Exact Equilibrium Shapes of Ising Crystals on Triangular/Honeycomb Lattices

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The anisotropic surface tension for an Ising system below the critical point on a triangular or a honeycomb lattice can be computed through duality. Using the Wulff construction, the equilibrium shape of a crystal (droplet of one phase inside a sea of the other) is found. An exact and simple equation for this shape is derived.

1. INTRODUCTION

Below its critical point, a two component, Ising-like system may display phase coexistence, with an interface separating the phases. The surface tension, the free energy per unit area of the interface, is in principle calculable from the microscopic Hamiltonian within the standard framework of statistical mechanics. In practice, however, even the *bulk* free energy is not susceptible to exact analysis of this kind, except for two-dimensional systems on certain regular lattices. The problem of finding the surface tension on these lattices is more complex, since the tension is generally *anisotropic*. That is, it is not a single number like the bulk energy, but a function $\sigma(\mathbf{n})$ of $\mathbf{n}$, the normal to the (planar) interface. With a recent increase in interest[1-3] in interfacial physics and crystal shapes, the question of finding these anisotropic tensions once again has attracted attention.

For the square Ising model, Onsager undoubtedly knew the anisotropic tension, although he only published the result for two specific

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orientations.[4] Seeveral authors[6,7,13] discovered (and rediscovered) it, ending with an explicit formula.[7] Here, we give a formula for $\sigma(\mathbf{n})$ in the triangular/honeycomb Ising model.

One application, for a given $\sigma(\mathbf{n})$, is to construct the associated minimum energy surface via geometric methods of Wulff[8] or analytic ones of more recent authors.[9 11] Such a surface is believed to describe the average shape of a (macroscopic) droplet of one phase in equilibrium with a background of the other. For historical reasons, these are called "equilibrium crystal shapes." Here, we use the $\sigma(\mathbf{n})$ appropriate for the triangular/honeycomb Ising model to construct hexagonal "Ising crystals" in both cases. Apart from the figures, which are found necessarily by numerical methods, we present a simple (and exact) equation for the shapes.

The outline of this paper is as follows. The setup, the duality transformation, and the surface tension function are given in Section 2. Following a brief review of the relationship between an anisotropic surface tension and the minimum energy shape, we show the Ising crystals in Section 3. A summary and some remarks form the concluding section.

2. SURFACE TENSION IN TRIANGULAR/HONEYCOMB ISING MODELS

First, we study an Ising system with nearest neighbor coupling on a honeycomb lattice. The triangular case, related to this by a decimation (or a $\Delta-Y$ transformation), will follow as an easy generalization. The most homogeneous system of this kind would have three distinct coupling energies E_α ($\alpha = 1, 2, 3$; see Fig. 1). For the sake of clarity, we restrict ourselves to equal energies ($E_\alpha = E$) during the derivation of $\sigma(\mathbf{n})$. Then we give a result for the general case at the end.

Following standard notation, we define the coupling

$$K \equiv \beta E \equiv E/k_{\rm B} T \tag{1}$$

so that "below criticality" means $K > K_c$. For the surface tension, we should in principle apply boundary conditions $+/-$ to induce an interface between the two phases that would coexist on the low-temperature system. By a gauge transformation, this problem can be changed to one with an antiferromagnetic seam[4],2 *and homogeneous* (say, all $+$) boundary conditions. Now, in principle we need to study a system with a seam from one edge to another and take the thermodynamic limit appropriately. In prac-

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2 For a rigorous proof that an antiferromagnetic seam may also be used instead of $+/-$ boundary conditions, see Ref. 5.
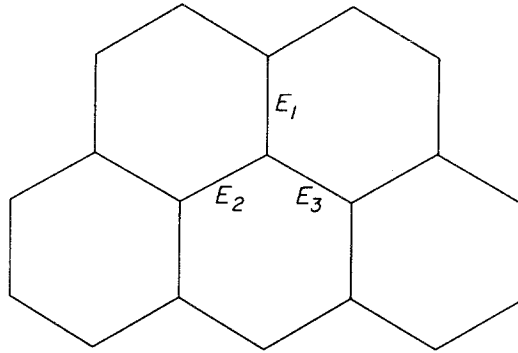
Fig. 1.   Honeycomb lattice with Ising nearest neighbor energies E_i.

tice, it is easier to consider an infinite system with a seam stretching between a hexagon and another one a distance R away. Then, the excess free energy of this system over the homogeneous one, divided by R, is expected to approach a constant as $R \to \infty$. In general, it will depend on the orientation of the line joining the hexagons, i.e., the interface between the $+/-$ phases. Characterizing the orientation by $\mathbf{n}$, the normal to the "interface," this excess energy per unit length is defined as the anisotropic surface tension $\sigma(\mathbf{n})$.

Obviously, to find $\sigma(\mathbf{n})$ on the original system is no easy task. Fortunately, it is related to the correlation length ξ on the dual system and that length can be extracted from the known[12] two-spin correlation function. The dual system relevant here is an Ising model on a triangular lattice with coupling K^*, which is related to K by

$$\tanh K^* = \exp(-2K) \tag{2}$$

Thus, for $K > K_c$, the dual system will be above *its* criticality, i.e., $K^* < K_c^* = \operatorname{arctanh}(2 - \sqrt{3})$. (In general, there is an E_α^* dual to each E_α; see Fig. 2). Now, the relation between σ and ξ is extremely simple,[13]
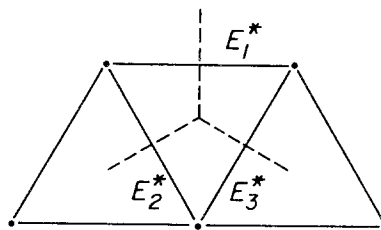
$$\beta\sigma(\mathbf{n}) = 1/\xi(\mathbf{u}) \tag{3}$$

Fig. 2.   The triangular dual lattice with energies E_i^*. Dashed lines represent bonds on the honeycomb lattice.

where **u** is the unit vector normal to **n**. Note that ξ depends on T and E via the duality condition (2).

To be more specific, associated with each hexagon is a site on the dual lattice. Thus, the two ends of the antiferromagnetic seam correspond to two sites on the triangular lattice. Label these $(0, 0)$ and (M, N), so that

$$R\mathbf{u} = M\mathbf{e}_1 + N\mathbf{e}_2 \tag{4}$$

where the $\mathbf{e}_i$ are the unit basis vectors on the dual lattice. (See Fig. 3. We normalize all physical distances by the lattice spacing on the triangular one.) Since $\mathbf{e}_1 \cdot \mathbf{e}_2 = -\frac{1}{2}$, $R^2 = M^2 + N^2 - MN$. The correlation length may be extracted from the correlation function $\langle SS \rangle$ via

$$1/\xi(\mathbf{u}) = \lim_{R \to \infty} -\ln \langle S_{00} S_{MN} \rangle / R \tag{5}$$

and is orientation dependent in general.

The full $\langle SS \rangle$ is quite complex, involving many terms and factors of integral representations [see Eqs. (5)–(7) in Vaidya's paper[12]]. For the leading large M, N behavior of $\langle SS \rangle$, we need only one factor,

$$\iint_{-\pi}^{\pi} d\phi_1 \, d\phi_2 \exp(-iM\phi_1 - iN\phi_2)/\Delta(\phi_1, \phi_2) \tag{6}$$
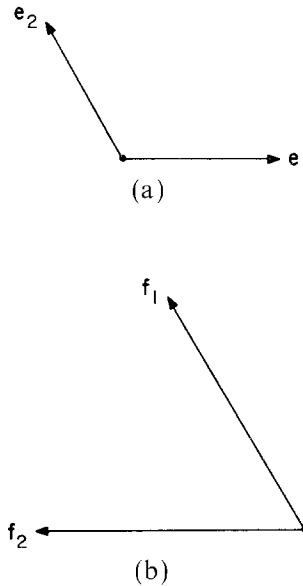

(a)

(b)

Fig. 3.   (a) Basis (normalized) vectors for the dual triangular system. (b) Basis vectors for the original honeycomb system. $\mathbf{f}_i \equiv h_{ij}\mathbf{e}_j$ rotated by 90°.

and the value of the integrand at the saddle point. Since overall constants are unnecessary, we write Δ in a simplified fashion:

$$\Delta = A - \cos\phi_1 - \cos\phi_2 - \cos(\phi_1 + \phi_2) \tag{7}$$

where

$$A \equiv [(\cosh 2K^*)^3 + (\sinh 2K^*)^3]/\sinh 2K^* \tag{8}$$

One can check that $A \geqslant 3$, with the equality holding only at criticality. Thus, the saddle point occurs at complex $\phi_i = -i\psi_i$ (Latin indices running over 1, 2 only), with the ψ's satisfying

$$F(\psi_1, \psi_2) \equiv A - \cosh\psi_1 - \cosh\psi_2 - \cosh(\psi_1 + \psi_2) = 0 \tag{9a}$$

and

$$\frac{M}{N} = \frac{\sinh\psi_1 + \sinh(\psi_1 + \psi_2)}{\sinh\psi_2 + \sinh(\psi_1 + \psi_2)} = \frac{m_1}{m_2} \tag{9b}$$

The equality in (9a) is actually an asymptotic condition, i.e., $F = O(1/R)$. So, to leading order, $-\ln\langle SS \rangle$ is just $M\psi_1 + N\psi_2$. In (9b) we use the equivalents of sines and cosines for a triangular coordinate system, defined by

$$m_1 \equiv M/R \qquad \text{and} \qquad m_2 \equiv N/R \tag{10}$$

Thus the m's depend on $\mathbf{u}$ and $\mathbf{e}$ through $\mathbf{u} = m_i \mathbf{e}_i$ (summation implied). Explicitly, we have

$$m_i = h_{ij}(\mathbf{e}_j \cdot \mathbf{u}) \tag{11}$$

where $h_{11} = h_{22} = 2h_{12} = 2h_{21} = 4/3$ is the inverse of the metric on the triangular system. Since $\mathbf{u}$ is just $\mathbf{n}$ rotated by 90°, we define, for later convenience, $\mathbf{f}_i$ to be $h_{ij}\mathbf{e}_j$ rotated by 90° (Fig. 3b). Then, (11) becomes

$$m_i = \mathbf{f}_i \cdot \mathbf{n} \tag{12}$$

showing the explicit dependence of the m's on $\mathbf{n}$.

Summarizing, we have

$$1/\xi(\mathbf{u}) \sim -\ln\langle SS \rangle/R \sim m_i\psi_i \tag{13}$$

and

$$\beta\sigma(\mathbf{n}) = m_i\psi_i \tag{14}$$

where the ψ's depend on $\mathbf{n}$, E, and T via Eqs. (9a), (9b). Note that, although these equations appear transcendental, they are in fact algebraic ones for cosh ψ_i. Unfortunately, the equation for each is sixth order and no simple explicit form like that for the square Ising case is found. Nevertheless, as will be shown in the next section, a simple explicit equation for the equilibrium shape can be derived.

For completeness, we simply note that if the E_α are not all equal, the form (14) would still be true with modifications for Eq. (9a). For the general case, we have

$$H_1 \cosh \psi_1 + H_2 \cosh \psi_2 + H_3 \cosh(\psi_1 + \psi_2) = 1 \qquad (15)$$

where

$$H_1 = 2Z_2(1 - Z_1^2)(1 - Z_3^2)/D$$

$$H_2 = 2Z_1(1 - Z_2^2)(1 - Z_3^2)/D$$

$$H_3 = 2Z_3(1 - Z_1^2)(1 - Z_2^2)/D$$

$$D = (1 + Z_1^2)(1 + Z_2^2)(1 + Z_3^2) + 8Z_1 Z_2 Z_3$$

$$Z_\alpha = \exp(-2\beta E_\alpha)$$

We conclude this section with some remarks about surface tensions on the Ising model on a triangular lattice. Note that this system must also be below its criticality, not to be confused with the dual triangular system. It is well known[14] that, by decimating every other spin on a honeycomb system, one arrives at a triangular one (a $\Delta-Y$ transformation). Although microscopically there is a difference between these two systems, surface tensions and correlation lengths are macroscopic quantities. For example, on a finite hexagonal system, it is possible to place the $+/-$ boundary conditions so that the interface is pinned between a spin that is to be decimated and one that is not. Ambiguity for the triangular system, in principle, arises. But, in the thermodynamic limit, when we are interested in an energy that goes to infinity like R, we could presumably just as well consider an interface pinned only by surviving spins. Now the ambiguity disappears.

Another concern may be the antiferromagnetic bonds introduced to facilitate the calculation of surface tensions. This question is related to the one above, in that there are two types of triangles: up/down-pointing ones. To make a honeycomb system out of a triangular one, only one of the two types needs decoration (inverse of decimation). Thus, as long as the two ends of the antiferromagnetic seam terminate in the same type of triangle, every triangle of the other type would have two antiferromagnetic bonds
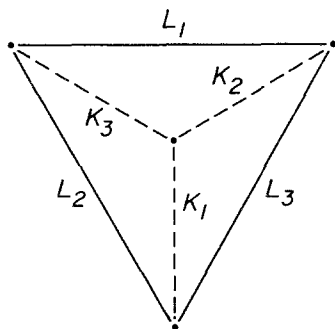
Fig. 4. Decorating a triangle with energies L_α/β.

(i.e., not frustrated) and may be decorated. We give the example, for completeness, of decorating down-pointing triangles (Fig. 4). If the original couplings are L_α and $\omega_\alpha \equiv \tanh(L_\alpha)$, then decorated ones are given by

$$\tanh K_1 = (\omega_1 + \omega_2\omega_3)/(1 + \omega_1\omega_2\omega_3)$$

and two other equations with 123 permuted cyclically. From such equations, it is clear that, as long as two (or no) L's are negative, two (or no) K's will be negative and real. By contrast, decoration with real K's cannot be performed on a frustrated triangle.

3. EQUILIBRIUM HEXAGONAL ISING CRYSTALS

Given a $\sigma(\mathbf{n})$ and a fixed volume V, Wulff[8] has shown how to construct a minimum-energy surface S that encloses V. Although this beautiful geometric construction has much intuitive appeal, it is not very useful in extracting analytic results. There is an alternative route—the parametric representation. Originally due to Burton et al.[9] in work connected with ledge shapes in two dimensions, this representation was generalized to higher dimensions in a coordinate-invariant way.[10,11]

The basic idea is to write $\mathbf{X}$, the position vector associated with a point on S, as a function of $\mathbf{n}$, the normal of the surface at that point. Thus, $\mathbf{n}$ acts a parameter and $\mathbf{X}$ sweeps out S as $\mathbf{n}$ varies over the unit sphere. There are no caveats provided $\sigma(\mathbf{n})$ is a smooth (twice-differentiable) function and the resultant S is not self-intersecting. If these conditions are not satisfied, there would be facets and corners in S and there are standard methods to supplement the parametric equations.[10,11] For our case, there are no facets or corners (except at the limiting case of $T = 0$) and we will stay with the simple equation. It is

$$\mathbf{X}(\mathbf{n}) = \lambda'\{\mathbf{n}\sigma(\mathbf{n}) + \nabla\sigma(\mathbf{n})\} \tag{16}$$

Here, the gradient is taken with respect to **n**. In practice, it is convenient to take the derivative as if **n** is not normalized and then project out the **n** component with $\mathbf{I} - \mathbf{n} \otimes \mathbf{n}$. Finally, λ' is a scale that can be used to satisfy the volume constraint.

From (14), it is clear that $\beta\sigma$ is the natural, dimensionless function to consider. Thus, defining $\lambda \equiv \lambda'/\beta$, we have

$$\mathbf{X}(\mathbf{n}) = \lambda\{m_1\psi_i\mathbf{n} + \nabla m_i\psi_i\}$$

with λ being a pure length scale factor. This equation simplifies even further due to $m_i \nabla\psi_i = 0$, which follows from taking the gradient of (9a), using the chain rule, and applying (9b). Thus,

$$\mathbf{X}(\mathbf{n}) = \lambda\{m_i\mathbf{n} + \nabla m_i\}\,\psi_i$$

But the m's are just linear functions of **n**, so that $\nabla m = \mathbf{f} - \mathbf{n}(\mathbf{n} \cdot \mathbf{f})$, which is just $\mathbf{f} - \mathbf{n}m$. We arrive at the final result:

$$\mathbf{X}(\mathbf{n}) = \lambda\psi_i\mathbf{f}_i \tag{17}$$

These equilibrium shapes[3] are shown in Fig. 5 with $\lambda = 1/\beta$, for various values of T. If the volume is fixed as V, then one only needs to scale λ appropriately. Specifically, if we denote the area inside a $\lambda = 1/\beta$ curve as W, then setting $\lambda = (V/W)^{1/2}$ will give the correct crystal shape.

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[3] These shapes resemble those obtained by Slotte and Hemmer,[20] who considered an *antiferromagnetic* Ising model in an external field, a model with very interesting properties. Distinct from the model studied here, the resemblence is entirely coincidental.
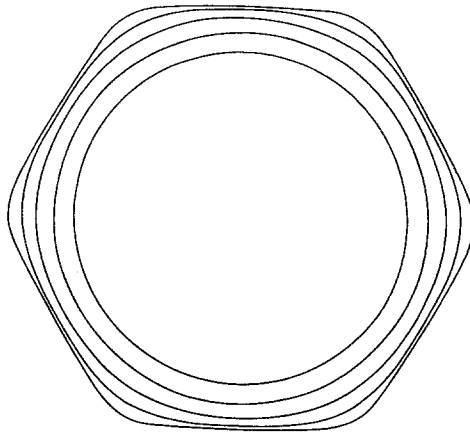

Fig. 5.   Equilibrium shapes of Ising crystals for some temperatures. The outermost figure is for $T = 0.1T_c$; successive inner ones represent $0.2T_c$, $0.3T_c$, $0.4T_c$, and $0.5T_c$.

Although $\lambda = 1/\beta$ may be thought of as a mere convenience, especially since the different shapes do not overlap in such a representation, the various figures in fact carry further information. In particular, distances here have units of energy (per lattice spacing). Thus, W represents the square of a surface tension. From the general result of Ref. 11, the total surface energy of an equilibrium crystal, with any V, is given simply by $(2WV)^{1/2}$ (in two bulk dimensions). Other information concerns certain points on each figure: those furthest and nearest to the center. At these points, the surface tension and equilibrium shape curves coincide in the Wulff construction. Thus, the distances from the center to these points are in fact the tensions for those normal directions. Furthermore, for $T = T_c/10$ (the outermost figure), the tension for a surface breaking the least bonds per unit length, i.e., a "near facet," is practically $2E$. So, although for the sake of clarity both the surface tension curves and the axes are omitted, this information gives a clear sense of the energy scale. As expected, since the figures carry information on surface tension instead of the physical volume, they shrink monotonically with temperature, approaching zero as T_c is reached.

Due to the extremely simple dependence of the equilibrium shape on the ψ's, it is possible to give another representation of these curves, one analogous to $x^2 + y^2 = r^2$ for a circle. Not surprisingly, we need coordinates natural to our system: the familiar triangular ones $\{x_1, x_2, x_3\}$ satisfying

$$x_1 + x_2 + x_3 = 0 \tag{18}$$

Figure 6 shows that a point given by $\mathbf{X} = \lambda \psi_i \mathbf{f}_i$ would have $x_i = \lambda \psi_i$, so that the desired equation is just

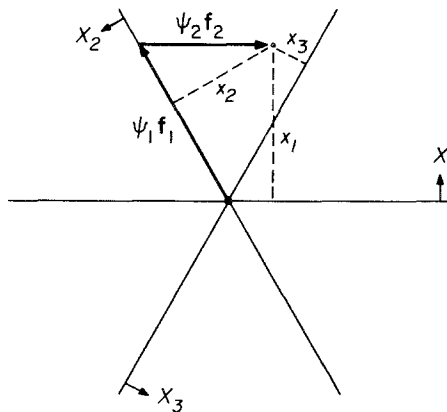$$\cosh(x_1/\lambda) + \cosh(x_2/\lambda) + \cosh(x_3/\lambda) = A \tag{19}$$

Fig. 6.   Triangular coordinates $\{x_\alpha\}$.

In addition to the explicit figures, let us analyze this formula directly near $T = T_c$ and 0, and check that it interpolates between a circle and a hexagon. Recall that A approaches 3 as $T \to T_c$. Thus, x_α/λ must be small and only two terms in each cosh contribute, leading to the equation

$$x_1^2 + x_2^2 + x_3^2 = \text{const} \tag{20}$$

Note that the x's are not small; they are dictated by the volume. However, λ does get large. That this is the equation of a circle in triangular coordinates can easily be checked. So, isotropy in the leading critical behavior is recovered.

On the other hand, A goes to infinity as $T \to 0$, and the x_α/λ values must also get large (again, this is achieved by λ becoming small and not by x getting large). However, for most values of the x's, their disparity means that one of the three terms in (19) saturates the left-hand side. Thus, major portions of the figure are given by one of the x's being nearly a constant while the other two vary. The crossover from one x_α to another occurs only when those two are (in the limit $T = 0$) equal. Such a figure is a hexagon.

We conclude this section by quoting the equation for the droplets if the E_α are not all equal. Referring to Eq. (15) and those following, the generalized equation (19) is

$$H_1 \cosh(x_1/\lambda) + H_2 \cosh(x_2/\lambda) + H_3 \cosh(x_3/\lambda) = 1 \tag{21}$$

4. CONCLUDING REMARKS AND SUMMARY

Before the summary, we point out certain highlights associated with these exact equilibrium crystal shapes.

4.1. Facets, Corners, Roughening, and All That

As expected,[9,15] in two-dimensional systems with short-ranged interactions, the roughening transition occurs at $T = 0$. Here, we confirm that result by the explicit, facetless shape [Eq. (19)] found for all $T > 0$, since faceted interfaces are associated with temperatures below roughening (see, e.g., Ref. 16). To display the approach to a faceted shape at $T = 0$ analytically, we consider the curvature as a function of $\mathbf{n}$. Ordinarily a tensor[11] with $d - 1$ nontrivial eigenvalues, its content is contained, in our case of $d = 2$, in its trace. Since $\mathbf{X}(\mathbf{n})$ is available explicitly, the trace of its inverse is just $\nabla \cdot \mathbf{X} = \lambda \mathbf{f}_i \cdot \nabla \psi_i$. The last factors can be obtained by taking gradients of Eqs. (9a), (9b):

$$F_1 \nabla \psi_1 + F_2 \nabla \psi_2 = 0 \tag{22}$$

$$m_2(F_{11} \nabla \psi_1 + F_{12} \nabla \psi_2) - m_1(F_{21} \nabla \psi_1 + F_{22} \nabla \psi_2) = F_2 \mathbf{f}_1 - F_1 \mathbf{f}_2 \tag{23}$$

where the subscripts on F denote derivatives with respect to ψ_i. Define κ, the physical curvature multiplied by λ, so that it is also dimensionless. Reducing everything to $\gamma_i \equiv \cosh \psi_i$, we find

$$\kappa = \frac{A(1 - m_1 m_2) - \gamma_1 m_1 (m_1 - 2m_2) - \gamma_2 m_2 (m_2 - 2m_1)}{[(A - \gamma_1)(A - \gamma_2) + \gamma_1^2 + \gamma_2^2 - A - 3]^{1/2}} \tag{24}$$

For the direction associated with a facet, we have $m_1 = m_2 = 1$ and $\gamma_1 = \gamma_2 = [(2A + 3)^{1/2} - 1]/2$ and recognize that $A \to \infty$ as $T \to 0$. Thus, κ vanishes like $\exp(-E/k_B T)$ for this normal direction. Further, one verifies that, since one cannot reach temperatures below roughening, the entire facet develops at once at $T = 0$. We could also ask how the corners develop as $T \to 0$. Here, we have $\gamma_1 = (A - 1)/2$, $\gamma_2 = 1$, and $m_1 = 2m_2$, for example. Then, κ approaches a constant. To translate this result into the physical curvature, we need to recall that the scale factor λ approaches zero, so that, for a fixed volume, the curvature diverges. It is easy to check that $\lambda \sim T$ in this limit and the corner emerges via the radius of curvature going to zero like T.

4.2. Approach to Isotropy near T_c for Equal Couplings E_α

We have seen that, as T_c is approached, one manifestation of isotropy is in the circularity of the equilibrium crystal shape. Beyond that, we could ask how the deviation from isotropy vanishes with $t \equiv (T_c - T)$. The answer lies first in $A = 3 + O(t^2)$. Thus, x/λ is of the order of t. The corrections to circularity must come from the higher order terms in the expansion of the cosh function in Eq. (19). Explicitly, one can check that the x^4 terms combine into $(x_1^2 + x_2^2 + x_1 x_2)^2$, which is isotropic. This can be traced to the lack of an anisotropic quartic invariant in the permutation group of three objects. On the other hand, there is a cubic invariant, so that anisotropy emerges at the sixth order. Thus, the corrections to circularity first appears at $O(t^4)$. Consequently, by contrast to the square lattice system, where the first correction are at $O(t^2)$, these droplets display a more isotropic form for T far below T_c.

4.3. Scaling near T_c for Unequal Couplings E_α

The critical behavior of this Ising system is the same, according to universality,[17] as that of the square one. In particular, we expect isotropy to be restored as T_c is reached, for arbitrary ferromagnetic couplings E_α. This expectation turns out not to be true *unless* a general linear transformation on the spatial cordinates is performed. In the language of the renor-

malization group, these operations would be labeled redundant.[18] In our case of two-dimensional space, two parameters are available: the relative scale of x vs. y and a rotation. Thus, the general elliptical crystal shape near T_c can be brought into a circle. To be specific, refer to Eq. (21) and verify that $\sum H_i \to 1$, $\lambda \to \infty$, and the equation reduces to a homogeneous quadratic one in $x \equiv (-x_1 - 2x_2)/\sqrt{3}$ and $y \equiv x_1$. Diagonalize this quadratic form and rescale x, say, and arrive at the equation for a circle.

4.4. Nonscaling for Arbitrary T and General E_α

Like the square lattice case, the full shape equation (21), as opposed to the critical limit, cannot be cast into the equal E_α one [Eq. (19)] by any coordinate transformation. Nonlinear transformations spoil translational invariance, while linear ones cannot handle nonlinear terms. Unlike the square case, however, even the $T = 0$ limit cannot be transformed into a regular hexagon. This is clear, since the shape in this case consists of six facets with different lengths and 120° angled corners. So, apart from having two relative scale factors to contend with, rescaling x vs. y generally spoils the 120° angles.

Another distinct mansfestation of the general E_α case lies in corrections to scaling near T_c. Once the leading shape is transformed into a circle, the leading corrections are $O(t^2)$. By contrast, in the equal E_α case, this order is absent. The difference may be traced to the lack of permutation symmetry in this situation, so that the raison d'être for the absence of x^4 is missing.

In these ways, the equilibrium shape reveals the relative strength of the couplings and cannot be "transformed away" by any coordinate transformations of the underlying lattice structure.

To summarize, we have presented the exact equilibrium shapes of crystals of the Ising model on honeycomb/triangular lattices. As expected, for both lattices, the shapes are hexagonal at low temperatures, gradually rounding off to circles as criticality is approached. For all positive T, the shapes are smooth, while at $T = 0$, they have nothing but six facets and corners. The analytic form of these shapes is exceedingly simple. However, the anisotropic surface tension is not explicitly available, though it is given by elementary functions of the root of a sixth-order polynomial.

It would be interesting to carry out a Monte Carlo study on these lattices and check against the shapes, the surface tensions, and, more easily, the total surface energy associated with a droplet. So far, only the total energy on a square lattice has been studied,[19] where the data agree well with analytic calculations like the present.[11] With the vast improvements

of microcomputers since that Monte Carlo simulation, such a study should be easy to persue.

ACKNOWLEDGMENTS

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