Buckling and metastability in membranes with dilation arrays

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We study periodic arrays of impurities that produce local dilations, embedded in 2D crystalline membranes. These arrays provide a simple elastic model of shape memory. As the size of each dilational impurity increases (or the relative cost of bending to stretching decreases), it becomes energetically favorable for each of the M impurities to buckle up or down into the third dimension, thus allowing for of order $2^M$ metastable surface configurations. With both discrete simulations and the nonlinear continuum theory of elastic plates, we explore the buckling of both isolated dilations and dilation arrays at zero temperature, guided by analogies with Ising antiferromagnets. We conjecture ground states for systems with triangular and square impurity superlattices, and comment briefly on the possible behaviors at finite temperatures.

I. INTRODUCTION

In this work, we study a periodic array of dilational impurities, placed within a deformable network that approximates a thin elastic sheet. Past a buckling threshold, each impurity becomes a site of bistability, and can relieve in-plane stresses by escaping up or down into the third dimension. A complete understanding of such a system would address several important questions in soft matter.

First, this system provides a simple microscopic model of out-of-plane plastic deformation. Such surfaces can be deformed into metastable states indexed by the up/down puckers of M impurities, somewhat analogous to how two dimensional crystals in flat space are remodeled by point-like dislocations pinned at discrete positions determined by a periodic Peierls potential. Beautiful models exist for visualizing in-plane plastic deformations— notably, the classic movies and paper by Bragg and Nye using bubbles in a soap solution that are drawn together by capillary forces to form a triangular lattice \cite{bragg1926}. However, much less is known about the microscopic features that mediate out-of-plane shape changes, and the development of simple models for these features may suggest productive directions for understanding plastic deformation into the third dimension.

Second, a special case of the model studied here was introduced as an elastic model for a shape memory material in the recent work of Oppenheimer and Witten \cite{oppenheimer2019}. Inspired by memory effects in crumpled paper, these authors focused on surfaces without preprogrammed target structures that deform in response to sufficiently strong external forces and remain in a deformed state when forces are removed. These shapeable surfaces are of both theoretical and practical interest, with potential applications to soft robotics and deployable structures \cite{oppenheimer2019, nelson2019}. Oppenheimer and Witten \cite{oppenheimer2019} suggested the model we are interested in, a lattice of adjacent bistable nodes, to illustrate that simple, ordered systems can display shapeability. Ultimately, they found that more complex lattices with additional sources of frustration resulted in superior shape memory capabilities, and focused their analysis on these more intricate systems. We return here to their simplest model, and its generalizations, in order to work with a system that is easier to treat theoretically. We extend previous work both by characterizing more precisely the buckling transition of individual impurities \cite{oppenheimer2019} and also by considering larger arrays with a variety of ordered initial conditions and varying impurity spacing. A number of interesting models similar to the one we investigate here have recently been introduced as methods for shaping thin sheets in three dimensions \cite{oppenheimer2019, nelson2019, radzihovsky2019, radzihovsky2019b, radzihovsky2019c}.

Finally, the model studied here may be of interest to those examining atomically thin materials. Experimentally, large impurity atoms such as silicon and germanium in two dimensional materials such as graphene have been observed to buckle out of plane in a manner qualitatively similar to our model \cite{bostwick2018, bostwick2019}. Theoretically, interest in impurity disorder embedded in tethered surfaces (two-dimensional generalizations of linear polymer chains) dates back to at least the early 1990’s \cite{bostwick2018, bostwick2019}. In particular, Nelson and Radzihovsky \cite{nelson2019} introduced a model similar to the one studied here, with dilational impurities embedded at random positions in an elastic sheet. However, as shown below, ordered arrays of impurities highlight new effects that do not appear for the disordered case.

We begin our study by investigating the buckling behavior of a single dilational impurity with simulations. We then note that the use of periodic boundaries (with stress relaxation) in a single impurity simulation in fact creates an array of image impurities, identically buckled in a same-side “ferromagnetic” configuration, and develop a general analytic framework in Fourier space to predict the buckling transition point. We use this framework together with an analogy with Ising models to explore candidate ground states in more general buckled configurations. Finally, we introduce a discrete model with a square host lattice, which lacks the geometric frustration of an underlying triangular mesh, and revisit each of the above topics in this new system. We conclude by

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discussing prospects for future work, including speculations about the behavior of periodic arrays of dilations at finite temperatures.

II. SINGLE IMPURITY

A back-of-the-envelope calculation suggests a threshold at which buckling of an isolated dilational impurity becomes energetically favorable. Consider the in-plane displacement field for a dilational impurity in an isotropic solid initially confined to a two-dimensional plane (a discrete version of a dilation is shown in the inset of Fig. 1).

$$u(r) = \frac{\Omega_0 r}{2\pi r^2},$$

(1)

where $\Omega_0$ measures the extra area added to the surface, discussed in more detail in the next section. We integrate the in-plane stretching energy from $r = \delta$, a microscopic cutoff of order a lattice constant or surface thickness, to $r = \infty$ to get \[13\]:

$$E_s = \frac{1}{2} \int d^2r (2\mu u_{ij}^2 + \lambda u_{kk}^2) = \frac{\mu\Omega_0^2}{2(1+\nu)\pi \delta^2} = \frac{Y\Omega_0^2}{4(1+\nu)\pi \delta^2},$$

(2)

where $\mu$ and $\lambda$ are Lamé coefficients, and $Y$ and $\nu$ are the 2D Young’s modulus and Poisson’s ratio respectively.

On the other hand, if we calculate the bending energy of a buckled impurity, assuming for simplicity a Gaussian height profile, $f(r) = H_0 e^{-r^2/2\sigma^2}$ where $\sigma$ is a buckling length scale to be determined, we have

$$E_b = \frac{\kappa}{2} \int d^2r (\nabla^2 f)^2 = \frac{\pi \kappa H_0^2}{\sigma^2}. $$

(3)

If we consider a buckled state that relaxes stretching completely, we can relate $\Omega_0$ to $H_0$ through the extra surface area generated by the dilation in the Monge representation.

$$\Omega_0 = \int d^2r \sqrt{1 + \left(\frac{df}{dr}\right)^2} - 1$$

$$\approx \pi \int dr r \left(\frac{df}{dr}\right)^2 = \frac{\pi H_0^2}{2},$$

(4)

which leads to $E_b \approx 2\Omega_0 \kappa/\sigma^2$. Note that in contrast to Eq. 3 this energy is linear in the extra area $\Omega_0$. If we now ask when the bending energy of a buckled dilation is lower than the stretching energy, we find that it occurs above a critical value of the dimensionless Föppl-von Kármán number $\gamma$, constructed with the impurity size $\Omega_0$,

$$\gamma_c \equiv \frac{Y\Omega_0}{\kappa} \sim \frac{\delta^2}{\sigma^2}. $$

(5)

This simple argument is meant to provide a qualitative rather than quantitative estimate of when it makes sense to trade stretching energy for bending energy, as our assumption of an inextensible sheet is only valid far above the transition point, and we made an ad hoc assumption about the height profile (a more realistic exponentially decaying height profile gives similar results, as shown in the Appendix). However, the finding that the transition is controlled by $\gamma$ will be confirmed by a more detailed analysis presented below. Note that, unlike classic defect problems, such as the buckling of dislocations and disclinations, $\gamma$ does not diverge with system size \[5, 10\]. We note in passing that dilations can also cause buckling in a one-dimensional semiflexible polymer embedded in two dimensions if the endpoints are held fixed. In this case, the transition is controlled by a dimensionless parameter that depends on system size, and the buckled state is a smooth, global deformation.

A. Discrete model

Following Refs. \[16\] and \[17\], we build our model from a two-dimensional triangular lattice. An alternative square lattice model with diagonal bonds will be discussed in Sec. \[V\] Neighboring nodes at positions $r_i$ and $r_j$ are connected by harmonic springs and neighboring triangular faces are penalized when their normals, $n_\alpha$ and $n_\beta$, are not aligned. The energy of the system is then the sum of stretching and bending energies,

$$E = \frac{k}{2} \sum_{\langle i,j \rangle} (|r_i - r_j| - a_{ij})^2 + \tilde{k} \sum_{\langle \alpha,\beta \rangle} (1 - n_\alpha \cdot n_\beta),$$

(6)

where the first sum is over neighboring nodes, and the second is over neighboring faces. The rest length of the springs connecting nodes $i$ and $j$, $a_{ij}$, is varied to insert dilational impurities. With all $a_{ij}$ equal to $a_0$, this energy approximates the bending and stretching energies used in the previous section in the continuum limit with $k = \sqrt{3}Y$, $\tilde{k} = \frac{2}{\sqrt{3}}\kappa$ and a Poisson ratio $\sigma = 1/3$ \[16\].

Next, we insert a dilational impurity at node $i$ by setting $a_{ij} = a_0(1 + \epsilon)$ for all neighboring nodes $j$, thus making the rest length of the springs connecting $i$ to the rest of the lattice an amount $a_0\epsilon$ longer (see inset of Fig. 1). We describe a periodic array of impurities by a pair of integers $(n, m)$, such that one moves between isolated dilations by taking $n$ steps along one lattice direction, turning counterclockwise by 60°, and taking $m$ more steps, as shown for $(n, m) = (2, 2)$ in Fig. 1. The energy minimizations used to explore the metastable configurations in this paper were performed using FIRE \[18\], with key results verified with BFGS \[19\]. All distances are measured in units of $a_0$, the spacing of the background triangular lattice.

Boundary effects can be strong, especially close to the buckling transition. Impurities can more easily buckle near free edges, and often buckle on the same side of the host surface and then curl beneath it. We observe a dramatic example of this effect if we place the impurities
FIG. 1: A schematic defining relevant quantities for our discrete model of dilation arrays. Large nodes with colors mark the impurity sites, here arranged in a (2, 2) tiling. Impurities that have buckled up are shown in red, and those that buckled down are in blue. Two normals corresponding to neighboring faces that contribute to the bending energy are shown in green. Inset: A top down view of a single impurity. Bonds with rest lengths \( a_0 \) and \( a_0(1 + \epsilon) \) are highlighted in teal and purple respectively.

close to one another in a (1, 1) array. As shown in Fig. 2a, smooth cap-like structures are then preferred, even for membranes that would be too stiff to buckle with the relaxed periodic boundaries used in the remainder of this paper. This behavior bears some resemblance to recently observed hemispherical configurations of kinetoplasts, which are “chainmail” structures composed of rings of DNA [20, 21]. It would be interesting if the “maxicircles” of DNA woven into the chainmail structure composed of DNA “minicircles” could be related to the dilations studied in this paper.

To avoid these interesting distortions, we implement tension-free periodic boundaries on a hexagonal domain. Domain size is quoted in terms of the radius \( R \), which is defined as the distance from the hexagon center to its corner when all bond lengths are equal to \( a_0 \) (Fig. 2b). We alternately perform local minimizations and global minimizations. Local minimizations move all nodes according to the gradient of Eq. 6 with respect to vertex positions, with the nodes on the top right edge of the periodic hexagon identified with the nodes on the bottom left edge, etc. Global minimizations take place over all possible affine deformations of the background hexagonal simulation cell. Optimization to a local minimum energy configuration is complete when the magnitude of all components of both the 3\( N \) dimensional local deformation gradient, where \( N \) is the number of nodes that can move independently from one another, and the three dimensional global affine deformation gradient are below a threshold value.

In what follows, we will use continuum theory to make predictions about buckling in a discrete flexible membrane with an array of point-like dilations. In order to verify these predictions with simulations, we must be able to translate between the continuum limit parameters and corresponding microscopic simulation parameters. As mentioned, the mapping of the continuum parameters \( \gamma \) and \( \kappa \) to \( k \) and \( \tilde{\kappa} \) respectively has been described for this model [16]. However, we still must find an expression for continuum parameter \( \Omega_0 \), the extra area added to the surface in flat space due to the dilation (as in Eq. 1), in terms of the dilational parameter \( \epsilon \). We will show that we can relate these parameters by considering a coarse-grained description of the surface’s preferred metric, and verify this relation with simulations.

Consider our discrete lattice with a single impurity at the center of the simulation cell, as in Fig. 2a. We divide the mesh into hexagons composed of six triangular faces such that the impurity is at the center of one of the hexagons as in the inset of Fig. 3. Each of these hexagons in isolation has an unambiguously defined preferred surface area that minimizes the stretching associated with the nearest neighbor springs. The hexagons without the impurity at the center prefer an area \( 3a_0^2\sqrt{3}/2 \), composed of six equilateral triangles with side length \( a_0 \), and the hexagon with the impurity prefers a surface area of \( 3a_0^2\sqrt{(1 + \epsilon)^2 - 1/4} \), composed of six isosceles triangles with side length \( a_0(1 + \epsilon) \) and base length \( a_0 \) (thus forming a three-dimensional prismatic structure). We then define the extra preferred surface area of the impurity...
hexagon

\[ \Omega_0 = 3 \varepsilon^2 \left[ 1 + \varepsilon^2 - 1/4 - \varepsilon^2 \sqrt{3}/2 \right] \approx 3.46 \varepsilon^2 - 0.577 \varepsilon^2 + O(\varepsilon^3). \]  

Note that the extra area \( \Omega_0 \) scales linearly with the dilation parameter \( \varepsilon \) for small \( \varepsilon \). Upon placing the impurity at the origin, we can summarize this description in the continuum limit in terms of a preferred metric for the surface, \( \bar{g}_{\alpha\beta} \) [12]:

\[ \bar{g}_{\alpha\beta} = \delta_{\alpha\beta} \left( 1 + \Omega_0 \delta^2(r) \right). \]  

This same energy expression follows if we instead consider the impurity to be a source of stress \( \sigma_{ij}^{\text{imp}} \) in an infinite two-dimensional medium, of the form [22, 23]

\[ \sigma_{ij}^{\text{imp}} = (\mu + \lambda) \Omega_0 \delta_{ij} \delta^2(r). \]  

Therefore, a coarse-grained metric description allows us to write down an expression for \( \Omega_0 \) in terms of the microscopic model geometry, which we check by measuring macroscopic expansion in two dimensions.

Figure 3 confirms the approximately linear relationship between the extra area \( \Delta A \) and \( \varepsilon \) with the predicted slope. At large values of \( \varepsilon \), the data are slightly lower than the theory, due to the neglected negative quadratic term in Eq. 8. Note that this relation is independent of the Young’s modulus, as expected from Eq. 8.

This approach of approximating dilations with delta functions (as we do in Eq. 9) should be contrasted with the complementary back-of-the-envelope calculation presented at the beginning of this section (Eq. 2), where we instead controlled the short distance singular behavior of the dilation by imposing a microscopic cutoff distance \( \delta \).

We now survey parameter space, first for a single dilation embedded in a large patch of a triangular host lattice. As anticipated by the simple argument in Sec II, the buckling transition occurs at a critical value of the dilation Föppl-von Kármán number \( \gamma \), which we can define in terms of both macroscopic elastic parameters and microscopic simulation parameters as

\[ \gamma \equiv \frac{Y \Omega_0}{\kappa} \approx \frac{4(3.46 \varepsilon^2)k}{3 \kappa}. \]  

Important parameter regimes appear in Fig. 4, where we plot the rescaled height \( f_0 \) of the impurity at the center of our periodic cell as a function of \( \gamma \), for fixed system size \( R \). The rescaling of the height by a function of \( \varepsilon \) is chosen to match the expected height in the inextensible limit (\( \gamma \to \infty \)). In this prismatic limit, only the impurity is displaced in the \( z \) direction, and all other nodes are undisturbed (inset c of Fig. 4). The ease with which we can identify the inextensible limit is a nice feature of the discrete model.

For small \( \gamma \lesssim \gamma_c \approx 14 \), the flat state is energetically favorable. At a critical value of \( \gamma_c \approx 14 \), the buckled state becomes preferred, with the height of the impurity rising continuously as a function of \( \gamma \) from 0 until it reaches a maximum value at \( \gamma \approx 40 \). In this regime, the height profile of the buckled surface drops off smoothly from the center of the dilation, with vertical displacements slowly going to zero at the boundaries of our periodic hexagonal domain (inset a of Fig. 4). As \( \gamma \) continues to increase, the height of the impurity drops (inset b of Fig. 4), slowly approaching the prismatic \( \gamma \to \infty \) limit (inset c of Fig. 4), where the lowest energy configuration is an isolated pyramid centered on the impurity with zero stretching energy.

We now focus on values of \( \gamma \gtrsim \gamma_c \) just past the transition, such that \( \gamma \) is not exceptionally large, the buckled profile is smooth, and the physics is more likely to be
FIG. 4: Height of a single impurity in units of the lattice constant $a_0$, rescaled by the height in the prismatic limit of small $\kappa$ and large $\gamma$, as a function of the dilation Föppl-von Kármán number $\gamma$. Insets a-c show the height profile defined in inset a of Fig. 6 at specific values of $\gamma$, for $R = 15a_0$.

describable in terms of continuum elasticity. The height of the impurity in the transition region just above $\gamma_c$ is plotted in Fig. 5. As shown in the inset, the height grows like the square root of $(\gamma - \gamma_c)/\gamma_c$. This behavior is reminiscent of the mean field behavior of the zero field ferromagnetic or antiferromagnetic Ising model, with $\gamma$ playing the role of temperature and the impurity height playing the role of an order parameter. We will introduce a theory that reproduces this result in Sec. III.

The height profile of the buckled state close to the transition is also of interest, especially because it influences the interactions between dilations when multiple buckled impurities are present. When the vertical displacement caused by each impurity falls off quickly, as illustrated in inset c of Fig. 4 for $\gamma \to \infty$, well-separated impurities no longer influence each other. The length scale with which vertical displacements decay determines the interaction range of dilations on a lattice.

Remarkably, the dominant length scale close to the buckling transition with $\gamma \gtrsim \gamma_c$ appears to be the system size itself. At first sight, a dilation seems like a relatively minor perturbation to an elastic sheet (compared to a more extreme, topological lattice defect such as a disclination [16]), unlikely to have an effect on distant nodes. However, near the buckling transition, these systems prefer to distribute the dilation deformations globally and pay a penalty in stretching energy in order to avoid bending further. We show this effect in Fig. 6. The height of the impurity also increases with $R$ close to the buckling transition, as shown in inset b of Fig. 6; however, this dependence disappears as $\gamma \to \infty$. Thus, although the system size was not varied in Fig. 4, its variation would clearly have had an effect sufficiently close to the buckling transition. We note that the buckling threshold itself also has a correction due to finite system size that decays as $1/R$, which we will discuss in detail in Sec. III.

These observations are supported by recent work of Oshri et al. [24] who studied buckling in a closely related system in which a small disk at the center of a larger circular region with free boundaries experiences dilational in-plane growth. This model can be thought of as an alternative continuum version of our discrete model for

FIG. 5: Close to the transition, the height of the impurity in its buckled state scales as $\sqrt{\frac{\gamma}{\gamma_c} - 1}$. Inset: Data on a log-log scale as a function of $(\gamma - \gamma_c)/\gamma_c$, clearly showing a slope of 1/2 close to the transition. Data shown are for a periodic hexagon with radius $R = 15a_0$, $\epsilon = 0.1$, and $\gamma$ is changed by varying $\kappa$. 

a single impurity with free boundaries, derived with a different coarse-graining procedure. Oshri et al. [24] also found that there is a near-threshold regime just past the buckling transition where height profiles are “extensive” and the deformation spreads out over the whole system, and a far-from-threshold regime where the energy minimizing configurations are localized (analogous to our approach to the prismatic limit). For another paper related to this work, see Efrati et al. [25].

We note that there is also a weak dependence of the buckling transition point on $\epsilon$ that is not captured by our single dimensionless parameter $\gamma$. As we increase $\epsilon$, $\gamma_c$ is slightly depressed. This effect is too small to be observed in Fig. 4 but is more visible in Fig. 13, the corresponding figure for a square lattice system that we introduce in Sec. V. However, we restrict our simulations to small values of $\epsilon$ and $\gamma$ is changed slightly depressed. This effect is too small to be observed in Fig. 4 but is more visible in Fig. 13. The preceding discussion relied heavily on observations emerging from a particular discretized “tethered surface” model. We now supplement this investigation of a single impurity with a more quantitative theory for the location of the buckling threshold and the scaling behavior of the impurity height near the transition.

We first observe that our periodic boundary conditions are equivalent to considering an array of impurities. For the hexagonal system of radius $R$, the periodic image impurities are separated along the $(R/a_0, R/a_0) \equiv (n, n)$ direction (using the notation defined in Fig. 1 and Fig. 2), and are all constrained to buckle identically on the same side of the host membrane. For comparison with later results, we call this configuration of buckled periodic images a “ferromagnetic” array. Treating a single impurity system as a periodic array with a large defect spacing simplifies the analysis.

We model our system with the elastic energy functional corresponding to the Föppl-von Kármán equations with a contribution due to impurity defects from the generalization of Eq. 10,

$$E = \int d^2r \left[ \frac{\kappa}{2} \left( \nabla^2 f \right)^2 + \mu \sum_{\alpha=\beta}^2 \gamma_{\alpha\beta} c^2 - (\mu + \lambda) \Omega_0 u \gamma c(r) \right],$$

with

$$u_{\alpha\beta} = \frac{1}{2} \left( \frac{\partial u_{\alpha}}{\partial x_{\beta}} + \frac{\partial u_{\beta}}{\partial x_{\alpha}} + \frac{\partial f}{\partial x_{\alpha}} \frac{\partial f}{\partial x_{\beta}} \right),$$

$$c(r) = \sum_i \delta^2(r - r_i),$$

where $c(r)$ is the concentration of impurity defects—a sum of delta functions centered at regularly spaced impurity sites $\{r_i\}$.

We now minimize the energy functional, Eq. 14, with respect to $u_{\alpha}$, the in-plane displacements, for a fixed function of the out-of-plane displacement field, $f(r)$. We will then substitute the minimizing displacements, $u_{\alpha}^*$, back in to the original energy functional to arrive at an

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expression that depends only on \( f(r) \) and the fixed impurity concentration \( c(r) \).

This procedure is related to that used at finite temperatures by Nelson and Radzihovsky [13] (among others [26]) to integrate out in-plane phonons in a partition function. We will follow similar steps to isolate the effect of in-plane displacements and simplify the minimization.

After separating the strain tensor into \( q = 0 \) and \( q \neq 0 \) Fourier modes and decomposing \( A_{\alpha\beta}(q) \) into longitudinal and transverse parts in terms of the functions \( \phi_\alpha(q) \) and \( \Phi(q) \), we find

\[
u_{\alpha\beta}(r) = u_{\alpha\beta}(0) + A_{\alpha\beta}(0)
+ \sum_{\mathbf{q} \neq 0} \left( \frac{i}{2} q_\alpha u_\beta(q) + q_\beta u_\alpha(q) \right) + A_{\alpha\beta}(q) e^{i\mathbf{q} \cdot \mathbf{r}},
\]

(18)

\[
u_{\alpha\beta}(0) + A_{\alpha\beta}(0) + \sum_{\mathbf{q} \neq 0} \left( \frac{i}{2} q_\alpha (u_\beta(q) + \phi_\beta(q))
+ q_\beta (u_\alpha(q) + \phi_\alpha(q)) \right) + P^{T}_{\alpha\beta}(q) \Phi(q) e^{i\mathbf{q} \cdot \mathbf{r}},
\]

(19)

where \( P^{T}_{\alpha\beta}(q) = \delta_{\alpha\beta} - \frac{q_\alpha q_\beta}{q^2} \) is the transverse projection operator. Upon taking the Fourier transform of Eq. 14 expression that depends only on \( c \)

\[\Phi(q) = P_{\alpha\beta}(q) P_{\alpha\beta}^{T}(q) \Phi(q) = P_{\alpha\beta}(q) A_{\alpha\beta}(q), \]

(22)

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(22)

\[\phi_\beta(q) = \frac{-i}{q^2} \left[ 2q_\alpha A_{\alpha\beta}(q) - q_\beta P^{\mu}_{\mu\nu}(q) A_{\mu\nu}(q) \right], \]

(23)

where

\[A_{\alpha\beta}(q) = \frac{1}{2A} \int d^2r \left( \frac{\partial f}{\partial x_\alpha} \frac{\partial f}{\partial x_\beta} \right) e^{-i\mathbf{q} \cdot \mathbf{r}}, \]

(24)

the Fourier transform of the nonlinear part of the strain tensor, defined above in Eq. 16 and \( P^{\mu}_{\mu\nu}(q) = q_\nu q_\mu / q^2 \)
the longitudinal projection operator. These expressions fully determine our in-plane displacements.

We can also use in-plane displacements to eliminate the energetic contribution from the \( q = 0 \) mode by setting

\[u_{\alpha\beta}(0) = -A_{\alpha\beta}(0) + \Omega_0 c(0) \delta_{\alpha\beta}. \]

(25)

Since \( c(q) = \frac{1}{4\pi} \int d^2rc(r) e^{-i\mathbf{q} \cdot \mathbf{r}}, c(0) \) is simply the number density of impurity atoms, \( N_{\text{imp}} / A \). The \( q = 0 \) mode is free to assume this value in our model because of our tension-free periodic boundary conditions.

We can now write our energy minimized with respect to in-plane displacements in both Fourier and real space (neglecting terms of order \( \Omega_0^2) \) as

\[E = \frac{A}{2} \sum_{\mathbf{q} \neq \mathbf{0}} \left( \kappa q^4 |f(q)|^2 + Y |\Phi(q)|^2 - Y \Omega_0 \Phi(q)(-\mathbf{q}) \right), \]

(26)

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

(27)

where the prime on the integral reminds us that the \( q = 0 \) mode is excluded.

We focus first on the terms quadratic in \( f(q) \). Upon rewriting in terms of \( f(q) \) and specifying \( c(q) \) as in Eq. 17 the portion of the energy quadratic in \( f(q) \) reads

\[E_2 = \frac{A}{2} \sum_{\mathbf{q} \neq \mathbf{0}} \left( \kappa q^4 |f(q)| f(-\mathbf{q}) \right) + \frac{Y \Omega_0}{2v} \sum_{\mathbf{q} \neq \mathbf{0}} \sum_{\mathbf{q}^\prime \neq \mathbf{0}} P_{\alpha\beta}^{T}(q + q^\prime) \delta_{\alpha\beta} f(q) f(q') \delta_{\mathbf{G}, -\mathbf{q} - \mathbf{q}^\prime} \]

(28)

where \( v \) is the real space area of the unit cell and \( \mathbf{G} \)

is a reciprocal lattice vector, both corresponding to the impurity superlattice.

Since we require that the system is invariant under translations respecting the periodic boundary conditions, we expand \( f(q) \), the out-of-plane displacement, in the corresponding set of superlattice reciprocal lattice vectors \( \{ \mathbf{G} \} \),

\[f(r) = \sum_{\mathbf{G}} f(\mathbf{G}) e^{i\mathbf{G} \cdot \mathbf{r}} \]

(29)

For impurities in a periodic \((u, n) = (R/a_0, R/a_0)\) array, the primitive vectors of the reciprocal lattice lattice
\[ G_1 = -\frac{2\pi}{3R} \hat{x} + \frac{2\pi}{R\sqrt{3}} \hat{y} = -\frac{g_0}{2} \hat{x} + \frac{g_0\sqrt{3}}{2} \hat{y}, \]
\[ G_2 = \frac{2\pi}{3R} \hat{x} + \frac{2\pi}{R\sqrt{3}} \hat{y} = \frac{g_0}{2} \hat{x} + \frac{g_0\sqrt{3}}{2} \hat{y}, \]
with \[ g_0 = \frac{4\pi}{3R}, \] the lattice spacing in reciprocal space, and a real space area of the unit cell \[ v = R^3 \frac{3\sqrt{3}}{2}. \]

We will also work with a third reciprocal lattice vector of the same magnitude,
\[ G_3 = G_2 - G_1 = \frac{4\pi}{3R} \hat{x} = g_0 \hat{x}. \]

We expect the lowest energy buckled states near the transition can be approximately described by the smallest reciprocal lattice vectors, corresponding to the longest wavelength deformations possible under our assumption of a periodic array. As a first approximation, we assume \( f(r) \) is a linear combination of only the six smallest reciprocal lattice vectors, \( \{G_1, G_2, G_3, G_4, G_5, G_6\} \), with labels shown in the inset of Fig. 8.

We now write the energy given this limited subspace for both \( f(q) \) and \( c(q) \). Since our displacements are real, we require \( f^*(G_3) = f(-G_3) \). We impose this constraint and work in the six-dimensional subspace composed of the real and imaginary parts of \( f(G_1), f(G_2) \) and \( f(G_3) \), and introduce the shorthand \( \text{Re}[f(G_j)] = f_j^R \) and \( \text{Im}[f(G_j)] = f_j^I \). We then express the energy in matrix form such that
\[ \frac{E}{A} = H_{nm} f_n f_m, \]
with
\[ H = \begin{bmatrix}
\alpha & -\Delta & -\Delta & 0 & 0 & 0 \\
-\Delta & \alpha & -\Delta & 0 & 0 & 0 \\
-\Delta & -\Delta & \alpha & 0 & 0 & 0 \\
0 & 0 & 0 & \alpha & -\Delta & \Delta \\
0 & 0 & 0 & -\Delta & \alpha & -\Delta \\
0 & 0 & 0 & -\Delta & -\Delta & \alpha \\
\end{bmatrix}, \quad \text{and} \quad f = \begin{bmatrix}
f_1^R \\
f_2^R \\
f_3^R \\
f_1^I \\
f_2^I \\
f_3^I \\
\end{bmatrix}, \]
where \( \alpha = \kappa g_0^4 \) and \( \Delta = \frac{3Y\Omega_g^2}{8v} \). To determine the stability of the unbounded state, we solve for the six eigenvalues and eigenvectors of \( H_{nm} \). The eigenvalues are
\[ \left( \kappa g_0^4 + \frac{3Y\Omega_g^2}{8v}, \kappa g_0^4 - \frac{3Y\Omega_g^2}{8v}, \kappa g_0^4 + \frac{3Y\Omega_g^2}{8v}, \kappa g_0^4 - \frac{3Y\Omega_g^2}{8v}, \right), \]
where the second and third eigenvalue are doubly degenerate. Since all of the variables in Eq. 30 are positive, only the first two eigenvalues listed can attain negative values. The first eigenvalue will always give the lowest energy. We thus find that our system becomes unstable to the corresponding buckling eigenvector provided \( \gamma > \gamma_c \), where
\[ \gamma_c = \frac{\sqrt{3}v}{\kappa} = \frac{4v g_0^2}{3} = \frac{4 \pi^2}{3 \sqrt{3}} \approx 61. \]

The second eigenvalue becomes negative at \( \gamma = 2\gamma_c \). Note that the dilation Föppl-von Kármán number \( \gamma \) emerges naturally in this calculation, and all factors of \( R \) cancel in the product \( v g_0^2 \). This calculation provides a rough estimate of the buckling threshold, but it is greater than that measured in simulations by approximately a factor of four.

We now consider the eigenvector corresponding to the lowest energy eigenvalue (the first mode to go unstable). We define \( m_1 \) as its magnitude. This eigenvector gives the real space deformation
\[ f(x,y) = \frac{2m_1}{\sqrt{3}} \left[ \cos(G_1 \cdot r) + \cos(G_2 \cdot r) + \cos(G_3 \cdot r) \right], \]
pictured in Fig. 7. Although at this level of approximation the eigenvector is independent of \( \gamma \), this independence will not hold when more reciprocal lattice vectors are included in Eq. 29.

We assume that just past the transition, the deformation \( f(q) \) can be approximately described by this eigenvector. Contributions from the quartic stretching energy term can then easily be included. In general, the quartic term in the energy Eq. 14 is given by
\[ E_4 = \frac{Y A}{8} \sum_{q_1 \neq q_2} \sum_{q_3 \neq 0} \sum_{q_4 \neq 0} \sum_{q_5 \neq 0} \sum_{q_6 \neq 0} P_{\alpha\beta}(q_1 + q_2) q_{1\alpha} q_{2\beta} \]
\[ \times f(q_1) f(q_2) P_{\mu\nu}(q_3 + q_4) q_{3\mu} q_{4\nu} f(q_3) f(q_4) \delta_{q_5,0} \delta_{q_6,0} \]
(39)

Unlike the quadratic energy term, the quartic term is strictly positive, ensuring a finite value for the impurity height that minimizes the energy. The total energy as a function of \( m_1 \) is then
\[ E = \frac{\kappa g_0^4}{\gamma_c} (\gamma_c - \gamma) m_1^2 + \frac{5Y g_0^4}{24} m_1^4, \]
(40)
strikingly similar to the Landau free energy of the Ising model. We find the usual mean field “critical exponent” \( \beta = 1/2 \) when we minimize the energy with respect to \( m_1 \) when \( \gamma > \gamma_c \).

\[ m_1 = \pm \sqrt{\frac{12\kappa}{5Y \gamma_c} (\gamma - \gamma_c)} = \pm \sqrt{\frac{12\Omega_g^2}{5\gamma_c} \left( \frac{\gamma}{\gamma_c} - 1 \right)}. \]
(41)

This scaling behavior agrees with our data, shown in Fig. 5, and is consistent with prior work in real space by Carraro and Nelson [5].
In fact, this result does not require assuming that only the buckling mode with magnitude $m_1$ is present. At this level of approximation we can prove that the two other eigenvectors that can have negative eigenvalues do not appear, even for $\gamma > 2\gamma_c$. If we consider a height field composed of an arbitrary mixture of the first three eigenvectors, the energy to quartic order has a symmetry such that $m_2$ and $m_3$ always appear in the combination $m_2^2 + m_3^2 = \rho^2$. Given that $\frac{Y_{\text{bulk}}}{\nu} > 0$, it can be shown that $\rho$ will always be zero, and the only phase transition in the system is the continuous transition in $m_1$ that we have already observed.

Although including only six reciprocal lattice vectors in this continuum calculation allowed us to derive the scaling behavior we observe in our simulations, $\gamma_c$ is well above our numerical results, due we believe to our truncated basis in Fourier space. The energy minimizing structures we observe in simulations will surely have contributions from higher Fourier modes (see, for example, the short distance structure embodied in inset a of Fig. 8). We therefore repeat the calculation with higher Fourier modes to get a more accurate estimate.

Physically, as we raise the cutoff for the included Fourier modes, we allow for better resolution of the degrees of freedom embodied in the space between the impurity sites, while keeping the strength of the impurity and the distance between impurities the same. Systematically raising the maximum allowed $|\mathbf{G}| = G_{\text{max}}$ in the expansion Eq. 29 is similar to measuring $\gamma_c$ in our discrete model for a single impurity with periodic boundaries as we progressively increase $R = na_0$ for a fixed value of $a_0$. We have explored the agreement between theory and simulations by looking for trends in $\gamma_c$ as we increase both the number of included modes in the theory and the size of the system in the simulations. We do not expect quantitative agreement for small systems/few Fourier modes, since the discreteness of the host lattice has a large effect and the approximation of our impurities as delta-function dilations in our theoretical treatment breaks down in this limit. Agreement between the theory and numerics should improve as the maximum reciprocal lattice vector and system size are increased and we approach the continuum limit.

It is helpful to examine this argument in more detail in terms of our discrete model. The lattice constant of the reciprocal lattice associated with the triangular mesh of the host lattice is $g_n = \frac{4\pi}{\sqrt{3}a_0}$. However, the lattice constant of the impurity reciprocal lattice for an $(n, n)$ array is $g_0 = \frac{4\pi}{\sqrt{3}a_0} = \frac{4\pi}{\sqrt{3}na_0}$. Because the primitive vectors of these two lattices are at an angle of 30 degrees to one another, the first Brillouin zone of the host lattice is a hexagon of radius $G_{\text{max}} = \frac{4\pi}{3a_0}$ (see inset of Fig. 8). When the radius of the real space lattice is increased from $R = na_0$ to $R = (n+1)a_0$, the lattice spacing of the impurity reciprocal lattice shrinks. If we measure in units of the impurity reciprocal lattice spacing, the radius of the first Brillouin zone also increases from $G_{\text{max}} = n g_0$ to $G_{\text{max}} = (n+1)g_0$. It therefore seems plausible that the approach to the continuum limit will have the same scaling behavior for both the theory and simulations when plotted in the correct variables.

We test these ideas in Fig. 8 and Fig. 9. In Fig. 8, we calculate $\gamma_c$ by numerically finding the eigenvalues of the energy matrix as in Eq. 35. In Figure 9 we estimate $\gamma_c$ for the discrete model by varying $\gamma$ with a resolution of 0.1. For a given value of $\gamma$, we displace the impurity node at the origin in the positive $z$ direction and minimize the energy. We compare this energy to the energy of the system with the same dilation when it is minimized in flat space. We set $\gamma_c$ to be the highest value of $\gamma$ for which the energy of the system that relaxes in two dimensions is equal (at our level of numerical precision) to the energy of the system that relaxes in two dimensions. For all values of $\gamma$ greater than this $\gamma_c$, the energy of the system that is allowed to buckle is lower.

The values of $\gamma_c$ in Fig. 8 and Fig. 9 approach a limit as we increase $R$ and $G_{\text{max}}$. We can estimate the infinite system size value of $\gamma_c$ by extrapolating $1/R$ and $1/G_{\text{max}}$ to 0. This extrapolation allows us to predict that $\gamma_c(\infty) \approx 16.3$ from the Fourier space theory (Fig. 8) and $\gamma_c(\infty) \approx 13.3$ from the simulations (Fig. 9). We seem to have approximate agreement in the limit in which continuum theory should apply. Furthermore, as hoped, the scaling behavior is the same: $\gamma_c$ appears to be a linear function when plotted as a function of $1/R$ and $1/G_{\text{max}}$. A linear dependence on these quantities is plausible, since we expect corrections to the continuum limit to scale as the ratio of the hexagon perimeter to the hexagon area in both real and reciprocal space.

### IV. OTHER IMPURITY ARRAYS

Having characterized the buckling transition for a single impurity with periodic boundary conditions, we now...
FIG. 8: Variation of $\gamma_c$ with $G_{\text{max}} = n g_0$. When plotted as a function of $G_{\text{max}}^{-1}$, a linear extrapolation to the infinite system size limit gives $\gamma_c = 16.3$. Results from various truncations of the Fourier expansion Eq. [29] used in our theory are shown for $n$ between 1 and 7, with $G_{\text{max}}$ measured in units of $g_0$. The green line connecting these points is a guide to the eye. Inset: Fourier modes used in the calculation when $G_{\text{max}} = 3g_0$ lie within or on the boundary of the dotted hexagon. Labeled wavevectors are the innermost ring of six included in the first calculation of $\gamma_c$ (Eq. [37]).

FIG. 9: Variation of $\gamma_c$ from simulations with $R = n a_0$. When plotted as a function of $R^{-1}$, a linear extrapolation to the infinite system size limit gives $\gamma_c = 13.3$. Data are shown for $n$ between 5 and 40, $\epsilon = 0.05$, and $\gamma$ is changed by varying $\kappa$. Inset: Periodic unit cell of the defect superlattice for $R = 3a_0$.

explore more general impurity arrays. Past the buckling transition, each dilation can buckle either up or down out of the plane, possibly influenced by interactions with neighboring impurities. This bistability gives us a complex energy landscape with many metastable states. Although phenomena such as phase transitions at finite temperatures are quite interesting (see Sec. 7), we focus here on the ground state (or ground states) of the system at $T = 0$. Understanding the ground state will provide insight into how the system organizes, and is a starting point for future investigations of fluctuations among the many metastable states at nonzero temperatures, where entropy can play an important role.

However, determining which configuration of an interacting triangular superlattice of possibly buckled dilations has the lowest energy is a challenging problem. If we consider states characterized by their up/down patterns of $M$ buckled impurities at fixed $\gamma > 2\gamma_c$, there are of order $2^M$ candidate ground states to test. Since we do not have the computational resources to test all of these states for large $M$, it is difficult to prove unambiguously which state has the lowest energy. Instead, we will use physical reasoning, simulations, and calculations to conjecture a likely ground state for $\gamma > 2\gamma_c$. We suspect, but cannot prove, that the buckling pattern of up and down dilations with the lowest energy for $\gamma \geq \gamma_c$ will remain the pattern with the lowest energy for $\gamma \gg \gamma_c$.

We will continue to probe the system by varying $\gamma$ and the number of mesh spacings of the host lattice separating impurities. Although we will focus on $(n, n)$ arrays, two other array families are also interesting. $(0, n)$ arrays buckle in ways that depend more strongly on $n$ and other microscopic details of the underlying host lattice. Chiral arrays, which have $(n, m)$ with $n \neq 0$, $m \neq 0$, and $n \neq m$ interact more strongly with our planar boundary conditions. The $(n, n)$ arrays we study in detail have a smoother approach to the continuum limit, and allow us to directly apply lessons from the single impurity case.

Even with $(n, n)$ arrays, the parameter space is massive. The problem becomes more tractable if we assume that the ground state is determined by pairwise interactions between neighboring buckled impurities. This approximation allows us to be guided by previous work on the Ising model on a fixed triangular lattice in flat space.

We make a direct mapping from impurity displacements to Ising spins, in real space. If an impurity buckles up, we assign it to be spin up, and vice versa. Sufficiently close to the buckling transition, the distortions caused by two nearby buckled impurities will overlap. The interaction energy will differ depending on whether the impurities are buckled in the same direction or different directions (i.e. if the “spins” are aligned or anti-aligned). Depending on whether the energy is lower for the aligned or anti-aligned configuration, the local interaction is ferromagnetic or antiferromagnetic respectively.

Since the interaction can be long range, especially close to $\gamma_c$, we allow for next nearest and third nearest neighbor interactions between impurities. The possible ground states of the Ising model on a rigid triangular lattice with up to third nearest neighbor interactions are known for coupling constants of all possible signs and magnitudes...
FIG. 10: The buckled zigzag state for a \((4,4)\) array of dilations in a hexagonal periodic domain of size \(R = 48a_0\) at \(\gamma = 17.9\), our conjectured ground state for the triangular host lattice. 

(a) Zigzag state viewed in perspective, with the vertical displacements multiplied by a factor of 10 for clarity. 

(b) Top down view of the zigzag state with impurities that have buckled up shown in red and impurities that have buckled down shown in blue.

We assume that the ground state of our system with puckered dilations is one of these, and test all seven candidate states using an energy minimization algorithm for our model with elastic interactions and a bending energy. Since the algorithm finds the nearest local minimum, we can probe metastable states by initializing the simulation with small positive and negative vertical displacements on the impurity atoms in the desired Ising spin pattern.

We find, out of the seven Ising candidate states, the “zigzag state” shown in Fig. 10 is the first state to buckle as \(\gamma\) is increased, and has the lowest energy for all values of \(\gamma\) tested once buckling has occurred. We plot the difference between the energy of the zigzag state and two other states in Fig. 11. In Fig. 11 and the rest of this work, we only show results for three states of interest: the conjectured zigzag ground state, the striped state, which is close in energy to the ground state and pictured in inset a of Fig. 11, and the “ferromagnetic” state, which has the highest energy of all metastable states we measured and is pictured in inset b of Fig. 11.

The finding that the zigzag state has the lowest energy, closely followed by the striped state, suggests that all pairwise interactions are antiferromagnetic, with the strength of the interaction falling off with distance. In fact, the zigzag and striped state have exactly the same energy in an Ising model when only nearest neighbors are included—longer range interactions are required to break the degeneracy of the ground state. This conclusion is supported by measurements of the energy of isolated pairs of up-up and up-down buckled impurities as the separation is varied. The strength of the antiferromagnetic interactions goes to zero as the separation between impurities is taken to infinity. These interactions also vanish in the prismatic limit \(\gamma \to \infty\).

Long-range antiferromagnetic interactions also follow from a continuum treatment of dilations [5]. Past the buckling transition, the behavior of the system is determined primarily by the bending energy. As in the first section, we can easily calculate the bending energy of two nearby aligned/anti-aligned impurities by assuming the height profiles are Gaussian. The conclusions once again hold if a more realistic height profile with an exponential decay is used (see Appendix). The superimposed Gaussian height profile

\[
f(x, y) = H_0 \left( e^{-((x-d)^2+y^2)/2\sigma^2} \pm e^{-((x+d)^2+y^2)/2\sigma^2} \right),
\]

(42)
gives us a bending energy of the form

\[
E_b = \frac{\pi\kappa H_0^2}{\sigma^6} \left( 2\sigma^4 \pm e^{-\frac{d^2}{2\sigma^2}} (d^4 - 4d^2\sigma^2 + 2\sigma^4) \right).
\]

(43)

The parameter \(\sigma\) measures the size of the buckled impurity. Upon neglecting stretching energy in the buckled configurations, the energy difference between the anti-aligned (antiferromagnetic) and aligned (ferromagnetic)
FIG. 12: Root mean square height at impurity sites (as a measure of the magnitude of up and down buckling) versus γ. The zigzag state buckles first, followed closely by the striped state, and then the ferromagnetic state. Data are for (4, 4) arrays with \( R = 96a_0 \), \( \epsilon = 0.1 \), and \( \gamma \) is changed by varying \( \kappa \).

puckers is

\[
E_{++} - E_{+-} = -\frac{2\pi \kappa H_0^2}{\sigma^2} e^{-\frac{\sigma^2}{2}} \left( \left( \frac{d}{\sigma} \right)^2 - 2 \right)^2,
\]

and is negative when \( d/\sigma > \sqrt{2 + \sqrt{2}} \approx 1.85 \). We therefore expect antiferromagnetic pairwise interactions for impurities with separations somewhat larger than the buckling size.

This argument also suggests the possibility of short-range ferromagnetic interactions, when impurity centers are very close. We observe a similar effect in a (2, 0) array, consistent with the behavior observed by Carraro and Nelson [5], although the continuum description cannot be straightforwardly applied in this limit. We have avoided this regime in our simulations.

In addition to measuring the energy of various states containing many dilations, we can measure the buckling threshold. As seen in Fig. 12, the zigzag state buckles first. This observation is consistent with a zigzag ground state, since the buckling transition is defined as the point at which a new configuration attains an energy lower than the flat state energy. It also means we can use our calculation of \( \gamma_c \) to better understand ground state behavior. As we now show, the analysis presented to study a single impurity as a ferromagnetic array readily generalizes to other types of deformations.

To proceed, we determine the subspace of wavevectors compatible with the periodicity of the zigzag and striped states. We can predict nonzero \( f(\mathbf{q}) \) modes by, in the language of solid state physics [28], looking for extinctions in the structure factor of the impurity superlattice. To simplify our analysis, we initially assume that each impurity has a height of either 1 or \(-1\), and all non-impurity sites have a height of zero. Under this assumption, we can find the Fourier transform of our height field by treating up and down impurities as two different types of atoms with atomic form factors of 1 and \(-1\) respectively.

We first describe a unit cell for the impurity height field with a basis. For the striped state, the lattice vectors

\[
\mathbf{a}_1 = 3na_0 \hat{x}, \quad \mathbf{a}_2 = \sqrt{3}na_0 \hat{y},
\]

with the basis

\[
\mathbf{d}_1 = 0, \quad \mathbf{d}_2 = \frac{3na_0}{2} \hat{x} + \frac{\sqrt{3}na_0}{2} \hat{y},
\]

give the appropriate description, with \( \mathbf{d}_1 \) and \( \mathbf{d}_2 \) corresponding to impurities that buckle up and down respectively. The reciprocal lattice vectors describing this deformation are

\[
\mathbf{G}(b_1, b_2) = b_1 \frac{2\pi}{3na_0} \hat{x} + b_2 \frac{2\pi}{\sqrt{3}na_0} \hat{y},
\]

where \( b_1 \) and \( b_2 \) are integers, whose amplitude is modulated by the geometrical structure factor

\[
S_{\mathbf{G}} = f_{\mathbf{d}_1}(\mathbf{G}) + f_{\mathbf{d}_2}(\mathbf{G}) e^{i\mathbf{G} \cdot \mathbf{d}_2}.
\]

If we rescale the atomic form factors \( f_{\mathbf{d}_1}(\mathbf{G}) \) to unity, corresponding to identical atoms, we regain the reciprocal lattice described by Eqs. 30 and 31. However, the choice \( f_{\mathbf{d}_1}(\mathbf{G}) = 1 \) and \( f_{\mathbf{d}_2}(\mathbf{G}) = -1 \), leads to

\[
S_{\mathbf{G}} = 1 - e^{i\pi(b_1 + b_2)}.
\]

If \( b_1 + b_2 \) is odd, the reciprocal vector characterized by \( b_1 \) and \( b_2 \) will survive. If \( b_1 + b_2 \) is even, \( S_{\mathbf{G}} \) will be zero, and this wavevector will not show up in the Fourier transform of the striped state. This rule leads to the lattice shown in Fig. 13b.

For the zigzag state, the lattice vectors

\[
\mathbf{a}_1 = 3na_0 \hat{x}, \quad \mathbf{a}_2 = 2\sqrt{3}na_0 \hat{y},
\]

with the basis

\[
\mathbf{d}_1 = 0, \quad \mathbf{d}_2 = \frac{3na_0}{2} \hat{x} + \frac{\sqrt{3}na_0}{2} \hat{y}, \quad \mathbf{d}_3 = \sqrt{3}na_0 \hat{y}, \quad \mathbf{d}_4 = \frac{3na_0}{2} \hat{x} + \frac{3\sqrt{3}na_0}{2} \hat{y},
\]

give the appropriate description, with dilations at \( \mathbf{d}_1 \) and \( \mathbf{d}_4 \) buckling up and those at \( \mathbf{d}_2 \) and \( \mathbf{d}_3 \) buckling down. The corresponding reciprocal lattice vectors

\[
\mathbf{G}(b_1, b_2) = b_1 \frac{2\pi}{3na_0} \hat{x} + b_2 \frac{\pi}{\sqrt{3}na_0} \hat{y},
\]

lead to a structure factor of

\[
S_{\mathbf{G}} = 1 - e^{i\pi(b_1 + b_2)} - e^{i\pi b_1} + e^{i\pi(b_1 + b_2)},
\]
FIG. 13: a. Fourier modes for the height field computed with Eq. 54 for a striped configuration on a (4,4) impurity array with \( R = 96a_0, \gamma \approx 68 \). b. Allowed wavevectors for a striped (4,4) impurity array using Eq. 49. c. Fourier modes for the zigzag state under the same conditions as in a. d. Allowed wavevectors for the zigzag state as in b using Eq. 53.

which gives the lattice shown in Fig. 13b.

To verify that we have identified the appropriate components of \( f(q) \), we plot the intensity that would result from a scattering experiment,

\[
I(q) = \frac{1}{N} \left| \sum_{r_i} f(r_i) e^{-i q \cdot r_i} \right|^2 ,
\tag{54}
\]

using simulation data of the states, where \( r_i \) is the \((x,y)\) location of node \( i \) on the lattice (both impurity and non-impurity sites included, \( N \) sites in total), and \( f(r_i) \) is the height of that node. These results are shown in Fig. 13a and Fig. 13c for the striped and zigzag state respectively.

Calculating \( \gamma_c \) as before for each of these structures in Fig. 14 we find that the analysis correctly predicts that the striped and zigzag states buckle before the ferromagnetic state when a reasonable number of Fourier modes are included. We truncate Fourier space for each calculation such that the magnitude of the largest superlattice reciprocal lattice vector included in the calculation (see Eq. 28) is greater than or equal to the magnitude of the largest Fourier space component of \( f \). Upon extrapolating to \( G_{\text{max}}^{-1} = 0 \) (corresponding to an infinitely fine mesh for the host lattice of our dilations) and using a linear fit that neglects the two points with the fewest Fourier modes included, we find an estimate of \( \gamma_c(\infty) = 16.3 \) for the ferromagnetic state, \( \gamma_c(\infty) = 16.2 \) for the striped state, and \( \gamma_c(\infty) = 16.0 \) for the zigzag state.

FIG. 14: Variation of \( \gamma_c \) with \( G_{\text{max}} = n g_0 \) for the ferromagnetic state (same values as in Fig. 8), striped state, and zigzag state. Results from various truncations of the Fourier expansion Eq. 29 used in our theory are shown for \( n \) between 1 and 7, with \( G_{\text{max}} \) measured in units of \( g_0 \).

FIG. 15: Root mean square impurity height (as in Fig. 12) for the zigzag state in our simulations as a function of \( \gamma \) with increasing impurity separation \((n,n)\). Data are for separations \( n = 2, 4, 6, \) and 8 in a hexagonal domain of size \( R = 96a_0 \) with \( \epsilon = 0.1 \), and \( \gamma \) is changed by varying \( \kappa \).

We can also see this effect in our simulations, illustrated in Fig. 15 for the zigzag state. As we increase the separation between impurities embedded in \((n,n)\) lattices, effectively increasing the number of wavevectors that can be included, the buckling threshold decreases in a manner consistent with a smooth approach to a limiting \( \gamma_c(n) \) as \( n \to \infty \).
V. EXTENSION TO A SQUARE LATTICE

Explorations of the ground state for a triangular lattice of impurities are complicated by geometric frustration, a familiar difficulty in Ising-like systems [27]. Because pairwise interactions between neighboring dilations are antiferromagnetic, longer-range interactions are necessary to specify the ground state. A natural question to ask is: How do our results change for impurity buckling in a geometry where the ground state of, say, an antiferromagnetic configuration is not frustrated?

To study this question, we design a discrete model of a two dimensional isotropic solid whose underlying lattice has a local square symmetry. This model allows us to have a square lattice of dilational impurities, whose post-buckling ground state we expect to be an unfrustrated checkerboard configuration. We begin with a simple square lattice, and add diagonal bonds between a subset of next nearest neighbor pairs to remove floppy modes. Placing bonds between all next nearest neighbors is an appealing option, because it approximates a pair potential with a second minimum at \( \sqrt{2}a_0 \), thus favoring a square lattice. Unfortunately, with two diagonal bonds in each square unit cell, the definition of the normals used to calculate bending energy is somewhat complicated. We instead make a simpler choice, shown in the inset of Fig. 16, which results in uniquely defined normals, but two distinct types of sites on the lattice. Half of the sites have four bonds, all of which connect to nearest neighbors, and the other half have eight bonds, half of which connect to nearest neighbors. We place impurities only on sites that have eight bonds in order to better approximate isotropic dilations. The relative strengths of the diagonal and nearest neighbor springs will be adjusted to produce isotropic elastic behavior at long wavelengths, despite the local square symmetry.

To fully specify the geometry, we must provide rest lengths for the bonds, which, as before, are modeled as harmonic springs. In the absence of impurities, nearest neighbor bonds have rest length \( a_0 \), and diagonal next nearest neighbor bonds have rest length \( \sqrt{2}a_0 \). For a dilational impurity, we would like the \( \gamma \rightarrow \infty \) ground state to be an isolated pyramid with zero stretching energy, as we found for the triangular lattice. With this aim in mind, on each impurity site we extend the nearest neighbor bonds to have rest length \( a_0(1+\epsilon) \) and the next nearest neighbor bonds to have rest length \( a_0(\sqrt{2}+2\epsilon+\epsilon^2) \). We can then calculate the excess area of the planar state \( \Omega_0 \) as before to be

\[
\Omega_0 = \Delta A = 4(a_0^2(1+\epsilon) - a_0^2) = 4a_0^2\epsilon. \tag{55}
\]

Figure 16 shows the linear relationship between \( \Delta A \) and \( \epsilon \) with the predicted slope. Unlike for the triangular lattice, we do not see any deviation at high \( \epsilon \), presumably because there are no higher order corrections to \( \Omega_0 \) in this case. As for the triangular lattice, the magnitudes of the elastic constants drop out.

Having specified our lattice structure, we now follow standard methods to find the spring constants for the discrete model that approximate an isotropic solid in the continuum limit [15, 16]. While in principle the spring constants for nearest and next nearest neighbors could differ, we find that they are the same for this model, and equal to

\[
k = \frac{4}{3}Y, \tag{56}
\]

where \( Y \) is again the macroscopic 2D Young’s modulus. We also find a 2D Poisson ratio \( \sigma = 1/3 \), just as we had for the triangular lattice.

To find the correspondence between the discrete bending rigidity \( \kappa \) and the continuum limit parameter \( \kappa \), we roll the modified square lattice into a cylinder along a vertical axis such that faces only bend perpendicular to hinges formed by nearest neighbor bonds of length \( a_0 \) (Fig. 17a). This configuration allows us, both analytically and numerically, to show

\[
\hat{\kappa} = \kappa, \tag{57}
\]

where \( \kappa \) is the continuum value of the bending rigidity. In principle, we could have a different value of the bending rigidity associated with bending perpendicular to bond hinges of length \( \sqrt{2}a_0 \). We test for this possibility by rolling a section of the lattice into a cylinder such that diagonals are along the cylinder axis, with bending now occurring across both long and short bonds (Fig 17b). This
construction allows us to compare the measured bending energy of large cylinders to the expected bending energy from continuum theory, and confirm that using only one value of $\tilde{\kappa} = \kappa$ is a good approximation.

We can now define a dimensionless dilation Föppl-von Kármán number $\gamma$ as for the triangular lattice model, differing only in the form of $\Omega_0$:

$$\gamma \equiv \frac{Y\Omega_0}{\kappa} = \frac{4(4a_0^2 \epsilon)k}{3\tilde{\kappa}}. \quad (58)$$

As before, we find a buckling transition for an isolated dilation with increasing $\gamma$, as shown in Fig. 18 and impurity height scaling close to the transition that again goes as $\sqrt{(\gamma - \gamma_c)/\gamma_c}$ (inset of Fig. 18).

We can again systematically increase the size of the discrete system and the number of modes in the Fourier space theory to estimate the buckling threshold in the infinite system size limit. For the square lattice, the periodic image impurities appear along the $(0, n)$ direction. We find rough agreement between the measured values of $\gamma_c$ shown in Fig. 19 ($1/R$ extrapolation $\gamma_c(\infty) \approx 10.4$) and the predicted values shown by the purple line in Fig. 23 ($1/G_{\text{max}}$ extrapolation $\gamma_c(\infty) \approx 16.4$). We attribute these differences to approximating the dilations by delta functions in the continuum theory. As in the triangular lattice case, the simulation value is lower than the continuum elastic theory estimate. Note that we quote lattice size in terms of $R$, which we define to be half of the length of a side of the square domain defining our periodic boundary conditions (inset of Fig. 19), for the purpose of easier comparison with the hexagonal domain used for the triangular lattice study.

Finally, we study periodic arrays of interacting dilations inserted into our square host lattice. By analogy with an Ising model on a square lattice with nearest and next nearest neighbor interactions, we focus on only three candidate ground states [29]. The checkerboard state, a Néel configuration shown in Fig. 20, will be the ground state if (as we expect for our system) there is strong nearest neighbor antiferromagnetism. The ferromagnetic
FIG. 20: The checkerboard state for a \((0, 4)\) array in a square periodic domain of size \(R = 24a_0\) at \(\gamma = 26.7 > \gamma_c\), viewed in perspective with the vertical displacements multiplied by a factor of 5 for clarity. This configuration is our conjectured ground state for the square lattice. Impurities that have buckled up are shown in red and impurities that have buckled down are shown in blue. Compare to Fig. 10.

FIG. 21: Difference in energy between two metastable states, the striped state (inset a, stripes run vertically) and the ferromagnetic state (inset b), and our conjectured checkerboard ground state pictured in Fig. 20 for \(\gamma\) larger than \(\gamma_c\) of the checkerboard state. All data are for \((0, 4)\) arrays with \(R = 96a_0\), \(\epsilon = 0.1\), and \(\gamma\) is changed by varying \(\kappa\). Compare to Fig. 11 and note the difference in scale on the y axis.

FIG. 22: Root mean square height measured at impurity sites versus \(\gamma\). The checkerboard state buckles first, followed by the striped state, and then the ferromagnetic state. Data are for \((0, 4)\) arrays with \(R = 96a_0\), \(\epsilon = 0.1\), and \(\gamma\) is changed by varying \(\kappa\). Compare to Fig. 12.

VI. DISCUSSION

The dilational impurity arrays studied here provide a simple arena for exploring shape memory, instability, metastable states, and Ising-like phase transitions for
FIG. 23: Variation of $\gamma_c$ with $G_{\text{max}} = n g_0$ for the ferromagnetic state, striped state, and checkerboard state. Results are shown for $n$ between 1 and 7, with $G_{\text{max}}$ measured in units of $g_0$. Compare to Fig. 14.

two-dimensional surfaces embedded in three dimensions. Tools from continuum elastic theory allow us to predict the approximate location and nature of the buckling transitions as a function of the dilation Föppl-von Kármán number $\gamma = \frac{Y b^3}{S b}$. Furthermore, we have conjectured ground states for $(n, n)$ arrays embedded in triangular host lattices and $(0, n)$ arrays with square host lattices, and found that these conjectures are consistent with simulations, calculations, and an Ising model analogy.

The Ising analogy provided us with a set of states to test, and in our analysis we assumed that the true ground state was contained in this set. While this approach has an appealing physical motivation, it would also be of interest to design a numerical experiment allowing the system itself to select the ground state. We could, for example, start a triangular lattice with dilations at a high temperature, slowly cool it down, and hope to observe domains of the zigzag state forming. However, such simulations are technically challenging because of the large system size required, the many metastable states with energies close to the ground state, especially for the triangular lattice, and the energy barrier for flipping an impurity and nearby affected sites from up to down. Although beyond the scope of this work, these difficulties could certainly be overcome, and we feel that this avenue is worth pursuing.

An especially intriguing aspect of finite temperature simulations of a regular lattice of dilations embedded in one of the “tethered surfaces” studied here is the interplay between crumpling transitions and the Ising-like orderings of buckled dilations. Even in the absence of an array of buckled impurities, triangulated surfaces in a low temperature flat phase can undergo a transition to an entropy-dominated crumpled state above a critical temperature $T_c$. Recent results suggest that this crumpling temperature (in the absence of distant self-avoidance) can be lowered considerably by inserting a regular array of holes in a simple model of free-standing graphene. If these holes are replaced by the regular array of dilations studied here, the resulting membranes have additional internal degrees of freedom. There can now be a finite temperature buckling transition at a temperature $T_b$, as well as a transition to a low temperature phase where these bucklers then order into a zigzag or checkerboard state via an Ising model phase transition at temperature $T_I$. It would be intriguing to study such phase transitions in this system, which resembles a highly compressible Ising model: The host lattice itself may rise up and crumple for entropic reasons! Self-avoidance of the host polymer sheet might well play a role under some circumstances, as would the relative ordering of important temperatures such as $T_c$, $T_b$, and $T_I$.

Although we have focused here on dilations, which locally add extra area to the lattice, one could also study defect arrays that remove area, such as a lattice of vacancies. In two-dimensional flat space, removing a single particle from a triangular array typically produces a “crushed vacancy, whose elastic field has a dipolar character. Little is known about what happens to interacting arrays of crushed vacancy dipoles, especially when allowed to relax into the third dimension.

It would also be of interest to study more systematically other $(n, m)$ tessellations of dilations in the host lattice, particularly the chiral versions with $n \neq 0$, $m \neq 0$, and $n \neq m$, which play an important role in the capsids of viruses and in various phyllotaxis problems (see references therein).

Finally, while we studied systems with periodic boundaries to better approximate an infinite, approximately planar material, experimental realizations of this system and related systems will likely have free or clamped boundaries. Boundary effects should therefore be systematically studied, as they can profoundly change the behavior in small systems, as illustrated by Fig. 23.

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A. Results for dilation buckling with an exponential profile

We present here a back-of-the-envelope calculation that motivates the existence of a buckling threshold like that in Eq. 5 using however an exponential height profile that better mimics simulation results than a Gaussian (see insets a and b of Fig. 4). Upon assuming a
buckled state height profile of the form
\[ f(r) = H_0 e^{-r/\sigma}, \]  
(A1)
the bending energy is
\[ E_b = \frac{\kappa H_0^2}{2} \int d^2r \frac{e^{-2r/\sigma}(\sigma - r)^2}{\sigma^4 r^2}. \]  
(A2)
If we integrate this from \( r = \delta \), a microscopic cutoff, to \( r = \infty \), the result can be expressed in terms of the exponential integral function \( Ei(z) = -\int_{-\infty}^{\infty} e^{-t} t dt \),
\[ E_b = \frac{\kappa H_0^2}{2} e^{-2\delta/\sigma}(2\delta - 3\sigma) - 4\sigma Ei(-2\delta/\sigma). \]  
(A3)
Upon letting \( x = 2\delta/\sigma \), this simplifies to
\[ E_b = \frac{\kappa H_0^2}{8\sigma^2} (e^{-x}(x - 3) - 4Ei(-x)). \]  
(A4)
Since \( \delta \) is of order the lattice constant \( a_0 \) and \( \sigma \) is many lattice constants near the transition, we expand about \( x = 0 \). Close to zero, the expansion is dominated by the term:
\[ E_b \approx \frac{\kappa H_0^2}{2\sigma^2} (\log(\sigma/\delta) + \text{const.}) \]  
(A5)
As before, we consider a buckled state that relaxes stretching completely, and relate \( \Omega_0 \) to \( H_0 \) by considering the extra surface area generated by the dilation in the Monge representation (now integrating from \( r = 0 \) to \( r = \infty \)). This gives
\[ \Omega_0 \approx \frac{\pi H_0^2}{4}. \]  
(A6)

If we again ask when the bending energy of a buckled dilation is lower than the stretching energy (Eq. 2), we once again find that it occurs above a critical value of the dimensionless Föppl-von Kármán number \( \gamma \), constructed with the impurity size \( \Omega_0 \),
\[ \gamma_c \equiv \frac{Y \Omega_0}{\kappa} \sim \frac{\delta^2 \log(\sigma/\delta)}{\sigma^2}, \]  
(A7)
which agrees with Eq. 3 up to logarithmic corrections. We have assumed here that the microscopic bending and stretching cutoffs are both \( \delta \) and dropped the constant in Eq. A5.

We can also superimpose two exponential height profiles and calculate the difference in bending energy between the ferromagnetic and antiferromagnetic configurations, as we did for Gaussian profiles in Eq. 4. The superimposed exponential height profile (compare with Eq. 42) is now
\[ f(x, y) = H_0 \left( e^{-\sqrt{(x-d)^2+y^2}/\sigma} \pm e^{-\sqrt{(x+d)^2+y^2}/\sigma} \right). \]  
(A8)
We can now compute the difference in energy between the anti-aligned and aligned configurations \( (E_{++} - E_{--}) \), as in Eq. 44 by numerically integrating \( (\nabla^2 f)^2 \) over a large region that excludes an area of radius \( \delta \) around \( x = \pm d, y = 0 \). As in the case of superimposed Gaussians, we find short-range ferromagnetism and long-range antiferromagnetism.

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