Low-temperature phase of a stacked triangular Ising antiferromagnet

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We have investigated a model consisting of planes of Ising spins with antiferromagnetic nearest-neighbor interactions on a triangular lattice connected by nearest-neighbor ferromagnetic bonds in the third direction. It is shown that Landau theory, mean-field theory, and conventional low-temperature expansions are not reliable for this model. We have also studied a class of one-dimensional frustrated Ising models which have low-temperature expansions with irrational coefficients, indicating that the usual method of counting excitations about ground-state configurations cannot be used to construct their low-temperature series. The results from the one-dimensional models are used to obtain bounds on the free energy and information about the magnetization of the three-dimensional model at low temperatures.

I. INTRODUCTION

The two-dimensional nearest-neighbor antiferromagnetic Ising model on a triangular lattice is a classic example of a frustrated system.[1] Wannier solved the model and showed that it is disordered at all finite temperatures and that it has finite entropy at zero temperature.[1] Thus, the number of ground states varies as $e^{\alpha N}$, where N is the number of spins and α is a constant.

Recently there has been some interest in a three-dimensional model constructed by connecting infinitely many antiferromagnetic triangular Ising planes with ferromagnetic bonds in a third direction, yielding a three-dimensional hexagonal crystal.[2-4] This model may be relevant to the experimental system VI_2.[5] The Hamiltonian is

$$H=J\sum_{\langle i,j\rangle}^{xy} S_iS_j-J'\sum_{\langle i,j\rangle}^{z} S_iS_j\ , \tag{1.1}$$

where $J,J'>0$, $S_i=\pm 1$, and the summations are over nearest-neighbor pairs either in the x-y plane or along z. The number of ground states of this model varies as $e^{\beta N^{2/3}}$, where N is the number of spins and β is a constant, so the entropy at zero temperature is zero. Blankschtein *et al.*[2] have estimated the free-energy cost of a domain wall and found that it is finite. Therefore, it is not altogether shocking that Monte Carlo calculations indicate that a low-temperature phase with broken symmetry exists.[2] In fact, two phase transitions occur, so there are two different phases with broken symmetry.

Blankschtein *et al.*[2] have analyzed the phase diagram in terms of a Landau-Ginsburg expansion and Monte Carlo simulations. Although the Landau-Ginsburg theory results are consistent with their Monte Carlo data for the stacked nearest-neighbor antiferromagnet, we will show that models exist for which the Landau-Ginsburg analysis is inadequate at low temperatures. Therefore, one would like to find a method to determine directly the nature of the phases. In this paper we will concentrate on the low-temperature phase; the intermediate-temperature phase will be considered only briefly.

The usual method to elucidate phase diagrams near zero temperature is to expand the free energy using the low-temperature expansion. The stacked triangular Ising antiferromagnet does not obey the criteria described by Slawny[6] that ensure the existence of the low-temperature expansion constructed by counting excitations from the ground states of the system, and we will show that the low-temperature expansion for the model (if it exists) is quite pathological. In fact, we will demonstrate that the model has finite disorder even at arbitrarily low temperature. This result is compatible with the Monte Carlo work of Blankschtein *et al.*[2] but does not agree with extended mean-field theory calculations using the method of Nakanishi and Shiba.[7] We will also provide arguments that may indicate that the order parameter of the low-temperature phase proposed by Blankschtein *et al.*[2] never saturates, so that all the chains of the spins remain partially disordered even as the temperature is lowered towards zero. A by-product of this work is the construction of a class of simple one-dimensional Ising models with low-temperature expansions involving irrational coefficients, which implies that one cannot use simple counting rules to obtain the coefficients of the series. Using these models, one can obtain bounds to the free energy of the three-dimensional stacked antiferromagnet, but so far no bounds on the magnetization have been obtained.

The organization of the paper is as follows. Section II contains an outline of previous work; it describes the Landau-Ginsburg expansion, the Monte Carlo results, and the extended mean-field theory calculations. The importance of frustration is stressed, and we construct a simple model for which the Landau-Ginsburg expansion does not describe the low-temperature phase. In Sec. III the attempt to construct a low-temperature expansion for the model is described. This attempt yields insight into why a low-temperature phase may exist even though the two-dimensional Ising antiferromagnet has no long-range order. However, it is shown that attempts to construct a low-temperature expansion about an ordered ground state [similar to the methods employed for the three-

dimensional axial next-nearest-neighbor Ising (ANNNI) model[8] and the fcc Ising antiferromagnet[9,10]] yield inconsistent results, so such an expansion cannot exist. In Sec. III C we consider the low-temperature properties of several one-dimensional models that are used to provide bounds on the free energy, and also the proposed low-temperature phase of Blankschtein *et al.*[2] is investigated using this viewpoint. Our results are consistent with their proposed phase diagram, but we argue that the amplitude of the order parameter tends to a nontrivial value at very low temperatures. However, these arguments are not rigorous. Finally, Sec. IV contains conclusions, speculations, and suggestions for further work.

II. REVIEW OF PREVIOUS WORK

A. Landau-Ginsburg-Wilson analysis of the model

Blankschtein *et al.*[2,3] have performed a Landau-Ginsburg-Wilson (LGW) analysis of the model, which is reviewed here. The analysis is expected to be valid near the transition from the paramagnet to the intermediate-temperature phase, where the order parameter is small. On the other hand, it does not necessarily describe the low-temperature phase because frustration effects are neglected.

The method consists of expanding the free energy in a power series of polynomial invariants of the order parameter. Therefore, the first step is to find the order parameter. At this stage there are two usual ways to proceed—either one uses knowledge of the ground-state properties or one minimizes the Fourier-transformed coupling

$$J(\mathbf{q})=(1/\sqrt{N})\sum e^{i\mathbf{q}\cdot\mathbf{r}}J(\mathbf{r})$$

as a function of $\mathbf{q}$. For unfrustrated systems these two methods are equivalent. However, for the stacked Ising antiferromagnet the ground state is highly degenerate and the relevant order parameter is not obvious. However,

$$J(\mathbf{q})=J[\cos q_x+2\cos(q_x/2)\cos(\sqrt{3}q_y/2)]-J'\cos q_z$$

has two inequivalent minima at $\mathbf{Q}=\pm(4\pi/3)\hat{\mathbf{x}}$ (and other Q's related by reciprocal-lattice vectors). Thus, by using the second method, one expects an order parameter of the form $\psi=\psi_0\cos(\mathbf{Q}\cdot\mathbf{x}+\phi)$. However, the energy per spin of the system's ground states $E_{\text{g.s.}}=-J-J'$ is greater than the energy $E=-\frac{3}{2}J-J'$ that one obtains from the expression

$$H=\sum_q \psi_q\psi_{-q}J(q)\ , \tag{2.1}$$

where $\psi(\mathbf{Q})=\psi(-\mathbf{Q})=1/\sqrt{2}$ and $\psi(q)=0$ otherwise.[11] This discrepancy occurs because LGW description neglects the constraint on the spins $|S_j|=1$ which is responsible for the frustration. Blankschtein *et al.*[2] argue that the restraint on spin length is irrelevant under renormalization at criticality and ignore it thereafter, but we will return and examine this contention.

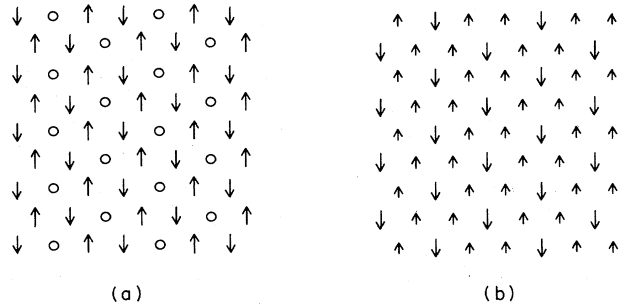
Given an order parameter $\psi(r)=\psi_0\cos(\mathbf{Q}\cdot\mathbf{r}+\phi)$, with $\mathbf{Q}=(4\pi/3)\hat{\mathbf{x}}$, one may construct the following invariant polynomial free energy up to sixth order in ψ_0:

$$F_{\text{LGW}}(\psi)=a_2|\psi_0|^2+a_4|\psi_0|^4+a_6|\psi_0|^6$$
$$+b_6|\psi_0|^6\cos(6\phi)\ . \tag{2.2}$$

If a net magnetic moment M is present, another term of the form $a_3M\psi_0^3\cos(3\phi)$ is also allowed. Depending on the sign of b_6, the minimum of F_{LGW} occurs at either $\phi=0$ or $\phi=\pi/6$. A mean-field analysis using the method of Bak and von Boehm[12] assuming $\psi(\mathbf{r})=\psi_0\cos(\mathbf{Q}\cdot\mathbf{r}+\phi)$ yields a positive value of a_6,[13] so it is plausible that $\phi=\pi/6$ and the intermediate phase has a three sublattice structure with one sublattice with no magnetic moment and the other two sublattices with equal and opposite moments [see Fig. 1(a)].

Because the Monte Carlo results indicate the existence of a second phase transition at lower temperature T_{cl}, Blankschtein *et al.*[2] argue that at T_{cl} the renormalized value of b_6 may change sign and cause a transition to a state with $\phi=0$ and order parameter $\psi=\psi_0\cos(\mathbf{Q}\cdot\mathbf{x})$. This order parameter corresponds to a three-sublattice state with magnetizations M, $-M/2$, $-M/2$ [Fig. 1(b)]. This conclusion is compatible with their Monte Carlo results, but since low-temperature Monte Carlo simulations are difficult, the very low-temperature portion of the phase diagram was not accessible.

We argue here that the LGW analysis, since it ignores frustration, may not be adequate to describe the low-temperature phases of the model. (Our interpretation of the lower phase transition is that the frustration becomes relevant at T_{cl}.) For instance, if one considers this model with weak second-nearest-neighbor attractive interactions in the plane (similar to a model considered by Mekata[14] in two dimensions) the LGW analysis yields identical results and the ground state has a three-sublattice structure, but each sublattice is fully ordered at low enough temperatures. One can also consider a model with, in addition to the antiferromagnetic nearest-neighbor coupling J', *third*-nearest-neighbor ferromagnetic coupling J_3 in the plane, where $J_3 \ll J'$ (and ferromagnetic coupling J_2 between planes). The LGW analysis is the same as above and yields the same low-temperature phase, but it is easily seen that the energy of the state shown in Fig. 2 is lower than the energy of any three-sublattice state, so at a



(a) (b)

FIG. 1. (a) Sublattice structure of the intermediate-temperature phase of the stacked triangular Ising antiferromagnet found by Blankschtein *et al.* (Ref. 2). (b) Sublattice structure of the low-temperature phase of the stacked triangular Ising antiferromagnet found in Ref. 2.

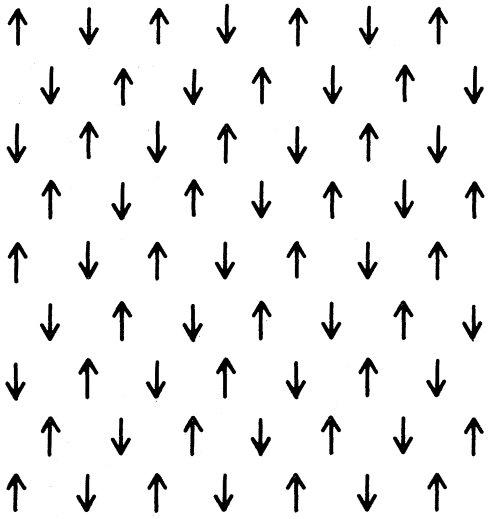


FIG. 2. Structure of the low-temperature phase of the stacked triangular Ising antiferromagnet with attractive third-nearest-neighbor couplings in the planes. This phase does not have a three-sublattice structure.

low enough temperature one expects the order parameter to be $\psi=\psi_0\cos(\mathbf{Q}_2\cdot\mathbf{x})$, where $\mathbf{Q}_2=(2\pi/\sqrt{3}a)\hat{\mathbf{y}}$ or $2\pi/\sqrt{3}a[\frac{1}{2}\hat{\mathbf{y}}\pm(\sqrt{3}/2)\hat{\mathbf{x}}]$. This phase does not have a three-sublattice structure.

This argument proves that models exist whose (finite number of) ground states cannot be found by minimizing $J(q)$. These models have well-defined low-temperature expansions for the free energy, so at low enough temperatures they have the same order parameters that describe their ordering in the ground state. This argument does not prove that the frustration becomes relevant at T_{cl} because, for example, one could imagine a third temperature T_{c3} at which the transition to the $\mathbf{Q}_2$ wave vector occurs, but the presence of two and only two transitions is a consistent and simple possibility.

B. Extended mean-field theory

Because the LGW theory has drawbacks, one is led to examine the low-temperature phase further using other techniques. For instance, one could use Mekata's mean-field (MF) theory,[14] where each sublattice i is assumed to have a uniform magnetization M_i when its affect on the other sublattice is calculated. For reasons that are discussed in the next section we assume that the Monte Carlo results indicating a three-sublattice structure in the low-temperature phase apply. The Hamiltonian is thus approximated by the form

$$H_{\rm MF}=\sum_{i=1}^{3}[3J(M_j+M_k)-2J'M_i]\times\sum_l S_i^l-3J(M_1M_2+M_1M_3+M_2M_3)+J'\sum_i M_i^2\ , \tag{2.3}$$

where i is a label for the sublattices and the sum over l is for the spins in each sublattice. Minimization of the free energy implies that the M_i satisfy the consistency condition

$$M_i=\tanh\{-\beta[3J(M_{i-1}+M_{i+1})-2J'M_i]\}\ , \tag{2.4}$$

where $1/\beta=k_BT$ and modular arithmetic is used for the sublattice labels.

We wish to find the solution to the three coupled equations (2.4) for the magnetizations in the limit of low temperatures. As $T\to0$ these equations have the solution $M_1=M_2=M_3=0$ as well as six equivalent solutions with broken symmetry, of which $M_1=1$, $M_2=M_3=-1$ is one. Thus, this mean-field theory predicts that the low-temperature phase has a net magnetic moment, in contrast to the LGW results.

However, this mean-field theory ignores fluctuations and frustration effects. One can visualize a possible problem by noticing that in this approximation the one-dimensional Ising model would have a nonzero moment in zero field. If the magnetization of sublattices 1 and 2 are exactly 1 and -1, respectively, then the third sublattice behaves as a one-dimensional Ising model in no field and thus has no moment at any finite temperature.

One can try to make less drastic approximations in the theory. Nakanishi and Shiba[7] have examined the stacked frustrated Ising model in a field using the extended mean-field theory method of Scalapino *et al.*[15] One treats each line of spins exactly, but uses an effective "mean field" to account for interactions between chains. (In this formulation, the chains are taken perpendicular to the triangular planes, i.e., along the z axis). We again assume the low-temperature phase has a three-sublattice structure and follow their method for the case with no applied field. The Hamiltonian (1) is broken into intrachain and interchain parts

$$H=H_z+H_{xy}\ , \tag{2.5}$$

where H_z is the Hamiltonian of a one-dimensional Ising model in no field.

The interchain part H_{xy} is approximated by

$$H_{xy}^{\rm MF}=\sum_i^z\sum_r^{xy}-h_{\rm MF}(\mathbf{r})S(i,\mathbf{r})+C\ , \tag{2.6}$$

where the "mean field" is assumed to be independent of i and

$$C=\sum_i\sum_r h_{\rm MF}\langle S(\mathbf{r})\rangle+\sum_i\sum_{\substack{r,r'\\ \rm NN}}J\langle S(r)\rangle\langle S(r')\rangle\ . \tag{2.7}$$

Minimization of the free energy yields the relation

$$h_{\rm MF}(\mathbf{r})=-\sum_{\substack{r'\\ \rm NN}}J\langle S(\mathbf{r}')\rangle\ . \tag{2.8}$$

(If two sublattices have exactly equal and opposite moments, the third sublattice has zero moment in this approximation.)

At sufficiently low temperatures one must solve the simultaneous equations

$$M_i=\frac{-\sinh[K(M_{i-1}+M_{i+1})]}{[y^2+\sinh^2K(M_{i-1}+M_{i+1})]^{1/2}} \tag{2.9}$$

with $y=e^{-2\beta J'}$ and $K=3\beta J$.

Note that if $M_1+M_2=M_2+M_3=M_1+M_3=0$, then $M_1=M_2=M_3=0$. Therefore, for a state with broken symmetry, at least one sublattice i must have $M_i=\pm 1$ up to corrections which can be shown to be very small. For definiteness, we shall let $M_1=1$. One must then solve two simultaneous equations for M_2 and M_3. One solution is $M_2=-1$, $M_3=0$. The "mean-field" free energy of this configuration is

$$\frac{F_{\rm MF}}{k_BT}=-\beta(J'+2J)-\tfrac{1}{3}e^{-2\beta J'} . \tag{2.10}$$

The other solution is

$$M_2=M_3=-1+\left[\frac{1}{2K}e^{-4\beta J'}\right]^{1/3}+O(e^{-8\beta J'/3}) . \tag{2.11}$$

The free energy of this configuration is lower, so as the temperature tends to zero the magnitude of the magnetization of all three sublattices tends to one, and the system has a moment, as in the simple MF theory but unlike the LGW theory results.

This review of previous work reveals that the LGW and MF analyses of the stacked triangular antiferromagnet are incompatible. Obvious questions arise: (1) Why does the low-temperature phase have a three-sublattice structure? (2) Does the low-temperature phase have a net magnetic moment? We will show the answer to (1) is revealed in entropy considerations, but the attempts to answer (2) will not be successful. We will obtain bounds on the free energy of the system that enable us to eliminate some candidates for the low-temperature phase and thus prove that the mean-field theory results cannot be correct. The results will be compatible with the LGW predictions, but we will argue that the magnitude of the order parameter may saturate at a nontrivial value. Thus, this paper will show that neither LGW theory nor MFT (extended or otherwise) gives totally reliable information about this frustrated system at very low temperatures.

III. THE LOW-TEMPERATURE EXPANSION

A. Naive approach

Since we are primarily concerned with low-temperature properties of the model, it is reasonable to attempt to construct a low-temperature expansion. Usually, one does so by simply counting the number of low-energy excitations of the system about the ground state. For instance, for an Ising ferromagnet on a square lattice in two dimensions with nearest-neighbor couplings J, the lowest-energy excitations are single spin flips with energy $8J$ which can occur at any site on the lattice, so the first term in the low-temperature expansion is $-(1/N)(Ne^{-8J/k_BT})$. The second term is found by counting the number of ways to place two adjacent flipped spins on the lattice, etc. Fisher and Selke[8] and Mackenzie and Young[9] have considered models with infinitely degenerate ground states; they find that one must perform the expansion about each ground state and find the one with the most low-energy excitations, which is then selected out in the low-temperature phase. Because of the nature of the method we consider only excitations about ground-state configurations. (This drawback will eventually lead to failure.)

B. Attempts to construct a low-temperature expansion

The method yields insight into why a low-temperature phase with broken symmetry could occur, even though the two-dimensional model is disordered at all temperatures. The system selects out one of its infinite number of ground states to form the low-temperature phase because of entropy considerations.

Slawny[6] has considered the conditions necessary for a low-temperature expansion to be valid, one of which is not satisfied by the stacked triangular Ising antiferromagnet. This condition is that the energy of any finite region of flipped spins must tend to infinity as the number of flipped spins does. To see how this condition is violated, consider the ground-state configuration shown in Fig. 3. The lines of spins corresponding to sublattices B and C are fully frustrated in the plane, since they have three satisfied and three unsatisfied bonds. Therefore, the only couplings keeping the chain magnetized are the couplings along z. One can note that the energy cost of flipping one spin $4J'$ is the same as the energy required to flip any number of adjacent spins along the line. The energy remains $4J'$ even as the number of flipped spins tends to infinity. This problem does not occur for any line of spins that is either partially or not frustrated in the plane.

This effect explains why the three-sublattice structure is preferred at finite temperatures. The argument above applies to any line of spins that is fully frustrated in the plane, and one can see that the three-sublattice structure

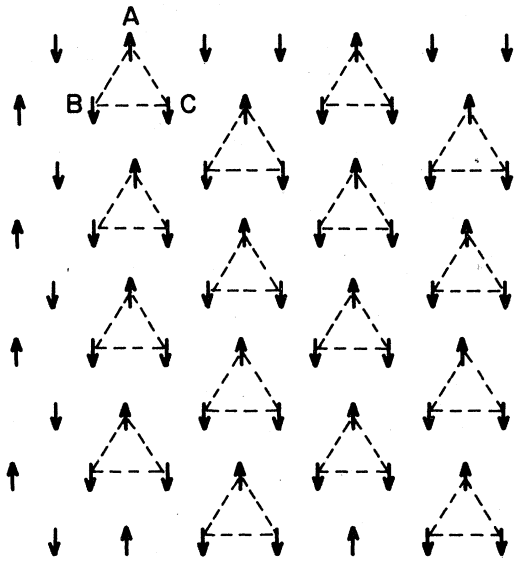


FIG. 3. Ground-state configuration that appears to be favored at low temperatures because of entropy considerations.

uniquely maximizes the number of frustrated lines of spins (up to a sixfold degeneracy), with $\frac{2}{3}$ of the spins frustrated in the plane. Although naively each frustrated line is equivalent to a one-dimensional Ising model in no field and thus should have no moment at finite temperature, if it demagnetizes, then an adjoining line of spins that formerly had large entropy because of frustration becomes unfrustrated and loses its entropy. Therefore, entropy tends to stabilize the $M=(1,1,-1)$ state, and the effect is much larger than in two dimensions because a line of spins is involved instead of one spin only.

At this point one may wonder at the reasonability of attempting to construct the low-temperature expansion. However, we point out that the one-dimensional Ising model with coupling $\tilde{J}$ in no field has exactly the same flaw as our model, but a low-temperature expansion can be constructed if one notices that the elementary excitations are kinks rather than spin flips. Thus, by counting possible locations for kinks, one finds the low-temperature expansion for the free energy

$$F=-k_BTN\left\{\frac{\tilde{J}}{k_BT}+e^{-2\tilde{J}/k_BT}-\tfrac{1}{2}e^{(-4\tilde{J}/k_BT)}+\cdots\right\}, \tag{3.1}$$

which can be verified by expanding the exact result. Using this point of view, one expects that starting from the $(1,1,-1)$ phase, the first term in the low-temperature expansion is just $-\frac{2}{3}e^{-2J'/k_BT}$, since $\frac{2}{3}$ of the spins are in "frustrated" chains.

We now attempt to construct more terms of the expansion for the stacked triangular Ising antiferromagnet and find an inconsistency, thus demonstrating that the method cannot be used. In addition, we show that the low-temperature phase is at least partially disordered. In the next section it will be argued (but not rigorously) that all the sublattices are partially disordered. It will not be shown that partial order does exist at low enough temperatures, so at this stage the exact nature of the low-temperature phase remains an open question.

We exhibit the problems by assuming a low-temperature expansion is possible. In this spirit, we expand about a ground state of the form $(1,-1,-1)$ and assume that all except for a finite number of sublattices are fully ordered. Thus, in lowest order we assume that all sublattices but one are ordered and count possible locations for kinks; thus one expects the lowest-order term to be $-\frac{2}{3}e^{-2J'/k_BT}$, as above.

The problems arise when one attempts to construct the second-order term. The calculation is done by assuming all sublattices but two are fully ordered and counting pairs of excitations; the only nontrivial case occurs when one considers two adjacent frustrated chains. Since all other chains are assumed to be ordered they cause an "effective" field on the two chains considered. The problem is equivalent to a one-dimensional two-chain Ising model in a field, shown in Fig. 4. There is an antiferromagnetic coupling J between chains and a ferromagnetic coupling J' along the chains, and the field is J. One finds that this model should have a net magnetization by arguing as outlined above for the full model. We note that the extended mean-field theory discussed in Sec. II B should describe this model if it is valid for the full problem. The two-chain Ising model is solved straightforwardly using the transfer-matrix method;[16] at low temperatures its free energy per spin is

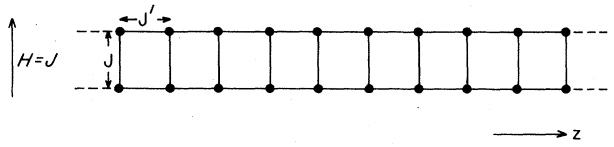


FIG. 4. Two-chain Ising model in a magnetic field that is equivalent to assuming all but two of the chains in the full model are ordered.

$$\frac{F}{k_BT}=-J'-\tfrac{1}{2}J-\frac{1}{\sqrt{2}}e^{-2J'/k_BT}+O(e^{-4J'/k_BT}), \tag{3.2}$$

so the *first-order* coefficient for two chains differs from that for one chain. Thus, there is no reason to expect the first-order coefficient of the full model to be given by the one-chain calculation. In addition, as the temperature is lowered towards zero, the magnetization per spin approaches $\frac{1}{2}$ rather than 1. (Since a field is present, finite magnetization is allowed even though the model is one dimensional.) Thus, the entropy effect that causes the magnetization to be nonzero does not cause the chains to order fully. These results do not depend on the ratio of the coupling constants J/J', as long as it is finite. One can rationalize this as follows—if the interchain coupling is weak, then the energy cost of creating a region with spins from both chains aligned opposite to the field is small, but it is still greater by a finite amount than the cost of flipping between ground states. Therefore, at low temperatures these regions are exponentially suppressed and they can be ignored. These exact results for the analytic form of the free energy and for the net magnetic moment disagree with the extended MF theory prediction, so that approximation must be regarded as unreliable.

It is now clear that the assumption of fully ordered sublattices in the stacked antiferromagnetic triangular Ising model is inconsistent. Since the chains for which excitations are allowed have only partial order at any finite temperature, and excitations are allowed on all chains, the low-temperature phase cannot consist of three fully ordered sublattices. Since the $(1,-1,-1)$ phase has lower free energy than any other fully ordered sublattice configuration, the low-temperature phase must have finite disorder. The low-temperature expansion cannot be performed by the method outlined above, because the disorder in each chain must be accounted for when calculating its effects on neighboring chains.

In summary, so far we have shown, by explicit construction of counterexamples, that none of the obvious methods—Landau-Ginsburg-Wilson analysis, mean-field theory, and naive low-temperature expansion—can describe adequately the low-temperature phase of a model with frustrated spins. We now describe some calculations done on bundles of chains that yield bounds on the free

energy and give some insight into the nature of the low-temperature phase.

C. Low-temperature calculations for small bundles of chains

This section describes finding the leading term in the low-temperature expansion for larger numbers of coupled chains. These calculations are used to obtain bounds on the free energy in the full model. Since any one-dimensional short-ranged model in zero field has no phase transition, knowledge of the magnetization is more difficult to obtain, but if one assumes that a fully ordered sublattice exists, bounds on the magnetization can be obtained. There are indications, however, that no sublattice of the full model is fully ordered.

We redefine the energy scale so the system has zero energy at zero temperature. The free energy per spin of the two-chain case has the form

$$F/k_BT=-(1/\sqrt{2})e^{-2J'/k_BT}+O(e^{-4J'/k_BT})$$

and the magnetization per spin is $\frac{1}{2}$. These results can be understood simply by noticing that as $T\to 0$ the system has three degenerate ground states labeled A, B, and C in Fig. 5(a). The system can change its configuration with an energy cost of $2J'$ by flipping one spin only. Therefore, to lowest order, transitions between A and B and between A and C are allowed, but transitions between B and C are not, as represented schematically in Fig. 5(b). The system breaks up into a series of domains. On average, the size of a given domain is independent of whether it is A, B, or C, so the probability P_α of finding each type of region can be found by writing an equation describing transitions between the various ground states. The regions

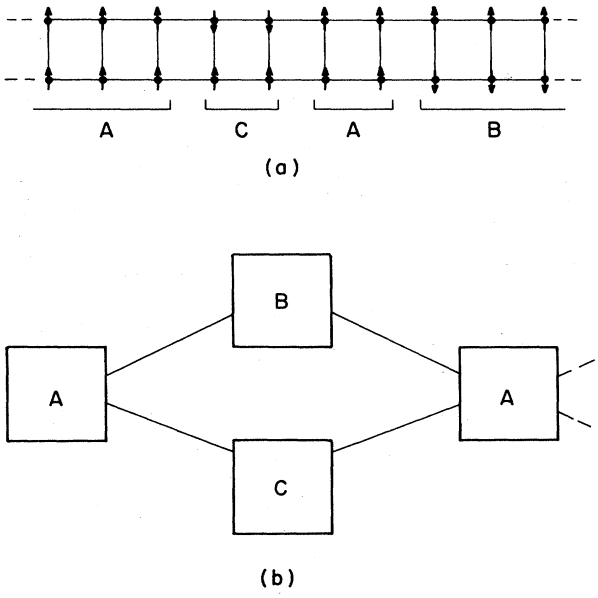


FIG. 5. (a) A "typical" configuration of the two-chain Ising model with walls separating the three ground states. (b) Schematic rendering of the lowest-energy transitions, which dominate at very low temperatures.

B and C can be connected only to regions of A while the A phase can be connected to either B or C, so

$$\begin{aligned} P_A&=P_B+P_C\ ,\\ P_B&=\tfrac{1}{2}P_A\ ,\\ P_C&=\tfrac{1}{2}P_A\ . \end{aligned} \tag{3.3}$$

Since $P_A+P_B+P_C=1$, $P_A=\frac{1}{2}$ and $P_B=P_C=\frac{1}{4}$. The magnetization per spin is $1(\frac{1}{2})+0(\frac{1}{4})+0(\frac{1}{4})=\frac{1}{2}$, the exact result found above. To calculate the first term in the low-temperature expansion of the free energy, notice that given n domain walls on a chain with N spins (i.e., $N/2$ sites long), the walls can be placed in $(N/2)!/[n!(N/2-n)]!$ different ways with an energy cost of $2nJ'$. Half the walls can also choose whether to go from A to B or A to C, while the other half have no freedom, since a given wall starting in a region of B or C must end in a region of A. Thus the partition function can be written as sum over the number of walls n:

$$\begin{aligned} Z&=\sum_{n=0}^{N/2}\frac{(N/2)!}{n!(N/2-n)!}(2^{n/2})e^{-2nJ'/k_BT}\\ &=(1+\sqrt{2}e^{-2J'/k_BT})^{N/2}\ . \end{aligned} \tag{3.4}$$

Thus, the free energy per spin is

$$F/k_BT=-\ln Z/N\simeq-(1/\sqrt{2})e^{-2J'/k_BT}\ .$$

It is possible to extend this method to deal with larger bundles of chains. For instance, Fig. 6 shows three chains coupled ferromagnetically with coupling J' along their length and antiferromagnetically with coupling J_1 between chains in an external field H which is $2J_1$ for the middle spin and J_1 for the end spins. (For simplicity we assume $J_1\gg J'$, but for the reasons explained above we expect the results to be independent of J_1/J'.) The ground states can be divided into classes with 0, 1, and 2 down spins, respectively, and the possible spin flips of a state with 1 spin down depends on whether it is in the middle or on the end (nearest-neighbor down spins are not allowed at low temperatures). The various states are shown in Fig. 7, and Fig. 8 is a diagram of possible transitions between them. Again, assuming that the probabili-

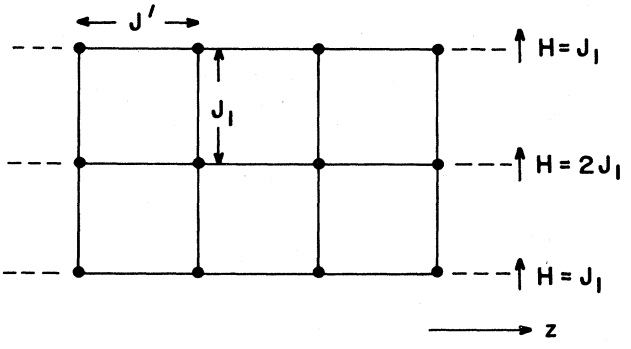


FIG. 6. A three-chain model whose low-temperature magnetization and free energy are found in the text.

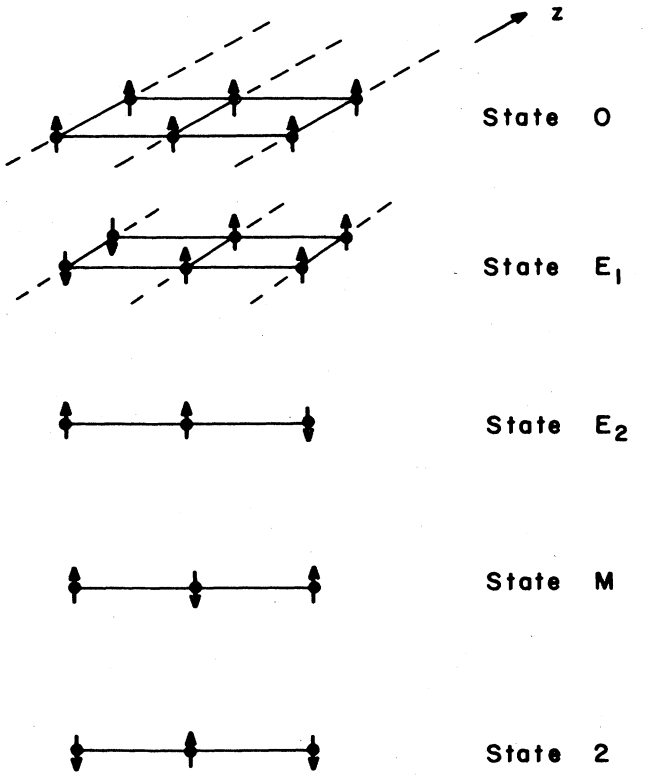


FIG. 7. Degenerate ground states of the three-chain model.

ties of all single spin flips are the same, one can write coupled equations for the probabilities of each type of domain:

$$\begin{aligned} P_0 &= \tfrac{1}{2}P_{E_1}+\tfrac{1}{2}P_{E_2}+P_M \ , \\ P_{E_1} &= P_{E_2} = \tfrac{1}{3}P_0+\tfrac{1}{2}P_2 \ , \\ P_M &= \tfrac{1}{3}P_0 \ , \\ P_2 &= \tfrac{1}{2}P_{E_1}+\tfrac{1}{2}P_{E_2} \ . \end{aligned} \tag{3.5}$$

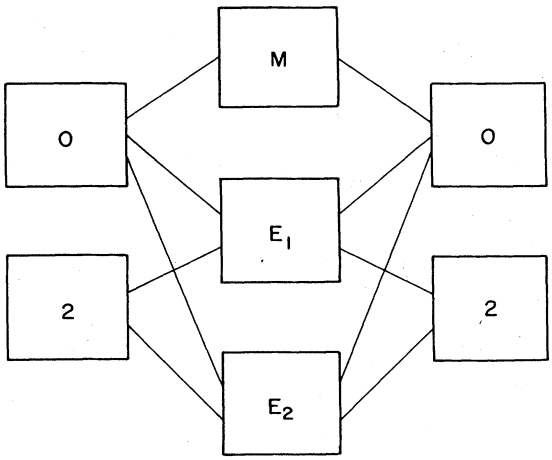


FIG. 8. Allowed transitions (to lowest order) between degenerate ground states of the three-chain model.

These equations, together with the condition that $\sum_\alpha P_\alpha = 1$, imply $P_0 = \frac{3}{10}$, $P_{E_1} = P_{E_2} = \frac{1}{5}$, $P_M = \frac{1}{10}$, and $P_2 = \frac{1}{5}$. The mean magnetization is $\frac{1}{5}$ for the end chains and $\frac{4}{5}$ for the middle chain.

The entropy calculation for this larger system is slightly more involved. Again we use the fact that n walls in a chain with N spins cost energy $2J'n$ and can be placed in $(N/3)!/[(N/3-n)!n!]$ locations. However, the number of choices at each wall location depends on the domain type.

Suppose the system is in state 0 (all spins up) at the point $x=0$. Then in the region just past the first wall, it can be in any of three states. After the second wall, each of the states can "flip" into either one of two states (from E_1 or E_2) or one state (from M), depending on the first wall.

This process goes on indefinitely. One can make a "branching diagram" of all possible allowed orderings similar to Fig. 9. To calculate the entropy, one must find the asymptotic dependence of the total number of distinct orderings of N domains $\alpha(N)$ on N. In this calculation, different states related by symmetry are distinguishable (i.e., there are three states with one spin down, etc.). Consider the chain after N flips and say the last domain has an even number of down spins. After two more walls there again is a region with an even number of down spins, so one can write the number of distinct orderings of $N+2$ domains $\alpha(N+2)$ in terms of $\alpha(N)$.

Say at the Nth step there are $\gamma_0(N)$ chains ending with a domain of type 0 and $\gamma_2(N)$ chains ending with a domain of type 2. A type-0 domain can flip into either a type-M domain or one of two type-E domains. A type-2 domain can flip into one of the two type-E domains. Similarly, a type-E domain can flip into either a type-0 or type-2, and a type-M can flip only into a type-0 domain. Thus,

$$\begin{bmatrix} \gamma_0(N+2) \\ \gamma_2(N+2) \end{bmatrix} = A \begin{bmatrix} \gamma_0(N) \\ \gamma_2(N) \end{bmatrix} \tag{3.6}$$

with

$$A = \begin{bmatrix} 111 \\ 110 \end{bmatrix} \begin{bmatrix} 11 \\ 11 \\ 10 \end{bmatrix} = \begin{bmatrix} 32 \\ 22 \end{bmatrix} .$$

By iteration, one finds

$$\begin{bmatrix} \gamma_0(2M) \\ \gamma_2(2M) \end{bmatrix} = A^M \begin{bmatrix} \gamma_0(0) \\ \gamma_2(0) \end{bmatrix} . \tag{3.7}$$

The asymptotic dependence of $\alpha(2M)=\gamma_0(2M)+\gamma_2(2M)$ on M for large M is determined by the largest eigenvalue λ_1 of the matrix A. One finds $\lambda_1 = \frac{1}{2}(5+\sqrt{17})$, so the first term of the low-temperature expansion of the free energy per spin is

$$-k_B T\{\tfrac{1}{3}[\tfrac{1}{2}(5+\sqrt{17})]^{1/2}\}e^{-2J'/k_B T} .$$

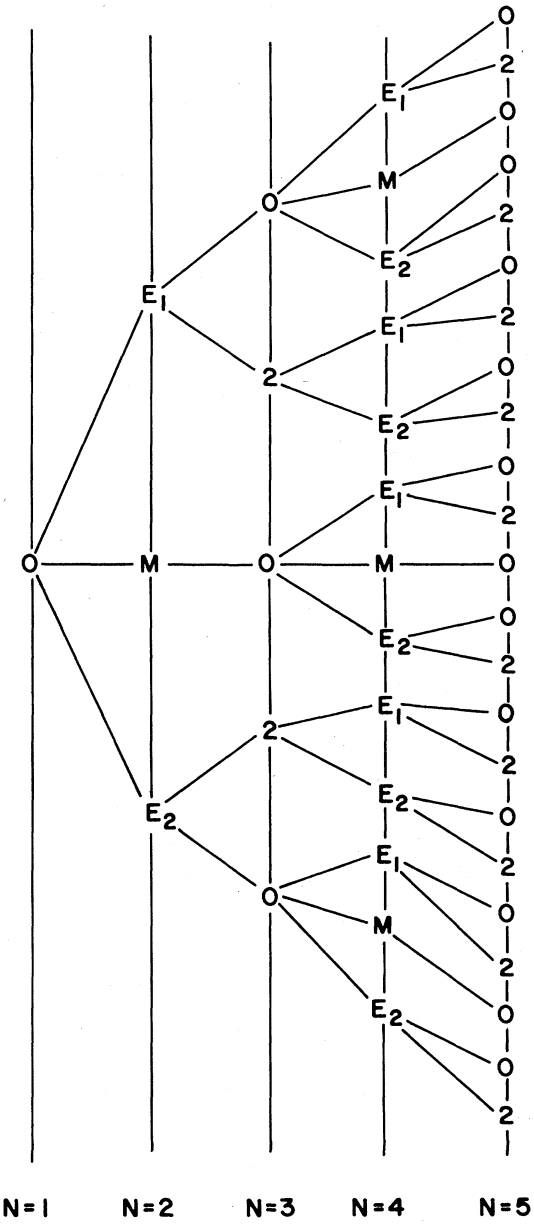


FIG. 9. "Branching" diagram showing all possible orderings of the domains of the three-chain model whose first domain has all spins up. By writing a recursive relation describing this tree, the lowest-order term in the low-temperature expansion of the free energy can be calculated.

This result can be verified by direct calculation using the transfer matrix.

This method can be applied to any finite bundle of chains. For instance, one can solve a ring of six chains in

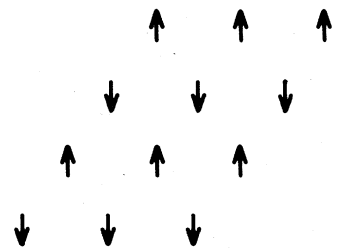


FIG. 10. Configuration of 12 spins that cannot be reached via low-energy excitations of a $(1,-1,-1)$ state. This configuration does not contribute to the free energy.

an external field $H=2J'$; one finds a mean magnetization of $\frac{1}{2}$ and a low-temperature free energy of

$$F/k_BT=-\tfrac{1}{6}[\tfrac{1}{2}(15+\sqrt{129})]^{1/2}e^{-2J'/k_BT}.$$

One can also solve models with 3, 6, 9, or 12 chains in no field with periodic boundary conditions imposed. For the larger bundles there are states which cannot be reached by single spin flips between low-energy configurations from the $(1,-1,-1)$ sublattice structure (i.e., the state pictured in Fig. 10), but they do not contribe to the first-order coefficient. The magnetization is zero, and the free energy per spin F_n for a model with n spins is

$$\frac{F_n}{Nk_BT}=-a_ne^{-2J'/k_BT}, \tag{3.8}$$

with $a_3=\frac{2}{3}$, $a_6=\sqrt{2}/3\simeq 0.47$, $a_9=\frac{1}{9}(8+2\sqrt{7})^{1/2}\simeq 0.405$, and $a_{12}=\sqrt{22}/12\simeq 0.391$. Since the a_n decrease as n increases, one may speculate that they are upper bounds to the value for the infinite system.

The method used above to calculate partition functions assumes that only one spin flip occurs for each step along the chains. This assumption is valid for any finite number of chains at low enough temperature, but as the number of chains goes to infinity, multiple spin flips become important. This complication can be accounted for straightforwardly.

Assume that every spin flip costs an energy $2J'$ and that it has (on average) α choices for its location in the plane. As the cross section A is increased, one expects α to increase proportionally, so one can write plausibly $\alpha=\gamma A$, where γ is a constant. (We will show $\frac{1}{3}<\gamma<\frac{2}{3}$, if it is well defined.) For each of the N_z steps along the chains, any number of spin flips can occur, each at an energy cost of $2J'$, so the partition function can be written

$$Z=\sum_{n_1=0}^{\infty}\sum_{n_2=0}^{\infty}\cdots\sum_{n_{N_Z}=0}^{\infty}\frac{1}{n_1!}\frac{1}{n_2!}\cdots\frac{1}{n_{N_Z}!}\exp\left[(-2J'/k_BT)\sum_{i=1}^{N_Z}n_i\right]\prod_i\left[\frac{\alpha!}{(\alpha-n_i)!}\right]$$

$$\cong\left[\sum_{n=0}^{\infty}\frac{1}{n!}(\alpha e^{-2J'/k_BT})\right]^{N_Z}=(e^{\alpha e^{-2J'/k_BT}})^{N_Z}=(e^{\gamma e^{-2J'/k_BT}})^{AN_Z}. \tag{3.9}$$

Thus, the first term in the low-temperature expansion for the free energy per spin is

$$F=-\frac{k_BT}{AN_Z}\ln Z\cong -k_BT\gamma e^{-2J'/k_BT}.$$

Therefore, if one knows γ, the thermodynamic limit can be taken straightforwardly.

A simple lower bound for γ (i.e., upper bound to the free energy) is found by noting that if all the spins in one frustrated sublattice are held fixed, the other frustrated sublattice decomposes into $A/3$ decoupled Ising chains. Since only a subset of the configurations are counted, it follows that $\gamma\geq\frac{1}{3}$. A more restrictive bound can be found by considering units of nine chains with five of them constrained as shown in Fig. 11. The constraints are chosen so that the units can fluctuate independently, and using the method described above, one finds that $\gamma\geq[(11+\sqrt{57})/2]^{1/2}/9$. Clearly, this type of calculation can be extended to larger units and more stringent bounds obtained.

Lower bounds on the free energy (i.e., upper bounds on γ) can also be obtained by considering groups of chains with free boundary conditions. For instance, one knows that not all the chains are simultaneously fully frustrated, so γ is bounded above by 1, the value achieved by A independent Ising chains. One can also consider larger groups of spins; the simplest calculation involves three coupled chains with free boundary conditions in the plane—since constraints between the groups of chains are ignored, one overcounts configurations and gets the bound $\gamma\leq\frac{2}{3}$. Once again, larger groups of chains can be considered to obtain more stringent bounds.

One can argue that all three sublattices must exhibit finite disorder even at arbitrarily low temperature, so that every sublattice magnetization M_i tends to a value different from one as the temperature approaches zero. First assume that one completely ordered sublattice exists. Consider the set of chains shown in Fig. 12. If the spins labeled A_1 through A_6 are fixed to be up, then the configuration with B_1 through B_3 up and C_1 through C_3 down is expected to occur with finite weight. If so, then the interior spin X is free to flip. Naively, one could estimate

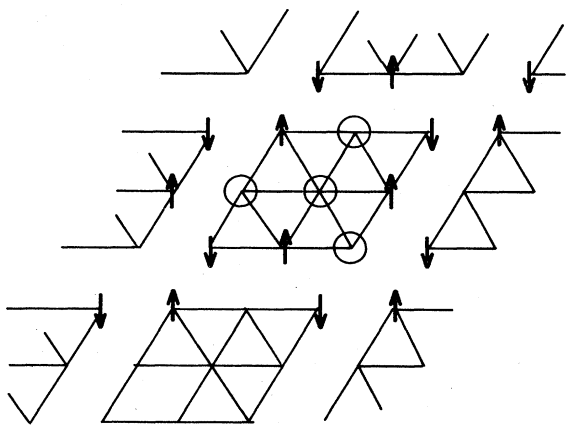


FIG. 11. Configuration of four chains that can be used to yield an upper bound on the free energy of the full model.

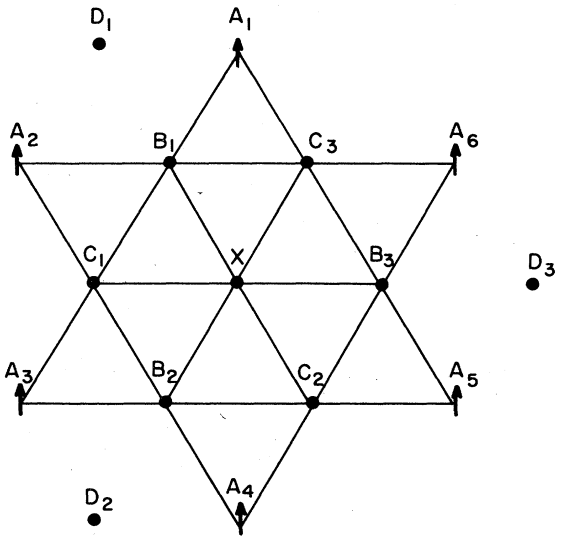


FIG. 12. Construction used to argue that if sublattice A consists of up spins only, then spin x should be free to flip a finite fraction of the time. This argument indicates that no sublattice is fully ordered even at arbitrarily low temperatures.

this effect by assuming a $(1,-\frac{1}{2},-\frac{1}{2})$ phase. One spin in each frustrated sublattice is flipped half the time, and the "right" spin will flip about half the time on each of three sublattices, so the central spin is free to flip about $(\frac{1}{4})^3=\frac{1}{64}$ of the time. Since spins B_1, B_2, and B_3 are also free to flip, the central spin will be down $(\frac{1}{4})(\frac{1}{64})=\frac{1}{256}$ of the time. This argument ignores correlations between the sublattices (i.e., the spins D_1, D_2, and D_3 must be down for B_1, B_2, and B_3 to be up), but there is no obvious reason why the configuration shown in Fig. 12 should not occur with finite weight. Alternatively, one can look at the set of seven spins shown in Fig. 13 and consider all

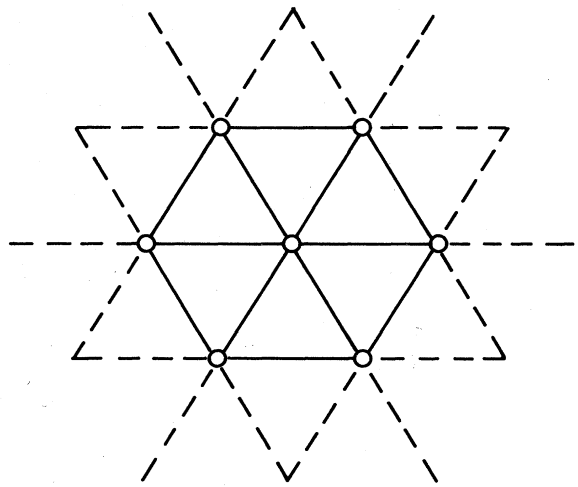


FIG. 13. Set of seven chains that can be examined with different boundary conditions, none of which are consistent with a $(1,-\frac{1}{2},-\frac{1}{2})$ state. This argument again indicates all three sublattices have finite disorder at any finite temperature.

possible states for a given configuration of the spins bounding this unit. One finds that no combination of boundary conditions is compatible with one fully ordered sublattice and two sublattices with magnetization $-\frac{1}{2}$. Thus we expect sublattice magnetizations of $(M_1,-M_2,-M_3)$, where M_1,M_2,M_3 approach nontrivial limits (i.e., not 0 or 1) as $T\rightarrow 0$.

The method of calculating the free energy of the bundles of chains may seem reminiscent of the time development of a finite number of spins. Thus, one may wonder if methods relating kinetic Ising models[17] in d dimensions to static models in $d+1$ dimensions are applicable,[18,19] especially since this correspondence tends to occur at Lifshitz points.[18] However, any connection is quite subtle. To see the complications, consider the simple two-chain model in a field of Sec. III A, whose mean magnetization is $\frac{1}{2}$ and free energy per spin is

$$F/k_BT=-(1/\sqrt{2})e^{-2J'/k_BT}+\cdots .$$

These results reflect the transitions between the three degenerate types of domains shown in Fig. 5(a). The domains do not occur with equal probability, but rather the A domain with both spins up is twice as likely as the B and C domains which have one spin up and one spin down. For any zero-dimensional model with three states of equal energy, each state occurs with equal probability in equilibrium. Any dynamical model which obeys detailed balance relaxes to the static equilibrium with equal weights for the three states and thus cannot describe the statistical mechanics of the one-dimensional coupled chains.

IV. DISCUSSION AND QUESTIONS

We have shown that Landau theory, mean-field theory, and standard low-temperature expansions cannot adequately describe the low-temperature phase of the stacked triangular Ising antiferromagnet. By solving one-dimensional models, one can obtain bounds on the lowest-order term of the low-temperature expansion of the free energy. One also can argue that all sublattices must have finite disorder at any nonzero temperature. These one-dimensional models have irrational coefficients in their low-temperature expansions, so these series cannot be obtained by the usual method of counting excitations about an ordered state.

However, many unanswered questions about the model remain. How does one prove that long-range order occurs at very low temperature (assuming that the Monte Carlo results indicating an ordered phase are correct)? Is the low-temperature phase ferrimagnetic or antiferromagnetic? These questions involve gross qualitative features of the low-temperature phase, and the fact that they are unanswered indicates the extent of our ignorance. In addition, more quantitative questions remain to be addressed. Is the parameter γ in Eq. (3.9) well defined, and if so, what is its value? What are the sublattice magnetizations at very low temperatures?

More work should be done to test the hypothesis that the low-temperature transition occurs when frustration becomes relevant. One test consists of doing a simulation on a stacked triangular Ising model with third-nearest-neighbor attraction and characterizing its phase diagram, as discussed in Sec. II A. Since we know that at very low temperatures the three-sublattice structure is unstable because of frustration effects, if only two transitions occur, it might indicate that the lower-temperature transition is caused by frustration becoming relevant. However, if three transitions occur (i.e., three ordered phases), then one may argue that the low-temperature phase in the absence of third-nearest-neighbor interactions corresponds to the intermediate ordered phase in the presence of these added couplings. This result would indicate that these frustrated models exhibit even more complexity in their phase structure than has been seen to date.

One would also like to test the claim that all three sublattices have finite disorder at any nonzero temperature. A Monte Carlo calculation of $\sum_i M_i^2$, the Edwards-Anderson order parameter, would be interesting, but it is very difficult to reach equilibrium at low temperatures, which is the region of interest here.

Finally, one may wonder whether the considerations discussed here are generally applicable. The major conclusion is that the low-temperature phase structures of frustrated systems can be very subtle, and that results obtained using many standard approaches must be regarded with caution. One concept used here that may be useful for future work on other systems is that of looking at the number of fully frustrated sites in a given configuration. This process is more difficult than counting frustrated rings,[20] because it depends on the particular spin configuration, but it yields insight into which configurations have the largest number of low-energy excitations and are thus preferred for entropic reasons. However, the details of the analysis are only expected to be applicable to other special nonrandom models with large degeneracy.

After completion of a draft of the manuscript, we learned of work by Dubois-Violette *et al.*,[21] who did a finite-size-scaling analysis of the low-temperature phase of this model. Their results are compatible with those of Blankschtein *et al.* and this work. In addition, we learned of Monte Carlo work by Grest[22] on a frustrated "comb" model on a cubic lattice, which may be amenable to the type of analysis presented here. However, investigating this question is beyond the scope of this paper.

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