

# Order by disorder in the antiferromagnetic Ising model on an elastic triangular lattice

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**Geometrically frustrated materials have a ground-state degeneracy that may be lifted by subtle effects, such as higher-order interactions causing small energetic preferences for ordered structures. Alternatively, ordering may result from entropic differences between configurations in an effect termed order by disorder. Motivated by recent experiments in a frustrated colloidal system in which ordering is suspected to result from entropy, we consider in this paper the antiferromagnetic Ising model on a deformable triangular lattice. We calculate the displacements exactly at the microscopic level and, contrary to previous studies, find a partially disordered ground state of randomly zigzagging stripes. Each such configuration is deformed differently and thus has a unique phonon spectrum with distinct entropy, lifting the degeneracy at finite temperature. Nonetheless, due to the free-energy barriers between the ground-state configurations, the system falls into a disordered glassy state.**

colloidal monolayer | geometric frustration | magnetoelasticity

**F**rustrated systems are characterized by interactions that may not be satisfied simultaneously (1). This leads to a degenerate and thus disordered ground state, and naively one would expect to have disorder down to zero temperature (2). However, the frustrated phase is very sensitive to small perturbations that can order it. These include anisotropic (3) or longer-range (4) interactions and lattice deformations (5–10), as well as entropic effects that may lift the ground-state degeneracy at finite temperature, in a process termed order by disorder (11–16). Frustration and its relief due to order by disorder are traditionally investigated in antiferromagnets and, in particular, in compounds that have a triangular-lattice structure (17–20). Recent experiments have demonstrated that artificial systems made of mesoscopic building blocks such as single-domain magnetic islands (21) or colloidal spheres (22) exhibit behavior that is similar to that of magnetic systems comprised of atomic-scale particles. Such mesoscopic systems enable direct visualization of the dynamics at the single-particle level and thus provide insight into the microscopic physical mechanisms responsible for the peculiar properties of frustrated matter. In this paper we address the theoretical underpinnings of these experimental systems and thus provide a vital step in understanding their unusual behavior and its connection to atomic-scale antiferromagnets.

A densely packed monolayer of hard spheres buckles out of its plane when it is confined between walls that are separated by slightly more than a single sphere diameter (23). Entropic forces depending only on geometry give rise to effective antiferromagnetic interactions favoring motion of neighboring spheres toward opposite walls (24), leading at high densities to stripes of alternating up and down spheres. The close-packed state is highly degenerate: The same maximal density is obtained by straight stripes or by any set of parallel stripes that zigzag within the hexagonally packed layer (22, 24–28). Recent experiments indicate a possible preference of the stripes to be straight rather than to zigzag randomly in the plane, suggesting that at densities below close packing there is an order-by-disorder effect giving an entropic advantage for straight stripes (22).

This quasi-two-dimensional problem is strongly related to the old, yet unsolved question of what is the stable high-density structure of hard spheres, fcc or hcp (29–31). In three dimensions, maximal density is obtained by stacking hexagonally packed layers with arbitrary sideways shifts between the close-packed positions. And, as for the buckled monolayer, slightly below close packing it is not clear which structure has the greatest entropy. Experiments on colloidal crystals that were grown slowly enough exhibit fcc order (32), whereas there is controversy on whether the theoretical estimates have reached the accuracy required to resolve the elusive entropic difference between fcc and hcp (33–38).

As for the fcc vs. hcp question, the entropy, or free volume, of buckled hard spheres is a collective function of the positions of all particles; thus the ability to obtain analytical results for it is very limited. Thus, instead of *approximating* the entropy of this hard-sphere system, we consider the antiferromagnetic Ising model on a deformable triangular lattice. This model has the same degenerate ground state of zigzagging stripes as the colloidal system, and for it we can *exactly* calculate the free-energy difference between the competing configurations. We find that straight stripes are always favored entropically. However, the free-energy barriers between various ground states are huge compared to this entropic gain, causing the system to fall into a disordered glassy state upon cooling.

## The Model and Its Ground States

In our Ising model, each site  $i$  at continuous position  $\vec{r}_i$  on a triangular network is occupied by a particle of discrete spin  $\sigma_i = \pm 1$ , and each nearest-neighbor bond comprises a harmonic spring of stiffness  $K$  and relaxed length  $a$ ; see Fig. 1A. The internal energy of the system depends only on the spin product  $\sigma_i\sigma_j$  of neighboring particles and on their relative positions,  $\delta r_{ij} = |\vec{r}_i - \vec{r}_j| - a$ . The model's Hamiltonian is given by the following sum over all nearest-neighbor pairs  $\langle ij \rangle$ :

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[ (1 - \epsilon \delta r_{ij}) J \sigma_i \sigma_j + \frac{K}{2} \delta r_{ij}^2 \right]. \quad [1]$$

The antiferromagnetic interaction strength is equal to  $J > 0$  for particles separated by the relaxed spring length  $a$  and decreases linearly with distance at a rate  $\epsilon > 0$ . This mimics the buckled colloidal monolayer in the following way: For hard spheres, free energy is determined by their free volume. However, it is convenient to reduce this to an effective two-particle repulsive interaction. Although the particles are confined to be quasi-two-dimensional, they do have a limited freedom to move

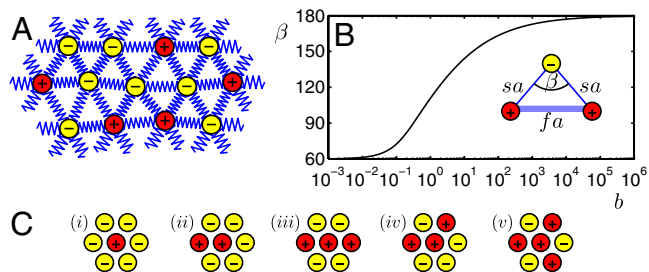
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**Fig. 1.** Model and its ground state. (A) Two-dimensional triangular network of antiferromagnetic Ising spins connected with harmonic springs. (B) Deformation angle  $\beta$  vs. the dimensionless ratio  $b \equiv \frac{Jc}{Ka}$  between the magnetoelastic interaction strength and the lattice rigidity, from Eq. 2. (Inset) Isosceles triangle with head angle  $\beta$ , satisfied bonds (thin lines) compressed by  $s$ , and frustrated bond (thick line) stretched by  $f$ . (C) The five possible configurations (up to rotations and spin inversions) of a particle and its neighbors, with a single frustrated bond in each triangle.

in the vertical direction, and thus nearest neighbors prefer to sit at opposite heights. We can now map the vertical position of each particle onto an Ising degree of freedom, due to a preference for either an “up” or “down” state. This leads to an effective two-dimensional antiferromagnetic Ising model, with the coupling between the elastic and the magnetic degrees of freedom as reflected in our Hamiltonian (Eq. 1); i.e., the effective antiferromagnetic interaction decays as the in-plane separation increases (24).

We model the in-plane entropic repulsion between neighboring spheres by a rotationally invariant central-force harmonic spring (Eq. 1). Approximate versions of this model have been studied previously: Ref. 6 uses a linearized long-wavelength elastic energy, invariant with respect to only infinitesimal rotations, that limits deformations to be small and to vary slowly in space, whereas in ref. 5 deformations are taken to be uniform.

In this paper we present results for the case in which the total area of the system is fixed to the area of a triangular lattice with lattice constant equal to  $a$ . However, we obtained qualitatively similar results for systems compressed or dilated with respect to this simple situation. From the equivalence between ensembles we thus expect to get the same results also when considering the fixed pressure case, which is probably more appropriate for the colloidal experiments.

As in the rigid triangular-lattice model (2, 3), the antiferromagnetic interactions along each three-particle loop in our deformable network cannot be satisfied simultaneously, and energy is minimized by having two satisfied ( $\sigma_i\sigma_j = -1$ ) bonds and a single frustrated ( $\sigma_i\sigma_j = 1$ ) bond around each triangular plaquette. Because of the magnetoelastic coupling, energy may be lowered by stretching the frustrated bonds by a factor  $f$  and compressing the satisfied ones by a factor  $s$ . We fix the area of each resulting isosceles triangle to be that of the initial equilateral plaquette with sides  $a$ , and thus we can parametrize the deformation by the head angle  $\beta$  (see Fig. 1B, Inset):  $f(\beta) = 3^{1/4}(\tan \frac{\beta}{2})^{1/2}$ ,  $s(\beta) = 3^{1/4}(2 \sin \beta)^{-1/2}$ . Minimizing the energy (Eq. 1) with respect to  $\beta$  yields (see *SI Appendix*)

$$(2s' - f')Jc + [2(s - 1)s' + (f - 1)f']Ka = 0. \quad [2]$$

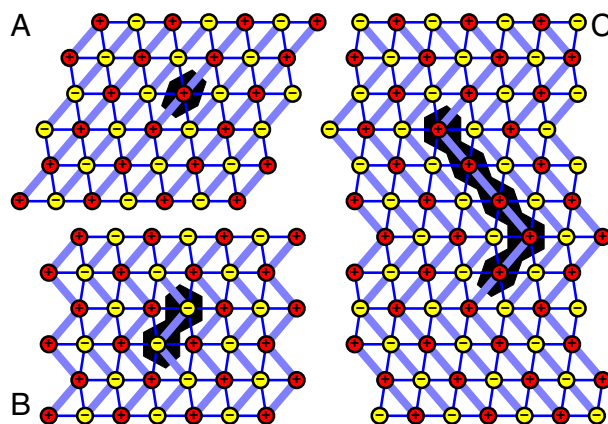
Fig. 1B shows how the triangles deform from  $\beta = 60^\circ$  toward  $\beta = 180^\circ$  as the ratio  $b \equiv \frac{Jc}{Ka}$  of the magnetoelastic interaction strength to the lattice rigidity grows.

Thus, each plaquette of the triangular lattice would minimize its energy by deforming into an isosceles triangle. We now show how this can be accommodated in the ground state by global deformations of the system. In the rigid triangular lattice, because each triangle must have exactly one frustrated bond, five nearest-

neighbor configurations are allowed in the ground state (Fig. 1C) (39). Requiring that the angle opposing each frustrated bond deforms to  $\beta > 60^\circ$  selects configurations (iii) and (iv) (22, 24), which give rise to zigzagging stripes. Thus, selecting a stripe of frustrated bonds defines a ground state of the lattice. It can be constructed by starting with a row of alternating spins (along the horizontal axis) and stacking copies of this row (along the vertical axis), as shown in Fig. 2. The intrarow and interrow separations are  $sa$  and  $sa \sin \beta$ , respectively, and the lateral shifts are determined by the arbitrary polarity of each row with respect to the one preceding it. Because these rows of alternating spins may be in any of the three principal directions of the network, and for a system of  $N$  particles, each of the  $\sqrt{N}$  rows may be in one of two states, the ground-state degeneracy is  $3 \cdot 2^{\sqrt{N}}$ .

This partially disordered ground state is realized by deformations that may vary rapidly in space; if deformations are assumed to be homogeneous (5) or to vary slowly (6), straight stripes are selected. In particular, the zigzagging stripes that minimize our microscopic Hamiltonian (Eq. 1) have a higher energy than straight stripes in the coarse-grained Hamiltonian considered in ref. 6. The ground state of zigzagging stripes that we find here is precisely the state that maximizes the packing density of buckled spheres (22, 24), which constitutes the connection of our model to the colloidal system.

Given the high ground-state degeneracy and the fact that it takes a discrete energy (of order  $J$ ) to flip a spin in the ground state, one might naively expect to find a stable phase of randomly zigzagging stripes at sufficiently low temperature ( $k_B T \ll J$ ). However, at positive temperature, the entropy of particle fluctuations around the energy-minimizing position is different in the different ground-state configurations. We will show that the state with straight stripes has lower free energy (or greater entropy) than other states, making it the stable thermodynamic phase at arbitrarily low temperature. Nonetheless, the resulting free-energy differences between the different zigzagging realizations are much smaller than the free-energy barriers between the ground-state configurations, and the system typically falls into disordered metastable configurations.



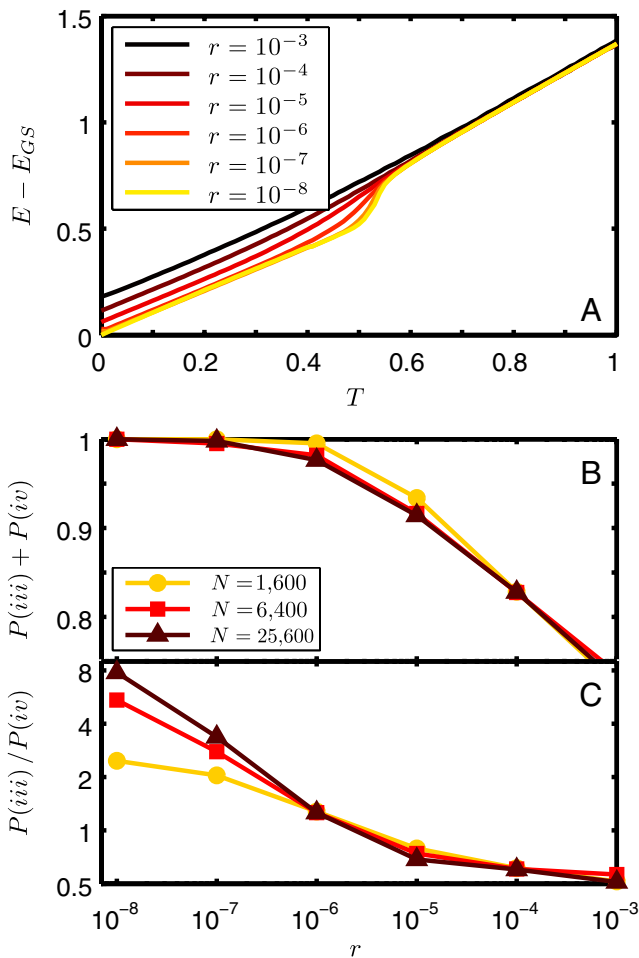
**Fig. 2.** Zigzagging stripes. Ground-state configurations are generated by stacking layers of particles with alternating spin and with arbitrary relative polarities between successive layers. The figure shows only simple configurations with periodic sequences of the straight (iii) and bent (iv) segments, as defined in Fig. 1C. However, ground-state configurations do not necessarily have a finite unit cell. (A) Straight stripes, for which all particles are in the state (iii). (B) Bent stripes, for which all particles are in state (iv). (C) Zigzagging stripes with a more complicated unit cell comprised of both (iii) and (iv). The shaded regions represent the unit cells used for the low-temperature expansion explained in the text. A, B, and C have one, two, and five particles per unit cell, respectively. Thick blue lines represent the stretched frustrated bonds, and thin blue lines represent the compressed bonds for which the antiferromagnetic interaction is satisfied.



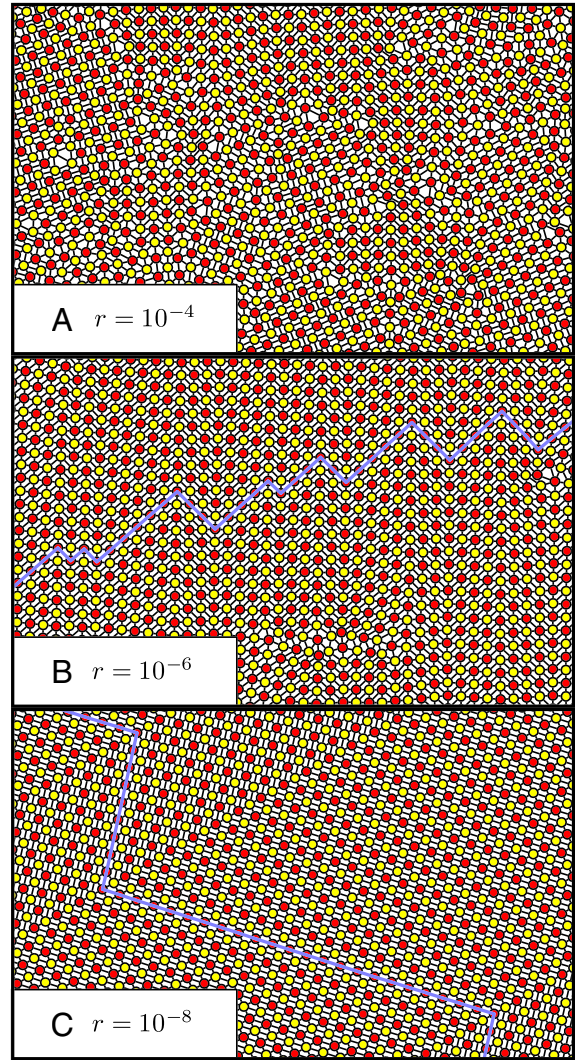
### Monte Carlo Simulations

To demonstrate the peculiar slow dynamics of this model, we used Monte Carlo simulations in which individual particles can flip their spin or move continuously in the plane and in which the simulation box may change its shape to accommodate the global deformations of the network (24). We start from a disordered state at high temperature, and we slowly cool the system at a rate  $r$  (time is measured in attempted Monte Carlo steps per particle). Fig. 3A shows that at high temperature the system follows an equilibrium curve irrespective of cooling rate, whereas below a certain temperature ( $T \approx 0.6$  for the parameter values shown here), the system's energy has a clear cooling rate dependence. Apparently, if the cooling is slow enough ( $r \leq 10^{-6}$  here), the system manages to reach the ground state.

Fig. 4A shows that indeed if the system is cooled too rapidly ( $r = 10^{-4}$ ) it falls into a disordered state with multiple small domains of a local stripy structure. For a slower cooling rate ( $r = 10^{-6}$ , Fig. 4B), the system finds a ground-state configuration with zigzagging stripes, such that the local configurations (iii) and (iv) defined in Fig. 1C are roughly equally represented, thus



**Fig. 3.** Monte Carlo simulations. (A) Energy  $E$  per particle above the ground-state energy  $E_{GS}$  vs. temperature  $T$  for different cooling rates  $r$  as indicated in the legend. System size is  $N = 6,400$ . Similar results were obtained for  $N = 1,600$  and  $N = 25,600$ . (B and C) Probabilities of finding the local configurations (iii) and (iv) defined in Fig. 1C at  $T = 0$  following cooling at different rates  $r$  for various system sizes  $N$ , as indicated in the legend. Note the logarithmic scale for the ratio  $P(iii)/P(iv)$  in C. Error bars are smaller than the symbols. Model parameters in all simulations are  $J = 1$ ,  $\epsilon = 2$ ,  $K = 8$ , and  $a = 1$ . Thus  $b \equiv \frac{\epsilon}{Ka} = 0.25$ . This yields a deformation angle of  $\beta = 86^\circ$  in the ground state and a difference in the free-energy coefficient, Eq. 4, of  $\mathcal{A}_0 - \mathcal{A}_1 = 0.025$  between straight and bent stripes.



**Fig. 4.** Ordering with decreasing cooling rate. Portion of the system in its final configuration after cooling at rates  $r = 10^{-4}$  (A),  $10^{-6}$  (B), and  $10^{-8}$  (C). Model parameters are  $J = 1$ ,  $\epsilon = 2$ ,  $K = 8$ , and  $a = 1$ . Thus,  $b \equiv \frac{\epsilon}{Ka} = 0.25$ , and in the ground state each triangular plaquette is deformed to an isosceles with head angle  $\beta = 86^\circ$ . System size is  $N = 25,600$ . For the fastest cooling rate, the system falls into a disordered state (A), for which the fraction of particles in the local configuration of a straight stripe (see Fig. 1C) is  $P(iii) = 0.31$  and the fraction in that of bends is  $P(iv) = 0.52$ . For the slower cooling rates, zigzagging stripes are formed (B and C). The blue lines are guides to the eye, which highlight a line of frustrated bonds, and emphasize that as the cooling rate is decreased from B to C, the stripes become straighter. In B,  $P(iii) = 0.54$  and  $P(iv) = 0.44$ , whereas in C,  $P(iii) = 0.9$  and  $P(iv) = 0.1$ .

corroborating this phase as randomly zigzagging stripes. Fig. 4C shows that for even slower cooling ( $r = 10^{-8}$ ), there is preference for ground-state configurations in which the stripes are more straight than bent, namely configuration (iii) is preferred over (iv). This is quantified in Fig. 3 B and C, which shows results obtained by averaging over multiple realizations for each system size and cooling rate. For very fast cooling, the system remains quite disordered with  $P(iii)/P(iv) \approx 0.5$ , in accordance with the combinatoric weights of these two local configurations. For  $r \leq 10^{-6}$ , the system manages to find its ground state, as seen by the fact that  $P(iii) + P(iv) = 1$ , namely all particles are in one of these two states. For  $r = 10^{-6}$  these two states are equally probable,  $P(iii) \approx P(iv)$ , as one would expect for randomly zigzagging stripes. However, as the cooling rate is decreased even more,



dence of the free energy  $\mathcal{F}$  in Eq. 3. At  $T = 0$ , entropy is irrelevant and all ground-state configurations are equivalent, but at  $T > 0$  this degeneracy is lifted and the configuration with the minimal free energy is selected. The frequencies of all vibrational modes scale as  $\omega_k^2 \propto K/M$ , where  $M$  is the mass of each particle. The normalized spectrum depends on the spin state  $\{\sigma_i\}$  and on the deformation angle  $\beta$ , which is set by  $b \equiv \frac{J_c}{K\alpha}$  (see Eq. 2 and Fig. 1B). Hence we define

$$\mathcal{A}(\{\sigma_i\}, \beta) \equiv \frac{1}{N} \sum_k \log \left( \sqrt{\frac{M}{K}} \omega_k \right) \quad [4]$$

and write  $\mathcal{F} = Nk_B T [\mathcal{A}(\{\sigma_i\}, \beta) + \text{const.}]$ . Thus minimizing  $\mathcal{F}$  is equivalent to minimizing  $\mathcal{A}$ .

We can analytically calculate the normal modes of vibrations of the deformed lattice for any ground state with a periodic repetition of straight (iii) and bent (iv) segments (see *SI Appendix*). In such cases the normal modes are phonons and the index  $k$  labeling them may be associated with their wave vector. The free-energy coefficient,  $\mathcal{A}$ , may then be obtained by numerically summing over the appropriate Brillouin zone. Fig. 2 highlights the unit cells in ground states with periods of one, two, and five particles. Now we consider ground states with larger unit cells and show that the free energy is mainly determined by the fraction  $p_s \equiv P(\text{iii})/[P(\text{iii}) + P(\text{iv})]$  of straight segments. To clarify which configurations we used in these calculations, we show in Fig. 5 several representative unit cells. For example,  $p_s = 1/3$  in a single configuration with three particles in its unit cell (Fig. 5A), but also in two distinct configurations with six particles (Fig. 5B), in six with nine particles (Fig. 5C), and in an increasing number of other configurations with larger unit cells. Similarly, for  $p_s = 1/2$ , we show the two configurations with four particles per unit cell in Fig. 5D and the six with eight particles in Fig. 5E.

Fig. 6A–C shows the results obtained by numerically evaluating Eq. 4 for ground states with unit cells consisting of up to 13 particles, plotted vs.  $p_s$  (see *SI Appendix* for more details and Tables S4–S5 for the entire data plotted). We find that the free energy is bounded between the extreme cases of straight stripes ( $p_s = 1$ , Fig. 2A) and bent stripes ( $p_s = 0$ , Fig. 2B). Moreover, the approximate collapse of the results for all calculated unit cells to a single curve provides strong support for the hypothesis that only the ratio between straight and bent segments is important and not the order in which they are positioned or the period of the pattern they form. Finally, the roughly linear dependence of  $\mathcal{A}$  on  $p_s$  indicates that the free energy may be approximated as a linear combination of contributions from the straight and bent segments. Because the ground state is determined by a one-dimensional sequence of straight and bent segments, this result implies that the system may be well approximated by an effective one-dimensional noninteracting Hamiltonian. For  $\beta = 155^\circ$  (Fig. 6C) we observe substantial deviations from the prediction of such a noninteracting Hamiltonian. It would be interesting to understand whether these deviations result from our limited numerical accuracy in evaluating so small free-energy differences, or from additional considerations that affect the physics of this system at such large deformations.

In our elastic Ising model, the low-temperature free energy of in-plane positional fluctuations is determined by the dimensionless ratio  $b \equiv \frac{J_c}{K\alpha}$  between the magnetoelastic interaction strength to the lattice rigidity. This ratio sets the deformation angle  $\beta$  of each isosceles triangle in the ground state (see Eq. 2 and Fig. 1B). In the corresponding system of buckled colloids, the deformation angle  $\beta$  of each isosceles plaquette in the close-packed state (which is equivalent to the Ising model's ground state) is similarly dictated by the ratio of each sphere's diameter to the separation between the confining walls (24). We expect the colloidal system

to exhibit a similar order-by-disorder effect that will be governed by the deformation angle  $\beta$ , and therefore we plot in Fig. 6D the free-energy difference  $\mathcal{A}_0 - \mathcal{A}_1$  between straight and bent segments vs. the geometrical parameter  $\beta$  rather than vs. the physical parameter  $b$ .

For extremely rigid lattices that hardly deform ( $\beta \approx 60^\circ$ ), straight and zigzagging stripes are almost equivalent in terms of their particle displacements and therefore  $\mathcal{A}_0 \approx \mathcal{A}_1$ . As  $\beta$  increases, straight stripes develop an entropic advantage that comes from the fact that the straight-stripe ground state is the most anisotropic and hence has the most nonuniform distribution of eigenfrequencies, and thus the maximal entropy (13). For extremely large deformations ( $\beta > 100^\circ$ ) we observe a decrease in  $\mathcal{A}_0 - \mathcal{A}_1$  that reaches a minimal value at  $\beta \approx 155^\circ$  and then increases again as  $\beta \rightarrow 180^\circ$ . In the *SI Appendix* we show how this nonmonotonic behavior results from the numerical structure of the dispersion relations. In particular, we identify in Fig. S10 the region in reciprocal space that dominates the free-energy difference between straight and bent stripes. The magnitude of the free-energy difference in this region grows monotonically with  $\beta$ ; however, the size of this region decreases, which gives rise to the nonmonotonic behavior seen in Fig. 6D for  $60^\circ \leq \beta \leq 155^\circ$ . For  $\beta \geq 155^\circ$  the free-energy difference has a different wave-vector dependence (see Fig. S10) that gives rise to the second increase in the total free-energy difference at such large deformations. It would be interesting to understand theoretically the deeper origins and possible implications of this nonmonotonic behavior. Note that for large values of  $\beta$ , the distance to some of the next-nearest neighbors becomes smaller than the distances between nearest neighbors. This introduces further complications beyond the analysis presented in this paper, which assumes only nearest-neighbor interactions. Overall, our numerical results show that for all ground-state deformations, straight stripes are preferred entropically; however, their entropic advantage is extremely small. We observed the same qualitative behavior when allowing the system's total volume to vary.

Before concluding we note that the free-energy coefficient  $\mathcal{A}$  given in Eq. 4 and plotted in Fig. 6 is defined for each specific realization of the stripes. Randomly zigzagging stripes that mix straight and bent segments are highly degenerate, and their free energy possesses also a configurational entropy (or entropy of mixing) that competes with the vibrational entropic advantage of straight stripes that we found. However, due to the one-dimensional character of the ground state, this configurational entropy is subextensive in system size and scales as  $\sqrt{N}$ . The vibrational free energy of Eq. 4 enters as an extensive quantity that scales linearly with  $N$  in  $\mathcal{F}$  and is thus dominant in the thermodynamic limit. As a result, straight stripes are favored. This may be the reason for the limited ability of our simulations to reach perfect straight stripes, and for the smaller values of  $P(\text{iii})/P(\text{iv})$  for smaller systems in Fig. 3C. To test this, we repeated the simulations at parameter values where the difference in  $\mathcal{A}$  between straight stripes and zigzags is smaller and indeed found a weaker preference for straight stripes.

## Summary and Conclusions

In summary, our exact microscopic description of this previously studied (5, 6) elastic Ising model reveals a highly degenerate, partially disordered ground state. We find a much richer behavior because entropy lifts the degeneracy at any positive temperature. However, large free-energy barriers between the ground-state configurations induce a glassy phase of zigzagging stripes. Although equilibration is strongly hindered in this glassy phase, the straight-stripe structures obtained in it result from the minute entropic differences in the equilibrium free energies of the various stripe realizations. The fact that this model is amenable to analytic treatment makes it appealing as a prototypical model for studying such order-by-disorder phenomena in a broader



context. Moreover, the current approach to experimentally studying frustration relief by lattice deformations in antiferromagnets is based on quite indirect measurements of lattice deformations (45, 46). Our work makes direct contact to a colloidal system (22) in which frustration and its relief are governed by similar physical mechanisms, yet it has the advantage that local deformations can be directly measured in it. Interestingly, several recent experimental works are focused on measuring the normal modes of vibration in colloidal systems (47–50). On top of understanding deformable antiferromagnets and mesoscopic model systems for

them, we expect that results obtained for our system will shed light on questions such as the statistical mechanics of sphere packings and the physical origins of glassy dynamics.

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