. Creating a highly tunable coefficient of thermal expansion, spanning both positive and negative values, is a goal of many metamaterial design efforts, and we are not aware of any other physically realizable 2D material expected to have this property [34–36]. This unusual anomaly could, for example, be leveraged to create nanoscale device components whose dimensions are insensitive to thermal changes at a particular operating temperature.

The model.— Since ab initio molecular dynamics [38] are computationally expensive for studying phase transitions and atomistic potentials for puckered materials are not yet developed, we use a coarse-grained discrete membrane model [39], tuned to approximate an isotropic elastic sheet in the continuum limit. Nodes are connected by harmonic springs (Fig. 1(a)) and there is an energetic cost when the normals of neighboring planes are not aligned (Fig. 1(b)). The total energy, adapted from

FIG. 1. (a) Square lattice model with background sites $A$ and a single dilation site $B$. (b) Schematic of normals of two neighboring triangular plaquettes $\alpha, \beta$. (c,d) Height profiles of relaxed membranes with a single (c) positive and (d) negative dilation at $T = 0$. The color represents the height relative to the zero plane in units of the lattice spacing $a_0$. The dilation nodes are indicated with a larger radius sphere. (e,f) Top views of membranes with a square array of positive (e) and negative (f) dilations in a $(0, 2)$ array at $T = 0$. Both display a checkerboard configuration characteristic of antiferromagnetism at $T = 0$ when spins are defined as the nodes that buckle out of plane. Node positions are visualized using OVITO software [37].
ref. [39], is given by
\[ E = \frac{k}{2} \sum_{\langle i,j \rangle} |\mathbf{r}_i - \mathbf{r}_j - a_{ij}|^2 + \hat{\kappa} \sum_{\langle \alpha,\beta \rangle} (1 - \mathbf{n}_\alpha \cdot \mathbf{n}_\beta), \]

where \( k \) is the spring constant, \( \hat{\kappa} \) is the microscopic bending rigidity, and \( a_{ij} \) is the rest length between two connected nodes, \( i \) and \( j \). The first sum is over connected nodes and the second sum is over neighboring triangular planes. This model (with \( a_{ij} = a_0 \), nodes on a triangular lattice) has been shown to produce mechanical [25] and thermal properties [29, 30] as well as height-height correlation functions [40] consistent with simulation results of 2D materials (e.g., graphene and MoS\(_2\)) using atomistic potentials [25, 41, 42] (Supplemental Material (SM) Sec. VII) [43]. Furthermore, anticipating our observations of critical phenomena, we expect aspects of the behavior of the system to be insensitive to microscopic details due to universality.

In this work, the nodes are arranged on a square lattice and the dilation sites \( B \) are embedded into a background matrix of standard, undilated sites \( A \). The dilations are modeled by changing the preferred lengths of the bonds between \( A \) and \( B \) sites [11], mimicking buckled monolayers (e.g., SnO and PbS [18, 21]). The rest lengths are:
\[ a_{AA} = a_0, \quad \sigma_{AA} = a_0 \sqrt{2}, \quad a_{AB} = a_0(1 + \epsilon), \quad \sigma_{AB} = a_0 \sqrt{2(1 + \epsilon + \epsilon^2/2)}, \]
where \( \epsilon \) is the fractional change in the bond length, and \( \sigma \) denotes a diagonal bond. For pristine membranes, the corresponding continuum Young’s modulus is \( Y = 4k/3 \) and the continuum bending rigidity is \( \kappa = \hat{\kappa} \) [11]. The continuum size of the dilation is defined as \( \Omega_0 = 4a_0^2 \epsilon \) [11]. We choose microscopic elastic parameters \( a_0 = 1, k = 100, \hat{\kappa} = 1, \epsilon = \pm 0.1 \). Here we study membranes with dilations that provide positive and negative extra area (\( \Omega_0 > 0 \) and \( \Omega_0 < 0 \) respectively) with periodic boundary conditions in \( x \) and \( y \) directions. See the SM Sec. IV for details on other parameter choices. Related tethered membrane models have been studied before [44–47], but with quenched random disorder rather than regular defect arrays.

**Mapping buckled structures to Ising spins.**—We first describe the behavior of the model at \( T = 0 \). As the cost of stretching/size of the dilation increases, the system crosses a buckling threshold, and a subset of the nodes will prefer to buckle out of the plane. As shown in Fig. 1(c) and (d), the relaxed configurations of isolated buckled positive and negative dilations differ. The positive dilations create localized, peaked structures, and the negative dilations lead to saddle-like deformations. This difference can be understood by considering the angular deficit/surplus at the dilation vertex in the inextensible limit—positive dilations have a local angular deficit (discrete positive Gaussian curvature) and negative dilations have a local angular surplus (discrete negative Gaussian curvature).

Despite these differences, we can assign Ising spin variables to dense, square arrays of either positive or negative dilations. In arrays of positive dilations at \( T = 0 \), the dilations themselves buckle out of the plane (Fig. 1(e)). In arrays of negative dilations, the dilations remain in a single plane at \( T = 0 \), and sites on the lattice dual to the dilation superlattice buckle (Fig. 1(f)). We assign a spin variable of \( \pm 1 \) to each buckled site depending on whether the dilation/dual site buckles up or down. At finite temperature, we assign spins using nodes’ positions relative to the local planes formed by their neighbors to account for thermal fluctuations. With this mapping, the buckled structures shown in Figs. 1(e) and (f) are equivalent to checkerboard spin configurations, mechanical analogs of a nearest-neighbor Ising antiferromagnet (AFM). Our simulations support the conclusion that the AFM state is the lowest energy state for the buckled positive and negative dilation arrays that we study. See the SM, Sec. III and V and [11] for further discussion of the buckling threshold and the ground states of arrays.

**Finite temperature simulations.**—As we are interested in the interplay between microstructure and temperature, we perform molecular dynamics (MD) simulations of both pristine membranes and membranes with positive and negative dilation defects at finite temperature using HOOMD [48]. The membranes have \( L_N \times L_N \) nodes with \( L_N \) ranging from 24 to 192. Systems with \( L_N^2 \) nodes have \( N_f = L_N^3/4 \) dilations. Temperatures are reported in units of the bending energy (\( \hat{\kappa} = 1 \)). See the SM Sec. IV and V for more simulation details.

**Magnetic ordering and phase transitions.**—The mapping between buckled structures and Ising spins suggests we can observe a “magnetic” phase transition at finite temperature in our MD simulations. We use the staggered magnetization per spin as the order parameter
\[ m_{st} = \frac{1}{N} \sum_i s_i (-1)^{x_i + y_i}, \]
where \( s_i = \pm 1 \) is the spin on site \( i \), and \( x_i, y_i \) are the site indices on a 2D square lattice (Fig. 3d,e). Figure 2 shows \( (m_{st}^2) \) for puckered membranes as a function of \( T \). We see clearly that pronounced AFM order for \( T < 0.2 \) rapidly becomes much smaller for \( T > 0.2 \). Snapshots of spin configurations for several temperatures are shown in Fig. 2 and the SM, Sec. V. Note that in our model the bond topology remains unchanged across the temperature range studied.

In studies of critical phenomena, it is typical to measure diverging quantities such as the magnetic susceptibility \( \chi \) and specific heat \( C \). Following standard methods [50–52], we calculate the staggered susceptibility as
\[ \chi' = \frac{N_{st}}{k_B T} \langle (m_{st}^2) - (\langle m_{st} \rangle)^2 \rangle. \]
This computationally convenient quantity differs from the true susceptibility by a constant factor above the transition and does not affect the susceptibility exponents [50, 51]. See the SM, Sec. VI for details. We also calculate the specific heat per site as
\[ C = \frac{1}{N k_B T} \langle (E^2) - (E)^2 \rangle. \] This measurement uses the total potential energy, so \( N \) includes all sites.
The staggered susceptibility and specific heat of membranes with positive dilations as a function of $T$ for a wide range of system sizes are shown in Fig. 3. We see that $\chi'$ and $C$ reach maxima at $T \approx 0.2$ and the peaks increase with system size, a signature of phase transitions in finite systems. Similar results for membranes with negative dilations appear in the SM, Sec. VI. In finite systems, the correlation length $\xi \sim |T - T_c|^{-\nu}$ cannot exceed the system size and thus the diverging quantities will reach a maximum when $\xi \approx L$. Finite size scaling allows us to extract critical exponents [50–52].

Upon fitting the data with power law functions, we measure $\gamma/\nu = 1.741 \pm 0.062, \alpha/\nu = 0.068 \pm 0.018$ for $\Omega_0 > 0$ and $\gamma/\nu = 1.684 \pm 0.061, \alpha/\nu = 0.074 \pm 0.016$ for $\Omega_0 < 0$. The measurement of $\gamma/\nu$ is consistent with the rigid 2D Ising model value, 7/4. The value of $\alpha/\nu$, on the other hand, appears to be approximately four standard deviations away from 0, the 2D Ising expectation. Although our specific heat data cannot completely exclude a rigid Ising model logarithmic divergence in the specific heat, this observation suggests that the universality class is not 2D Ising. We can plausibly attribute this departure to the long-range interaction between staggered magnetization and Gaussian curvature that arises in the phenomenological model introduced in the following section. In the SM, Sec. VI, we extract $\nu$ and present data for the exponents $\alpha, \gamma,$ and $\beta$.

Anomalous thermal expansion.—The order-disorder transition has a striking effect on the thermal expansion of the membrane as a function of temperature. We first examine the thermal contraction of a pristine membrane (no dilations) to establish a point of comparison. Thermalized membranes have been studied extensively using MD and Monte Carlo simulations [28–30, 40, 41, 53, 54], and their negative coefficient of thermal expansion $\alpha_T$ has been calculated analytically [55].

$$\alpha_T = \frac{1}{A_0} \frac{dA}{dT} \approx -\frac{k_B}{4\pi \kappa} \left[ \ln \left( \frac{l_{th}}{a_0} \right) + \frac{1}{\eta} - \frac{1}{2} \right],$$  \hspace{1cm} (2)$$

where the thermal length $l_{th} = \frac{\pi}{\eta} = \sqrt{\frac{16\pi^2\kappa^2}{3T\kappa^2}}$ and $\eta$ is a universal scaling exponent describing flexural phonons, $\eta \approx 0.8$ [55]. In our simulations, we vary $T$ from 0.100 to 0.400, which varies $l_{th}$ from $3.5a_0$ to $1.8a_0$. Figure 4(a) shows the average projected area divided by the area of a flat membrane as a function of $T$. Upon computing $\alpha_T = \frac{1}{A_0} \frac{dA}{dT}$, we find excellent agreement with Eq. 2 with no free parameters (red dashed line in Fig. 4(d)), using the zero-temperature values of the bending rigidity and Young’s modulus. The pristine membrane model therefore reproduces the negative coefficient of thermal expansion of materials such as graphene [56]. In contrast, positive thermal expansion has been measured in relatively thick freestanding transition metal dichalcogenides, possibly due to a higher bending rigidity suppressing flexural phonons [42, 57–59].

In contrast, $\langle A \rangle / A_0$ (and hence $\alpha_T$) for puckered mem-
FIG. 4. Top row: Normalized area ⟨A⟩/A0 as a function of T for (a) pristine membranes, (b) membranes with positive dilations, and (c) membranes with negative dilations for LN = 120. ⟨A⟩/A0 decreases with increasing T for pristine membranes whereas ⟨A⟩/A0 for puckered membranes shows non-monotonic behavior. Bottom row: The coefficient of thermal expansion αT as a function of T for (d) pristine membranes and (e) membranes with dilations. The theoretical prediction of αT for pristine membranes with no adjustable fitting parameters matches very well with simulations (red dashed line). Far below Tc, αT for membranes with dilations is negative, as for pristine membranes. Close to Tc, αT increases rapidly and reaches a positive value, decreasing again to a negative value for T well above Tc.

branes shows non-monotonic behavior. Here, the constant factor A0 is the projected area of the lowest energy state at T = 0, a buckled checkerboard as described above. We observe that, while there is shrinkage for T < Tc, as for a pristine membrane, the value of αT is less strongly negative. For T ≪ Tc, αT puckered/αT pristine ∼ 0.5, suggesting that membranes with ordered puckers stiffen. This observation is consistent with a theoretical argument based on [60], treating the buckled dilation texture as a frozen background metric (SM, Sec. II). The calculation predicts the existence of an increased bending rigidity at T = 0, κμ ≈ κ(1 + 3h0/2μ), where h0 is the amplitude of the buckled membrane. Close to the transition, however, αT increases rapidly and eventually reaches a positive value. Evidently, the swelling due to disordered up and down puckers on all length scales near Tc dominates the entropic shrinkage present in pristine sheets [55–57].

Phenomenological model. — To better understand the observed differences between the thermal expansion of pristine membranes and membranes with dilations, we introduce a “flexural Ising model,” with an effective free energy that couples an Ising order parameter to a thin elastic sheet that is allowed to fluctuate both in and out of the plane. We assume coarse-graining such that the short wavelength, impurity-scale phonons are accounted for by a staggered pucker order parameter mst, which interacts with a long wavelength nonlinear strain matrix, uij.

\[
F = \int d^2x \left[ \frac{\kappa}{2} (\nabla^2 f)^2 + \mu u_{ij}^2 + \frac{\lambda}{2} u_{kk}^2 + \frac{K}{2} (\nabla m_{st})^2 + \frac{r}{2} m_{st}^2 + g m_{st} u_{kk} \right],
\]

where uij is related to in-plane displacements uij and out-of-plane displacements f by

\[
u = \frac{1}{2} \left( \frac{\partial u_{ij}}{\partial x_i} + \frac{\partial u_{ij}}{\partial x_j} + \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_j} \right).
\]

The term proportional to g is the lowest order contribution allowed by symmetry coupling the phonon and order parameter fields. Similar free energies have been used to study flat compressible 2D Ising models in the limit f = 0 [62, 63]. We also note similarities to free energies used to model electron-phonon interactions in graphene [64–67].

Upon tracing out the in-plane phonons according to standard methods [62, 63, 68], Eq. 3 becomes:

\[
F_{eff} = \frac{g^2}{2A_0} \left( \frac{1}{2\mu + \lambda} - \frac{1}{\mu + \lambda} \right) \left( \int d^2x m_{st}^4 \right)^2 + \int d^2x \left( \frac{1}{8} \left[ P_{ij}^T \partial_i f \partial_j f \right]^2 + \frac{g\mu}{2\mu + \lambda} \left( m_{st}^2 P_{ij}^T \partial_i f \partial_j f \right) \right)
\]

\[
+ \int d^2x \left[ \frac{\kappa}{2} (\nabla^2 f)^2 + \frac{K}{2} (\nabla m_{st})^2 + \frac{r}{2} m_{st}^2 + \left( u - \frac{g^2}{2(2\mu + \lambda)} \right) m_{st}^4 \right],
\]

where Pij is the transverse projection operator and the primed integral omits q = 0 modes. Equation 4 has three terms that are not present for either pristine membranes or the Ising model. The first and final terms, proportional to g2, also appear for flat compressible 2D Ising models [62, 69]. The term proportional to g, however, is unique to the flexural Ising model. Since the Laplacian of \(-\frac{1}{2} P_{ij}^T \partial_i f \partial_j f\) is the Gaussian curvature S(x) in the Monge representation [70], this term represents a long-range interaction between the squared staggered magnetization and the Gaussian curvature of the form \(-\frac{1}{2\pi} \int d^2x \int d^2x' m_{st}^2(x) S(x') \ln |x - x'| \). A power counting argument suggests that the coefficient w = \(\frac{g\mu}{2\mu + \lambda}\) is a strongly relevant operator. We plan to examine the behavior of w in more detail in future work.

We can calculate the coefficient of thermal expansion αT by adding an in-plane pressure, and compare to the
simulation data in Fig. 4. As shown in Sec. I of the SM, we find the average change in area
\[ \langle \delta A \rangle = -gA_0\left(\frac{m^2}{\mu + \lambda}\right)_0^T - \frac{A_0}{2} \left( \frac{\partial f}{\partial x_i} \right)^2, \] (5)
and coefficient of thermal expansion
\[ \alpha_T = \left. \frac{1}{A_0} \frac{dA}{dT} \right| = \frac{d}{dT} \left( \frac{g(m^2)}{\mu + \lambda} \right) - \frac{d}{dT} \left( \frac{1}{2} \left( \frac{\partial f}{\partial x_i} \right)^2 \right). \] (6)

We expect that the microscopic couplings \( g \) and \( \mu + \lambda \) depend only weakly on temperature over the temperature range of interest, provided we are far below the high temperature crumpling transition. Therefore, the contribution from the first term is sharply peaked around \( T_c \), given the results in Fig. 2. We expect \( g > 0 \), as the antiferromagnetic state has a smaller projected area than the ferromagnetic state at \( T = 0 \), consistent with the positive peak in \( \alpha_T \) at \( T_c \) observed in Fig. 4(e). The second term is the usual entropic thermal shrinkage, also present for a pristine membrane [55].

**Conclusion.**—We observe a phase transition in the staggered magnetization of a puckered membrane, which provides a mechanical analog of a highly compressible antiferromagnetic Ising model. Furthermore, we find that the order-disorder transition produces an anomalous thermal response for puckered membranes. These observations suggest a strong coupling between flexural phonons and the ordering of the spins (buckled sites). We introduce a phenomenological “flexural Ising model” that anticipates a competing effect between entropic shrinkage due to out-of-plane deformations and swelling due to puckler disorder at the phase transition.

Our findings suggest that bistable buckled structures change the thermal response of 2D materials, leading to a tunable coefficient of thermal expansion. The ability to tune thermal expansion is important for combining different materials, as mismatched thermal expansion can affect the longevity of integrated materials [71]. Materials with tunable thermal expansion are rare and often require precise engineering [34].

Since the phase transition temperature in our model depends on the elastic constants of the host lattice and the separation between dilations, one could imagine constructing a nanocantilever or nanoactuator out of a puckered membrane designed to be insensitive to thermal expansion/shrinkage at the temperature at which it must work (\( \alpha_T = 0 \) at two temperatures, one above and one below \( T_c \)). Additionally, the mechanism of an inefficient packing of buckled structures resulting in a global expansion can be applied to macroscale materials with multi-stable units [10, 12, 35]. Moreover, our work suggests the possibility of studying novel universality classes in 2D materials with a coupling between spin and both in-plane and out-of-plane displacements, generalizing past work on compressible Ising models to include flexural phonons.

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See Supplemental Material [url] for details on theoretical derivations, extracting critical exponents, simulations and a supplementary video, which includes Refs. [72–78].

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## Supplemental Material

### Contents

| Section                                                                 | Page |
|------------------------------------------------------------------------|------|
| I. Phenomenological model                                              | 1    |
| II. Stiffening of puckered surfaces for $T = 0$                         | 3    |
| III. Buckling threshold and eigenvectors for negative dilations        | 6    |
| IV. Molecular dynamics simulations                                      | 8    |
| V. Equilibration                                                       | 10   |
|   A. Ground states at $T = 0$                                          | 10   |
|   B. Thermal equilibration for $T > 0$                                 | 10   |
| VI. Critical exponents                                                 | 14   |
|   A. Susceptibility $\chi$ and critical exponent $\gamma$             | 16   |
|   B. Specific heat $C$ and critical exponent $\alpha$                  | 19   |
|   C. Critical temperature $T_c$ and correlation length exponent $\nu$  | 21   |
|   D. Magnetization $m_{st}$ and critical exponent $\beta$             | 23   |
| VII. Comparison of fluctuations in empirical graphene model and membrane model | 23   |
| References                                                             | 25   |

Here, we provide theoretical derivations, as well as procedures for performing molecular dynamics simulations and extracting critical exponents.

### I. Phenomenological model

We calculate here the ensemble-averaged area change $\langle \delta A \rangle$ using our phenomenological model. To do so, we add an in-plane pressure term proportional to $\alpha$ to Eq. 3 of the main text the form

$$-\alpha \int d^2x \left( \frac{\partial u_k}{\partial x_k} \right) \equiv -\alpha \int d^2x \tilde{u}_{kk}. \quad (S1)$$
The change in the projected area is
\[ ⟨\delta A⟩ = \int \mathcal{D}u \int \mathcal{D}f \int \mathcal{D}m_{st} \left( \int d^2x \tilde{u}_{kk} \right) e^{-\beta F} = \frac{\int \mathcal{D}u \int \mathcal{D}f \int \mathcal{D}m_{st} \left( k_B T \frac{\partial}{\partial \alpha} e^{-\beta F} \right)}{\int \mathcal{D}u \int \mathcal{D}f \int \mathcal{D}m_{st} e^{-\beta F}} \]
\[ = \frac{k_B T}{Z} \frac{\partial Z}{\partial \alpha} = k_B T \frac{\partial \ln Z}{\partial \alpha}. \tag{S2} \]

We will evaluate this expression at \( \alpha = 0 \) to understand our simulations with tension-free periodic boundary conditions.

We now simplify \( Z \) by integrating out in-plane phonons so that our result can be expressed in terms of \( m_{st}(x) \) and the flexural phonon field \( f(x) \). We Fourier transform the free energy of Eq. 3, using the conventions \( \xi(x) = \sum_q \xi(q)e^{iq\cdot x} \) and \( \xi(q) = \frac{1}{2} \int \! d^2x \xi(x)e^{-i\mathbf{q}\cdot \mathbf{x}} \), and then separate the (nonlinear) strain tensor into \( q = 0 \) and \( q \neq 0 \) components.

\[ u_{ij}(x) = u^0_{ij} + A^0_{ij} + \sum_{\mathbf{q} \neq 0} \left( \frac{i}{2} (q_i u_j(q) + q_j u_i(q)) + A_{ij}(\mathbf{q}) \right) e^{i\mathbf{q}\cdot \mathbf{x}}, \tag{S3} \]

where \( A_{ij}(x) = \frac{1}{2} \left( \frac{\partial f}{\partial x_i} \frac{\partial f}{\partial x_j} \right) \). Since the Fourier transform of the pressure term only depends on \( u^0_{ij} \), we only need to consider the \( q = 0 \) modes of the terms in the free energy that depend on \( u_{ij} \) to evaluate Eq. S2 as a function of \( f \) and \( m_{st} \). Note that \( u^0_{ij} \) has three independent degrees of freedom, unlike \( \frac{1}{2} (\partial_i u_j + \partial_j u_i) \) evaluated at finite wavevector.

Upon defining
\[ \Psi(x) = m_{st}(x)^2, \tag{S4} \]
the \( q = 0 \) mode of the free energy (suppressing the pure Ising terms, which do not depend on \( u_{ij} \)) is

\[ F^0 = A_0 \left( \mu (u^0_{ij} + A^0_{ij})^2 + \frac{\lambda}{2} (u^0_{kk} + A^0_{kk})^2 + g\Psi(0) (u^0_{kk} + A^0_{kk}) - \alpha u^0_{kk} \right). \tag{S5} \]

We now shift \( u^0_{ij} \), defining \( u^0_{ij} = w^0_{ij} - A^0_{ij} - \frac{g\Psi(0) - \alpha}{2(\mu + \lambda)} \delta_{ij} \) [1]. This change of variables produces a quadratic function of \( w^0_{ij} \) that is easily integrated out. Our remaining \( q = 0 \) free energy, following the integration over in-plane phonons, is

\[ F^0 = \frac{A_0}{2(\mu + \lambda)} \left( -g^2\Psi(0)^2 + 2\alpha g\Psi(0) - \alpha^2 \right) + \alpha A_0 A^0_{kk}. \tag{S6} \]
Equation S2 evaluated at $\alpha = 0$ then gives

$$\langle \delta A \rangle = -\frac{g_0 A_0 \langle m_{st}^2 \rangle}{\mu + \lambda} - \frac{A_0}{2} \left\langle \frac{\partial f}{\partial x_k} \frac{\partial f}{\partial x_k} \right\rangle,$$

(S7)

where $A_0$ is the projected area of the buckled system at $T = 0$.

A complete understanding of the behavior of $\langle f(q)f(-q) \rangle$ and $\langle m_{st}(q)m_{st}(-q) \rangle$ would require evaluating the thermal averages with the effective free energy given by Eq. 4. However, as discussed in the main text, this model is still able to shed light on the simulation data for $\langle A \rangle / A_0$ in Fig. 4 and the observed divergence of the coefficients of thermal expansion.

II. Stiffening of puckered surfaces for $T = 0$

Using the techniques of ref. [2], we argue here that the corrugations associated with antiferromagnetically buckled impurities lead to an increase in the effective membrane bending rigidity at $T = 0$. Following shallow shell theory [3], we assume that there is a zero energy reference state described by

$$r_0(x_1, x_2) = (x_1, x_2, h(x_1, x_2)).$$

(S8)

In refs. [4, 5], among others, $h(x_1, x_2)$ describes a shallow section of a sphere, for example. For that case, $h(x_1, x_2) = R \left(1 - \sqrt{1 - \frac{x_1^2}{R^2} - \frac{x_2^2}{R^2}}\right)$. We instead assume that our reference metric corresponds to a checkerboard pattern of up and down puckers, like an egg carton. Thus, deviations from this checkerboard egg carton ground state [6] cost energy. This description should be accurate provided fluctuations are not able to invert any of the buckled impurities, as will be the case for $T \ll T_c$.

To describe deviations from the reference state, which cost energy, we separate the deviations into two components tangential to the reference surface and one normal to the reference surface,

$$r = r_0 + u_t \hat{t}_i + f \hat{n}^0.$$

(S9)

To extract the bending rigidity at $T = 0$, we also allow for a position-dependent force/area difference $p(x)$ to act across the membrane, with an energetic cost proportional to $f(x)$, the displacement in the direction normal to the reference state. Ref. [2] derives a self-consistent
equation for the linear response of the membrane in the presence of small $p$,

$$f(q) = \frac{p(q)}{\kappa q^4} - \frac{Y}{\kappa q^4} \sum_{q', q'' \neq 0} \frac{(q \times q')^2(q' \times q'')^2}{q'^4} h(q - q') h(q' - q'') f(q'').$$  \hspace{1cm} (S10)$$

Following [2], we assume that the term proportional to $Y/\kappa$ can be treated as a perturbation, and solve iteratively by inserting $f(q) = \frac{p(q)}{\kappa q^4}$ in the right hand side to get the first order correction, linear in $p(q)$,

$$f(q) \approx \frac{p(q)}{\kappa q^4} - \frac{Y}{\kappa q^4} \sum_{q', q'' \neq 0} \frac{(q \times q')^2(q' \times q'')^2}{q'^4} h(q - q') h(q' - q'') \frac{p(q'')}{\kappa q'^4}.$$

The substitutions

$$q' = q - q_1,$$  \hspace{1cm} (S12)$$q'' = q - q_1 - q_2,$$  \hspace{1cm} (S13)

lead to

$$f(q) \approx \frac{p(q)}{\kappa q^4} - \frac{Y}{\kappa^2 q^4} \sum_{q_1, q_2} \frac{(q \times q_1)^2((q - q_1) \times q_2)^2}{(q - q_1)^4} h(q_1) h(q_2) \frac{p(q - q_1 - q_2)}{(q - q_1 - q_2)^3}.$$  \hspace{1cm} (S14)

We now parametrize the deflections induced by the buckled impurities. The nonzero Fourier modes compatible with the checkerboard reference state can be found by direct calculation as in ref. [6], and are

$$B(m_1, m_2) = \left(\frac{(2m_1 + 1)\pi}{n a_0} \hat{x} + \frac{(2m_2 + 1)\pi}{n a_0} \hat{y}\right),$$  \hspace{1cm} (S15)$$

where $m_1$ and $m_2$ are integers and $na_0$ is the real space distance between impurities when $\epsilon = 0$.

Consider a long wavelength pressure $p(x) = \sum q p(q) e^{i q \cdot x}$, whose largest magnitude wave vector component is much smaller than the smallest wave vector describing the checkerboard pattern. For this case, the correction term will not contribute unless $q_1 = -q_2$. With this
condition, Eq. S14 simplifies to

\[ f(q) \approx \frac{\rho(q)}{\kappa q^4} \left( 1 - \frac{Y}{\kappa q^4} \sum_i \frac{(q \times q_i)^4 |h(q_i)|^2}{(q - q_i)^4} \right) \approx \frac{\rho(q)}{\kappa q^4} \left( 1 + \frac{Y}{\kappa q^4} \sum_i \frac{(q \times q_i)^4 |h(q_i)|^2}{(q - q_i)^4} \right), \]  

(S16)

where the sums are over wave vectors given by Eq. S15. We can now extract an effective bending rigidity with a correction that is strictly positive for a given pressure wave vector \( q \).

\[ \kappa_R(q) \approx \kappa \left( 1 + \frac{Y}{\kappa q^4} \sum_i \frac{(q \times q_i)^4 |h(q_i)|^2}{(q - q_i)^4} \right). \]  

(S17)

Thus, at long wavelengths, the buckled defect texture stiffens the membrane. The amount of stiffening depends on the direction and wavelength of the pressure being applied, as well as the details of the buckled defect structure encoded in \( h(q) \). We can simplify the expression further by applying the assumption \( q \ll q_i \) for all \( q_i \), which leads to

\[ \kappa_R(q) \approx \kappa \left( 1 + \frac{Y}{\kappa} \sum_i \sin^4 \theta_i |h(q_i)|^2 \right). \]  

(S18)

where \( \theta_i \) is the angle between \( q \) and \( q_i \).

If we assume that the buckled dilation height field is well approximated by the eigenvectors derived in Sec. III using the smallest four wave vectors given by Eq. S15 as a basis,

\[ h(x, y) = h_0 \cos \left( \frac{\pi x}{na_0} \right) \cos \left( \frac{\pi y}{na_0} \right), \]  

(S19)

for positive dilations, and

\[ h(x, y) = h_0 \sin \left( \frac{\pi x}{na_0} \right) \sin \left( \frac{\pi y}{na_0} \right), \]  

(S20)

for negative dilations, we can estimate the size of the correction. Upon carrying out an angular average over possible orientations of \( q \), we have

\[ \kappa_R \approx \kappa \left( 1 + \frac{Y h_0^2}{16 \kappa} \langle \sum_i \sin^4 \theta_i \rangle \right) \approx \kappa \left( 1 + \frac{3Y h_0^2}{32 \kappa} \right). \]  

(S21)

Close to the inextensible limit, we have \( h_0^2 \sim \Omega_0 \), the change in the surface area due to
the dilation. If we are instead close to the buckling transition, $h_0^2 \sim a_0^2 (\gamma - \gamma_c)$ [6]. For the parameters used in our simulations, $Y = 400/3, \kappa = 1$, and $h_0$ is measured to be approximately $0.4a_0$. In this case,

$$\frac{3Yh_0^2}{32\kappa} \approx 12.5\frac{h_0^2}{a_0^2} \approx 2.$$

(S22)

Therefore, according to this argument, we expect significant increases to the bending rigidity of the membrane due to the corrugated background of buckled dilations. Since this correction is large, violating the assumptions of our perturbation theory, we would need a more sophisticated calculation to estimate its magnitude more accurately.

III. Buckling threshold and eigenvectors for negative dilations

The continuum theory introduced in ref. [6] to treat positive dilations at $T = 0$ can also be applied to the case of negative dilations. We present these results here for checkerboard arrays composed of either positive or negative dilations. A high level overview of the calculation is given below; detailed explanations of the steps can be found in ref. [6].

We start with the energy in the continuum limit minimized with respect to in-plane displacements,

$$E = \frac{1}{2} \int d^2 r \left( \kappa (\nabla^2 f)^2 + Y \left( \frac{1}{2} P_{\alpha\beta} \partial_\alpha f \partial_\beta f \right)^2 - Y \frac{\Omega_0}{2} P_{\alpha\beta} \partial_\alpha f \partial_\beta f c(r) \right),$$

(S23)

where the density of dilational impurities at positions $\{r_j\}$ is given by $c(r) = \sum_j \delta^2(r - r_j)$ [6]. Note that the area added/subtracted by each dilation is explicitly included in the elastic model here, in contrast to the phenomenological model of Sec. I. We Fourier transform the height field, noting that the displacements are real ($f^*(q) = f(-q)$). We can now consider a particular pattern of defect buckling (ferromagnetic, checkerboard, striped, etc.). For each pattern, there is a subspace of wave vectors compatible with its periodicity that can be used as a basis for the height field. For example, the height field of the ferromagnetic state (equivalently, a single defect with periodic boundaries) can be written as a sum over the reciprocal lattice vectors of the defect superlattice.

As a first approximation, we truncate the wave vector basis so that it includes the vectors
with the smallest magnitudes that allow the impurities to couple to the flexural phonons. Since deformations are quite long range close to the buckling threshold [6], these longer wavelength modes are particularly important. We then write the quadratic contribution to the energy in matrix form, and diagonalize the energy matrix to find the eigenvalues and eigenvectors in reciprocal space. Next, we determine which eigenvalue first attains a negative value as the magnitude of $Y\Omega_0$ is increased—this corresponds to the deformation for which the flat state of the system is first unstable. The positive quartic term ensures the existence of stable states with $f \neq 0$ beyond the buckling threshold.

We find the first eigenvectors to become unstable for both positive and negative dilations in a checkerboard array in Fig. S1. These eigenvectors are shifted by a uniform translation relative to each other in position space, similar to what is seen in simulations of (0, 2) arrays.

![FIG. S1. Contour plots of the first unstable eigenvector in the continuum elastic analysis in real space using a basis of four wave vectors. Dilation nodes are located at the black dots. Negative deflections are blue, and positive deflections are red. The magnitude is arbitrary. (a) Positive $\Omega_0$. (b) Negative $\Omega_0$.](image)

In this approximation, both eigenvectors become unstable at the same magnitude of $\gamma_c$,

$$\gamma_c = \frac{Y|\Omega_0^c|}{\kappa} = 4\pi^2$$  \hspace{1cm} (S24)

To more accurately estimate the buckling thresholds, we increase the number of Fourier modes included in the basis, and extrapolate to the continuum limit. The results from these
extrapolations are shown in Fig. S2: Positive dilations buckle earlier than negative dilations as the resolution of the calculation is increased beyond the initial truncation, which agrees qualitatively with the simulations of (0, 2) superlattices presented in this paper. To pursue more quantitative agreement between the continuum theory and simulations for negative dilations, we would need to find the buckling threshold as a function of dilation separation, as we did for positive dilations in Fig. 19 of ref. [6]. We expect this continuum model is more accurate when dilations are further apart (and can be reasonably approximated as δ-function strains).

![Graph](image)

**FIG. S2.** Variation of the buckling threshold $\gamma_c$ as we increase the number of Fourier modes included in the calculation. $G_{\text{max}}$ is the magnitude of the largest Fourier mode in the basis, measured in units of $2\pi/d$, where $d$ is the separation between dilations in real space. $G_{\text{max}}^{-1} \to 0$ corresponds to the continuum limit, in which case an infinite number of Fourier modes are included. Linear extrapolations to the continuum limit are shown by the grey lines.

### IV. Molecular dynamics simulations

We study “magnetic” ordering of membranes with dilations that provide positive and negative extra area ($\Omega_0 > 0$ and $\Omega_0 < 0$ respectively) at finite temperatures with periodic boundary conditions in $x$ and $y$ directions. The energy, adapted from ref. [7] and described
in the main text, is given by

$$E = \frac{k}{2} \sum_{\langle i,j \rangle} \left( |\mathbf{r}_i - \mathbf{r}_j| - a_{ij} \right)^2 + \hat{\kappa} \sum_{\langle \alpha,\beta \rangle} (1 - \mathbf{n}_\alpha \cdot \mathbf{n}_\beta),$$

(S25)

where $k$ is the spring constant, $\hat{\kappa}$ is the microscopic bending rigidity, and $a_{ij}$ is the rest length between two connected nodes, $i$ and $j$. The first sum is over connected nodes and the second sum is over neighboring triangular planes.

For pristine membranes, the corresponding continuum Young’s modulus is $Y = 4k/3$ and the continuum bending rigidity is $\kappa = \hat{\kappa}$ [6]. The continuum size of the dilation is defined as $\Omega_0 = 4a_0^2\epsilon$ [6]. In ref. [6], it was shown that $\Omega_0 = 4a_0^2\epsilon$ for positive dilations. In fact, this relation holds for negative dilations as well, which can be confirmed by observing that the argument of [6] does not depend on the sign of $\epsilon$, and with a numerical check.

We choose microscopic elastic parameters $a_0 = 1, k = 100, \hat{\kappa} = 1, \epsilon = \pm 0.1$, corresponding to dilation Föppl-von Kármán number [6] $\gamma = \frac{Y|\mathbf{R}_a|}{\kappa} \approx 53$, and place neighboring dilations two lattice spacing apart. For these parameters, positive and negative dilation arrays are structurally similar. The arrays are dense and we are close enough to the buckling threshold ($\gamma_c \approx 21$ for positive dilations, $\gamma_c \approx 26$ for negative dilations) that the out-of-plane deformations of neighboring dilations overlap strongly, producing strong interactions.

We run molecular dynamics (MD) simulations with HOOMD [8] on NVIDIA Tesla V100 GPU and we accumulated roughly 1 terabyte of data. The simulations at finite temperatures are carried out within the $NPT$ ensemble (fixed number of particles $N$, pressure $P$, and temperature $T$) at zero pressure. The NPT integration is carried out via Martyna-Tobias-Klein barostat-thermostat with a time step $dt = 0.001$. For zero-temperature structural relaxation, we use Fast Inertial Relaxation Engine (FIRE) algorithm [8, 9] with a step size $dt = 0.005$, force and energy convergence criteria of $10^{-6}$ and $10^{-10}$, respectively.

We simulate membranes with $L_N \times L_N$ nodes on a square lattice with $L_N = 24, 36, 48, 60, 80, 96, 120, 160, 192$; these values correspond to membranes with a total number of sites ranging from 576 to 36864. Dilations are placed on a square lattice two unit spacings $(2a_0)$ apart along $x$ and $y$ directions to form a $(0, 2)$ tiling. We denote $L_1 \equiv L_N/2$ as the number of “spins” (buckled sites) along one axis which is the linear dimension of our mechanical antiferromagnet.

The coarse-grained model with no dilations [7] has been used to study the mechanical
and thermal properties of 2D materials (e.g. graphene) [10–14]. We fix $\kappa = 1.0$ and $k = 100$ and vary $T$ from 0.1 to 0.4. At each $T$, snapshots of positions are taken every 10,000 steps and a total run of $10^7$ steps is performed. For systems with $L_N > 96$ we perform $2 \times 10^7$ steps. For each $T$ we perform at least 10 independent runs—near $T_c$, we perform 15 to 50 independent runs. Longer runs and more independent runs are required for larger systems. The first half of the snapshots are discarded to allow for thermal equilibration.

V. Equilibration

A. Ground states at $T = 0$

To probe the ground state of our mechanical analog of an Ising AFM, we first performed structural relaxation on a system with $L_N = 4$ which has 2x2 dilations, with energy minimization and cell optimization at $T = 0$ via the Fast Inertial Relaxation Engine (FIRE) algorithm. The cell optimization allows the lattice parameters to deform during minimization to achieve a zero stress state. We initialize the buckled sites of positive dilations ($\Omega_0 > 0$) with two different initial conditions (i) AFM and (ii) FM with some noise. In both cases, the final relaxed configurations indicate antiferromagnetic buckled order, as found by ref. [6].

For $\Omega_0 < 0$, we also initialize the buckled background sites with AFM and FM states. We find that the relaxed state for the AFM initialization has a total energy per “spin” $E/N_I = 0.6475$. In contrast, when we initialize the system with an FM state, the final configuration has 3 “spins” pointing in the same direction, and $E/N_I = 0.8575$. Thus, for both positive and negative dilations the AFM state has the lower energy. In the following section, we also find a robust AFM ground state by quenching a large system ($L_N = 96$, 48x48 dilations) initialized with a disordered configuration to a low temperature ($T = 0.1$) using molecular dynamics.

B. Thermal equilibration for $T > 0$

In this section, we investigate the equilibration of puckered membranes at low and high temperatures, $T = 0.1, 0.3$. To test equilibration, we compare systems initialized with two
FIG. S3. (a) Snapshots of all nodes at low and high temperatures and the corresponding “spin” configurations. The heat map shows the height of each node relative to the $xy$-plane and scaled by the maximum and the minimum value at a given snapshot. In the spin configuration snapshots, black pixels represent upward buckles and white pixels represent the downward buckles. (b) Schematic side view of a chain of buckled sites (spins). Because of the curvatures due to thermal fluctuations, a local frame formed by neighboring nodes is used to define up and down spins (buckling direction).

different configurations: (i) the known AFM ground state at $T = 0$ and (ii) a disordered state. Here, we use 48x48 dilations hosted on a 96x96 square lattice ($L_N = 96$). For initialization with the AFM configuration, we first set the heights of impurities following AFM ordering and then add some small random deformations to all nodes. We then performed energy and stress minimizations at $T = 0$ using the FIRE algorithm. MD is implemented
following this relaxation. For disordered initializations, we set the heights of all nodes with random numbers.

Figure S3(a) shows snapshots of positions of all nodes at $T = 0.1$ and $T = 0.3$. The heat map indicates the position of a node relative to the reference $xy$-plane ($z = 0$). Based on the height profiles, we see that thermal fluctuations excite the out-of-plane mountains and valleys associated with flexural deformations, also present in membranes without dilations. Since the membrane is no longer perfectly flat at non-zero $T$, we need to use a local reference frame (formed by local neighbors) to identify whether a dilation is in fact buckled up or down (see Fig. S3(b)). To assign up and down spins to buckled sites at finite temperature, we use the nodes’ positions relative to the local planes formed by their neighbors. This procedure accounts for thermal fluctuations relative to the zero plane, which can be much larger than the typical dilation buckling amplitude in this highly compressible medium (approximately $0.4a_0$).

Using the filtering described above, we are able to map the positions of the buckled sites to spin configurations (see Fig. S3(a)). To study magnetic ordering in this system, we use the standard stagger operator to calculate the staggered magnetization per spin

$$m_{st} = \frac{1}{N_I} \sum_{i} s_i (-1)^{x_i + y_i}$$  \hspace{1cm} (S26)

where $N_I$ is the total number of dilations, $s_i$ is the dilation’s “spin” and $x_i, y_i$ are the site indices on a 2D square lattice.

Next, we monitor the total potential energy $E$ and the staggered magnetization per spin $m_{st}$ to check if the system is in equilibrium. Figure S4((a)-(d)) shows $E$ and $m_{st}$ as a function of time for systems prepared in both an AFM and a disordered state. At high $T$, we see $E$ and $m_{st}$ of the disordered and AFM states equilibrate after $\sim 5 \times 10^5$ MD steps. At low $T$, the systems prepared with an AFM state relaxes after $\sim 5 \times 10^5$ MD steps, whereas the system prepared with a disordered state relaxes after $\sim 1 \times 10^7$ MD steps. Similar results are found for membranes with negative dilations, shown in Fig. S5. We can see that for both cases the AFM state is robust even at finite temperature, provided $T < T_c \simeq 0.2$.

We could start with disordered states (high $T$) and quench the systems to low $T$ to initialize all simulations; however, this would take too many computing resources as the equilibration time is quite long at low $T$. To run $2 \times 10^7$ MD steps, it takes 1 hour using
FIG. S4. The total potential energy $E$ and the staggered magnetization per spin $m_{st}$ for membranes with $\Omega_0 > 0$ as a function of MD steps at high (a, b) and low (c, d) temperatures. The simulations were initialized in both a disordered state and an AFM configuration.

one GPU or $\sim$ 30 hours using one CPU. Since we know that the ground state at $T = 0$ and the equilibrated state at low $T$ (well below the critical temperature) is a mechanical analog of an Ising AFM state, we will use the AFM configurations (with some small noise) as the initial conditions for all simulations.
FIG. S5. The total potential energy $E$ and the staggered magnetization per spin $m_{st}$ for membranes with $\Omega_0 < 0$ as a function of MD steps at high (a, b) and low (c, d) temperatures. The simulations were initialized in both a disordered state and an AFM configuration.

VI. Critical exponents

In any system of finite dimensions, the correlation length $\xi$ cannot exceed the system size $L$ and divergent quantities, such as staggered antiferromagnetic susceptibility $\chi$ and
specific heat $C$, will reach a maximum at a pseudocritical temperature $T_c(L)$. Note that unlike the usual incompressible nearest neighbor 2D Ising model we do not know the true critical temperature ($T_c(L \to \infty)$) in the thermodynamic limit. We use a standard finite-size scaling (FSS) approach to extract critical exponents, similar to methods used to determine critical exponents from Monte Carlo (MC) simulation studies of Ising models and related models \[15, 16\]. Since $\xi \sim \left| T - T_c \right|^{-\nu}$ and $\xi_{T=T_c(L)} \simeq L$, we can rewrite the temperature dependence of susceptibility, specific heat, and magnetization at $T_c(L)$ in terms of system size,

$$
\chi \sim L^{\gamma/\nu}, \\
C \sim L^{\alpha/\nu}, \\
\sqrt{m^2} \sim L^{-\beta/\nu}.
$$

Using this finite size scaling approach with extensive simulations for $L_N = 24, 36, 48, 60, 80, 96, 120, 160$ and 192 we obtain $\gamma/\nu = 1.741 \pm 0.062$, $\alpha/\nu = 0.068 \pm 0.018$, $\beta/\nu = 0.127 \pm 0.038$, and $\nu = 0.99 \pm 0.09$ for $\Omega_0 > 0$ and $\gamma/\nu = 1.684 \pm 0.061$, $\alpha/\nu = 0.074 \pm 0.016$, $\beta/\nu = 0.111 \pm 0.036$, and $\nu = 1.05 \pm 0.25$ for $\Omega_0 < 0$.

The critical exponents of membranes with positive and negative dilation arrays and the exact critical exponents of the 2D Ising model are tabulated in Table I. Note that we directly measured $\gamma/\nu$, $\alpha/\nu$, and $\beta/\nu$ using finite size scaling. In order to calculate $\gamma$, $\alpha$, and $\beta$, we need to use $\nu$ obtained from other measurements. We use a power law fitting in the form of $L = c_0 |T_c(L) - T_c(\infty)|^{-\nu}$ to extract $\nu$. We excluded the two smallest systems ($L_N = 24, 36$) in all fittings. Details of the fitting for each critical exponent will be discussed in the subsections below.

| $\Omega_0$ | $\alpha$     | $\beta$   | $\gamma$    | $\nu$    |
|-----------|--------------|-----------|-------------|----------|
| $\Omega_0 > 0$ | 0.067 ± 0.018 | 0.126 ± 0.040 | 1.723 ± 0.162 | 0.99 ± 0.09 |
| $\Omega_0 < 0$ | 0.078 ± 0.025 | 0.117 ± 0.047 | 1.768 ± 0.425 | 1.05 ± 0.25 |
| 2D Ising   | 0(log)       | 1/8       | 7/4         | 1        |
A. Susceptibility $\chi$ and critical exponent $\gamma$

![Graph showing normalized staggered susceptibility and related quantities as a function of temperature](image)

**FIG. S6.** (a) The normalized staggered susceptibility for positive dilations $k_BT\chi = N_I \left( \langle m_{st}^2 \rangle - \langle m_{st} \rangle^2 \right)$ and the closely related quantity $k_BT\chi' = N_I \left( \langle m_{st}^2 \rangle - \langle |m_{st}| \rangle^2 \right)$ as a function of temperature $T$ for a membrane of size $L_N = 120$. The total number of up/down puckers $N_I = L_I \times L_I = 60 \times 60$. We can see $k_BT\chi$ converges to the system size $N_I = L_I \times L_I$ at low $T$. (b) For $T > T_c$, we see that $k_BT\chi'/(1 - 2/\pi)$ closely tracks $k_BT\chi$ [15].

The staggered susceptibility is given by

$$\chi = \frac{N_I}{k_BT} \left( \langle m_{st}^2 \rangle - \langle m_{st} \rangle^2 \right),$$

(S27)

where $N_I$ is number of dilations. True spontaneous symmetry breaking can occur in the thermodynamic limit only. For a finite system, there is a probability for the spontaneous staggered magnetization $\langle m_{st} \rangle$ to flip after a long finite time (unless we apply a small symmetry-breaking staggered field) and thus $\langle m_{st} \rangle$ is zero for all $T$.

Thus, for any finite system under zero external field $k_BT\chi = N_I\langle m_{st}^2 \rangle$. Consequently, $k_BT\chi$ is a monotonically increasing function that converges to $N_I$ as $T \to 0$ [15].
suggested by Binder [15], we can use $|m_{st}|$ to calculate a closely related quantity

$$\chi' = \frac{N_I}{k_B T} \left( \langle m_{st}^2 \rangle - \langle |m_{st}| \rangle^2 \right),$$ (S28)

which is better behaved. Below $T_c$, $\chi'$ converges to the true susceptibility in the thermodynamic limit $L_N \to \infty$. The quantity $\chi'$ is different from the true susceptibility for $T > T_c$ by a simple multiplicative factor $1 - 2/\pi$ [15]. We compare $k_B T \chi$ and $k_B T \chi'$ obtained

![Graph](image)

**FIG. S7.** The function $\chi'$ given by Eq. S28 for membranes with $\Omega_0 > 0$ as a function of temperature $T$ for different linear system sizes $L_N$. $\chi'$ increases with increasing system size and the location of pseudocritical temperature $T_c(L)$, where $\chi'$ is at its maximum, decreases with increasing system size. Note that for our (0, 2) dilation configuration on a square lattice, the linear system size of the spins is $L_I = L_N/2$. (b) Log-log plot of susceptibility peaks $\chi'_{\text{max}}$ as a function of linear system size $L_N/2$. The data points are fitted with a power law function and we find $\gamma/\nu = 1.741 \pm 0.062$.

from MD simulations of a membrane with $N_I = 60 \times 60$ positive dilations in Fig. S6. We indeed find that $k_B T \chi$ converges to the system size at low temperatures ($T < T_c$) and $k_B T \chi' \simeq k_B T \chi (1 - 2/\pi)$ at high temperatures. Since $\chi'$ is only different by a simple multiplicative factor, many Monte Carlo studies of spin systems use $\chi'$ to extract the critical exponent $\gamma$ [15, 16]. We use $\chi'$ to extract susceptibility exponents for membranes with
FIG. S8. The function $\chi'$ given by Eq. S28 for membranes with $\Omega_0 < 0$ as a function of temperature $T$ for different linear system sizes $L_N$. (b) Log-log plot of susceptibility peaks $\chi_{\text{max}}'$ as a function of linear system size $L_N/2$. The data points are fitted with a power law function and we find $\gamma/\nu = 1.684 \pm 0.061$.

$\Omega_0 > 0$ in Fig. S7. We see that the peak increases with increasing system size whereas the location of the peak decreases with increasing system size. The maximum value of $\chi'$ as a function of linear system size $L_I \equiv L_N/2$ in a log-log plot is shown in Fig. S7(b). By fitting the data to a power law function we obtain $\gamma/\nu = 1.741 \pm 0.062$. Similar trends are found for $\chi'$ in membranes with negative dilations, as shown in Fig. S8. For negative dilations we find $\gamma/\nu = 1.684 \pm 0.061$. Using $\nu$ obtained from FSS (see below), we obtain $\gamma_{\Omega_0>0} = 1.72 \pm 0.16$ and $\gamma_{\Omega_0<0} = 1.77 \pm 0.42$. 
FIG. S9. Specific heat of membranes with positive dilations $\Omega_0 > 0$ as a function of $T$ for nine different linear system sizes $L_N$. (b) Log-log plot of peaks of specific heat $C_{\text{max}}$ as a function of linear system size $L_N$. The data points are fitted with a power law function and we find $\alpha/\nu = 0.068 \pm 0.018$.

B. Specific heat $C$ and critical exponent $\alpha$

The average values of the energy and the square of the energy in thermodynamic equilibrium are given by

$$\langle E \rangle = \frac{1}{Z} \sum_j E_j e^{-E_j/k_B T} \quad (S29)$$

$$\langle E^2 \rangle = \frac{1}{Z} \sum_j E_j^2 e^{-E_j/k_B T}, \quad (S30)$$

where $Z$ is the partition function. Upon taking the derivative of $\langle E \rangle$ we find the specific heat for $N$ degrees of freedom:

$$NC \equiv \frac{\partial \langle E \rangle}{\partial T} = \frac{1}{k_B T^2} (\langle E^2 \rangle - \langle E \rangle^2), \quad (S31)$$
FIG. S10. Specific heat of membranes with $\Omega_0 < 0$ as a function of $T$ for different linear system sizes $L_N$. (b) Log-log plot of peaks of specific heat $C_{\text{max}}$ as a function of linear system size $L_N$. The data points are fitted with a power law function and we find $\alpha/\nu = 0.074 \pm 0.016$.

where $C$ is the specific heat per node. We can calculate $\langle E \rangle$ and $\langle E^2 \rangle$ from MD simulations by averaging over equilibrated configurations.

Note in the simulations we measure the total energy and so $N$ is the total number of nodes and not the total number of dilations $N_1$. In constrast, the staggered susceptibility was extracted just from the up/down fluctuations of the buckled sites. From finite-size scaling we have $C \propto |T - T_c|^{-\alpha} \propto L^{\alpha/\nu}$. In Fig. S9, we fit the peaks of $C$ with a power law function and find $\alpha/\nu = 0.068 \pm 0.018$ for positive dilations. For negative dilations (see Fig. S10), we find $\alpha/\nu = 0.074 \pm 0.016$. Note that these values are smaller than the 3D Ising class ($n = 1$ spin component) $\alpha/\nu \simeq 0.175$ and larger than the 2D Ising class $\alpha/\nu = 0(\log)$. Although we can also fit our data equally well with a logarithmic function, we note that these estimates are $\sim 3.5$ and $\sim 4.5$ standard deviations away from $\alpha = 0$, consistent with the possibility of a different universality class for this highly compressible Ising model.
C. Critical temperature $T_c$ and correlation length exponent $\nu$

![Image: A graph showing the fourth order Binder cumulant as a function of $T$ for different linear system sizes $L_N$. Data for $L_N = 160$ are not shown for clarity. $U_L$ approaches 2/3 at low $T$ ($T < T_c$) and goes to 0 at high $T$ ($T > T_c$).](image_url)

![Image: A log-log plot of linear system size $L_N/2$ as a function of $|T_c(L) - T_{c*}|$. $T_c(L)$ are obtained from the peaks of $\chi'$ and $T_{c*}$ is the estimated $T_c(\infty)$ obtained from the crossing of $U_L$ of the three largest systems. The data are fitted with a power law function and we find $\nu = 0.99 \pm 0.10$.](image_url)

**FIG. S11.** (a) Fourth order Binder cumulant for $\Omega_0 > 0$ membranes as a function of $T$ for different linear system sizes $L_N$. Data for $L_N = 160$ are not shown for clarity. $U_L$ approaches 2/3 at low $T$ ($T < T_c$) and goes to 0 at high $T$ ($T > T_c$). (b) Log-log plot of linear system size $L_N/2$ as a function of $|T_c(L) - T_{c*}|$. $T_c(L)$ are obtained from the peaks of $\chi'$ and $T_{c*}$ is the estimated $T_c(\infty)$ obtained from the crossing of $U_L$ of the three largest systems. The data are fitted with a power law function and we find $\nu = 0.99 \pm 0.10$.

The simplest way to estimate the correlation length exponent $\nu$ is to assume that, as in virtually all known critical phenomena, hyperscaling ($d\nu = 2 - \alpha$) holds, which in $d = 2$ dimensions gives $\nu = \frac{1}{1+\alpha/(2\nu)}$. Thus, from our specific heat data, we find $\nu = 0.967 \pm 0.008$ for positive dilations and $\nu = 0.964 \pm 0.007$ for negative dilations, results which are approximately 4-5 standard deviations away from the incompressible Ising value $\nu = 1$.

As a consistency check, we can also extract $\nu$ using finite size scaling by fitting $L = c_0 |T_c(L) - T_c(\infty)|^{-\nu}$, where $T_c(L)$ are obtained from the peak location of susceptibility, $T_c(\infty)$ is the critical temperature in the thermodynamic limit, and $c_0$ is a constant. Extracting $\nu$ by this method is rather difficult because we have three adjustable parameters and we do not know the true $T_c$ in the thermodynamic limit. We use the fourth order Binder...
FIG. S12. (a) Fourth order Binder cumulant for $\Omega_0 < 0$ membranes as a function of $T$ for different linear system sizes $L_N$. Data for $L_N = 160$ are not shown for clarity. (b) Log-log plot of linear system size $L_N/2$ as a function of $|T_c(L) - T_c^*|$. $T_c(L)$ are obtained from the peaks of $\chi'$ and $T_c^*$ is the estimated $T_c(\infty)$ obtained from the crossing of $U_L$ of the three largest systems. The data points are fitted with a power law function and we find $\nu = 1.05 \pm 0.25$.

cumulant [15, 16]

$$U_L = 1 - \frac{\langle m_{st}^4 \rangle}{3 \langle m_{st}^2 \rangle^2},$$

(S32)

to locate the phase transition and to estimate $T_c(\infty)$. $U_L$ approaches 2/3 in the low temperature broken symmetry phase and approaches 0 in the symmetric phase. $T_c$ is usually found by finding a $T$ where the two curves $U_L$ cross. Unfortunately, our data, for this highly compressible Ising model, are quite noisy, and we do not observe systematic dependence between intersection points and system size. We thus use the crossing of the three largest systems to estimate $T_c^* = T_c^{estimated}(\infty) \simeq 0.194$ for $\Omega_0 > 0$. With this value of $T_c^*$, we fit our data with a power law and find $\nu = 0.99 \pm 0.10$, shown in Fig. S11.

For membranes with $\Omega_0 < 0$ we find $T_c^* = T_c^{estimated}(\infty) \simeq 0.188$ and $\nu = 1.05 \pm 0.25$. Data are shown in Fig. S12. Note that while consistent with a value of $\nu$ near one, the error bars are much bigger than those obtained by using the formula $\nu = \frac{1}{1 + \alpha/(2\nu)}$. 
D. Magnetization $m_{st}$ and critical exponent $\beta$

FIG. S13. (a) Absolute staggered magnetization $\langle |m_{st}| \rangle$ for $\Omega_0 > 0$ as a function of temperature $T$ for different system sizes. Data for $L_N = 160$ are not shown for clarity. Log-log plot of $|m_{st}|$ at $T_c(L)$ as a function of linear system size $L_N/2$. The data with $L_N/2 > 20$ is fitted with a power law function and we find $\beta/\nu = 0.127 \pm 0.038$.

The quantity $|m_{st}|$ as a function of $T$ for $\Omega_0 > 0$ is shown in Fig. S13(a). One way to extract the staggered magnetization exponent $\beta$ is to fit $|m_{st}|$ at $T = T_c(L)$ with $L^{-\beta/\nu}$. In our system, it is hard to get reliable values of $\sqrt{m_{st}^2}$ at $T = T_c(L)$ with the error bars and the temperature steps we have in our simulations. Hence we obtain $\beta/\nu = 0.127 \pm 0.038$ with a large uncertainty. Data for negative dilations are shown in Fig. S14 and we find $\beta/\nu = 0.111 \pm 0.036$.

VII. Comparison of fluctuations in empirical graphene model and membrane model

The scaling behavior of critical systems, such as freestanding thermalized membranes, is known to be insensitive to microscopic details [17, 18]. To demonstrate this explicitly,
FIG. S14. (a) Absolute staggered magnetization $\langle |m_{st}| \rangle$ for $\Omega_0 < 0$ as a function of $T$ for different system sizes. Data for $L_N = 160$ are not shown for clarity. Log-log plot of $|m_{st}|$ at $T_c(L)$ as a function of linear system size $L_N/2$. The data points are fitted with a power law function and we find $\beta/\nu = 0.111 \pm 0.036$.

we performed computationally costly molecular dynamics simulations of graphene with AIREBO potentials [19] on Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [20] and measured the height-height correlations $|h(q)|^2$. Details on molecular dynamics procedures to simulate graphene with AIREBO potentials using LAMMPS can found in [21, 22]. The height fluctuations in Fourier space $\langle |h(q)|^2 \rangle$ at zero strain are given by

$$\langle |h(q)|^2 \rangle = \frac{k_B T}{A \kappa(q) q^4},$$

where $A$ is the area of the membrane, $\kappa(q)$ is the renormalized bending rigidity, and $q$ is magnitude of the wave vector. The effective $\kappa$ and $Y$ as a function of wavevector are given
by [11, 23]

\[
\kappa(q) \propto \begin{cases} 
\kappa_0 & \text{if } q \gg q_{\text{th}} \\
\kappa_0 \left( \frac{q}{q_{\text{th}}} \right)^{-\eta} & \text{if } q \ll q_{\text{th}}
\end{cases}
\]  
(S34)

\[
Y(q) \propto \begin{cases} 
Y_0 & \text{if } q \gg q_{\text{th}} \\
Y_0 \left( \frac{q}{q_{\text{th}}} \right)^{\eta_u} & \text{if } q \ll q_{\text{th}}
\end{cases}
\]  
(S35)

where \( \eta \sim 0.8 - 0.85 \) and \( \eta_u \sim 0.3 - 0.4 \). The thermal length is given by

\[
l_{\text{th}} = \sqrt{\frac{\pi^3 16 \kappa_0^2}{3 k_B T Y_0}} \equiv \frac{\pi}{q_{\text{th}}},
\]  
(S36)

where \( T \) is the temperature, \( k_B \) is the Boltzmann constant, \( \kappa_0 \) is the bending rigidity at \( T = 0 \), and \( Y_0 \) is the 2D Young’s modulus at \( T = 0 \). Thus we expect \( \langle |h(q)|^2 \rangle \propto q^{-4+\eta} \) for the temperature range we are interested in. Stiffening occurs when the observed length scale becomes comparable to the thermal length \( l_{\text{th}} \).[23].

For graphene with an AIREBO potential, \( \kappa_0 = 1 \) eV and \( Y_0 = 21 \) eV/Å\(^2\)—thus, \( l_{\text{th}} \sim 20 \) Å at room temperature (\( T = 300 \) K). We indeed see a collapse of the rescaled \( |h(q)|^2 \) in Fig. S15 when the data are plotted using Eq. S33. (see also Ref. [11]).

Next, we plot \( \langle |h(q)|^2 \rangle \) for a pristine membrane with the square lattice coarse-grained model used in the current work, shown in Fig. S16. We see that both models show \( \langle |h(q)|^2 \rangle \) with \( q^{-(4-\eta)} \) behavior with \( q \simeq 0.8 \).

[1] The corresponding expression in ref. [6], Eq. 24, is missing a factor of 1/2, and should instead read \( a^{0\alpha}_\alpha = -A^0_{\alpha\beta} + \frac{\Omega_{0\alpha}(0)}{2}\delta_{\alpha\beta} \). This change only produces corrections of order \( \Omega_0^2 \), which are dropped in this paper.

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FIG. S15. The collapse of normalized $\langle |h(q)|^2 \rangle$ as a function of $q/q_{th}$ for graphene modeled with an AIREBO potential simulated at $T = \{10, 20, 100, 300\}$ K. The membrane has a length of $L = 200$ Å and a width of $W = 50$ Å. Data are obtained by taking 100 snapshots during a total run of 10 ns ($10^7$ time steps). The time interval between snapshots is 100 ps. The dotted dashed line shows the $q^{-(4-\eta)}$ behavior.

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FIG. S16. Normalized $\langle |h(q)|^2 \rangle$ for a square lattice membrane of size $120a_0 \times 120a_0$ as a function of $q/q_{th}$ at different temperatures. The dashed line shows the $q^{-(4-\eta)}$ behavior.

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