

Kinetics of the order-disorder transition of the two-dimensional anisotropic next-nearest-neighbor Ising model

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(Received 26 July 1984)

The development of a modulated (2,2) antiphase ordering in the two-dimensional anisotropic next-nearest-neighbor Ising model is studied following a quench from the disordered phase to a low-temperature unstable state. The domain growth and the dynamical structure factor are found to evolve anisotropically. The structure factor is shown to satisfy scaling for a variety of choices of a length scale. The growth laws for different definitions of length scale are shown to be most consistent with an effective power-law exponent of $n \approx 0.50$. The dynamical roles of different types of domain walls and vertices present in this model are discussed, with an emphasis placed on the need for theoretical studies of this model.

I. INTRODUCTION

The kinetics of domain growth in systems which are quenched from a high-temperature disordered phase to a temperature below the critical temperature of an order-disorder transition has recently received considerable theoretical and experimental attention.¹⁻¹⁴ These nonlinear phenomena far from equilibrium are fundamental problems in statistical mechanics and are of practical interest in a variety of fields including metallurgy and surface science. In particular, models of physisorbed and chemisorbed systems are currently receiving considerable attention.^{6,14} This is warranted because these systems can be fairly realistically modeled by simple lattice-gas Hamiltonians.¹⁵

The quantities which are typically analyzed in these thermal-quench studies are the growth law for the average size $R(t)$ of the ordered domains, and the nonequilibrium structure factor $S(\vec{k}, t)$ where $\vec{k}$ is the wave vector and t is time. Both $R(t)$ and $S(\vec{k}, t)$ can be experimentally studied by electron microscopy and low-energy electron diffraction, respectively. For the nonconserved Ising model, with a doubly degenerate ground state, it is now known theoretically² and experimentally^{3,4,14} that

$$R(t) \sim t^n, \quad (1)$$

with $n = \frac{1}{2}$ and that the structure factor satisfies time-dependent scaling,

$$S(k, t) = R^d(t) F(kR(t)), \quad t \geq t_0 \quad (2)$$

where d is the dimensionality, F is the scaling function, and t_0 indicates the initial time period after which the scaling is valid.

Recently these properties have been tested in systems with a variety of features,⁵⁻¹² in order to find the unifying principles which may be operative for the growth process-

es. It is thought that there may exist universality classes in analogy to those known from critical phenomena, which would be partially determined by the degeneracy of the ground state⁵⁻¹² of the system. It has also been suggested⁷⁻⁹ that the domain wall "softness" may play a role in determining the universality class. This issue remains somewhat unsettled, however.¹⁰ In a recent Monte Carlo study by Sadiq and Binder⁶ the domain growth in a commensurate modulated structure was addressed. In a very careful study of the role of the conserved density combined with the ground-state degeneracy $p \geq 3$ they found a growth law with an exponent $n = \frac{1}{3}$. If true, this implies a new universality class. (Note that although the density is conserved in this model, the order parameter is not conserved.) For the case with a degeneracy $p = 2$ the conservation of the density does not play any role.^{4,14} The simple lattice-gas model studied by Sadiq and Binder shows a richness of different types of domain walls which can play a role in domain growth. Another class of modulated structures in incommensurate systems has recently been studied theoretically by Kawasaki.¹⁶ Such incommensurate structures appear for example in the N -state clock model [$N \geq 5$ (Ref. 17)] and the chiral clock model.¹⁸

In this paper we have chosen to study a two-dimensional anisotropic next-nearest-neighbor Ising (ANNNI) model,^{19,20} which is described by the following spin Hamiltonian:

$$\mathcal{H} = - \sum_{\langle ij \rangle} (J_1 s_{ij} s_{i+1j} - J_2 s_{ij} s_{i+2j} + J_0 s_{ij} s_{i,j+1}), \quad (3)$$

where $J_0, J_1, J_2 > 0$ and the sum is over all sites of a square lattice, each site being occupied by an Ising spin with $s_{i,j} = \pm 1$. This spin Hamiltonian can be simply transformed to a lattice-gas Hamiltonian. To our knowledge there is no physisorbed or chemisorbed system which is described by such a lattice-gas Hamiltonian. Nevertheless, this Hamiltonian offers yet another (very

simple) example of modulated commensurate and incommensurate structures. In addition it is perhaps the simplest model in which to study the effect of anisotropy on domain growth.

Figure 1(a) shows the phase diagram of the ANNNI model, which demonstrates the three different ordered structures possible in this model: a commensurate ferromagnetic phase, a modulated (2,2) antiphase, and an incommensurate phase. The boundary between these phases occur at a value of the parameter $\alpha = J_2/J_0 = \frac{1}{3}$ ($K = J_2/J_1 = \frac{1}{2}$). We have quenched to the (2,2) antiphase region of the ANNNI phase diagram. The modulated (2,2) antiphase has a ground-state degeneracy $p = 4$ and consists of an alternating sequence of two ferromag-

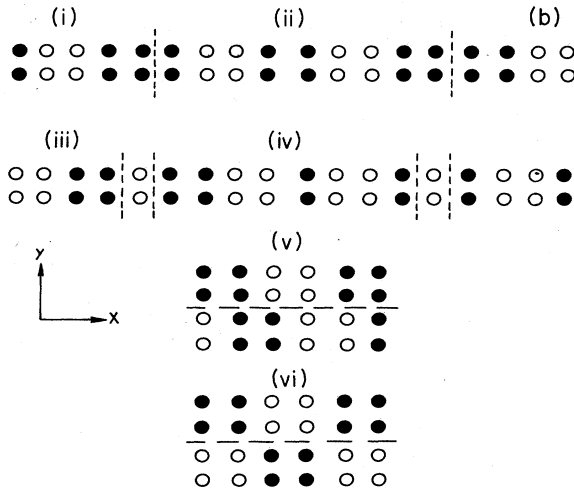
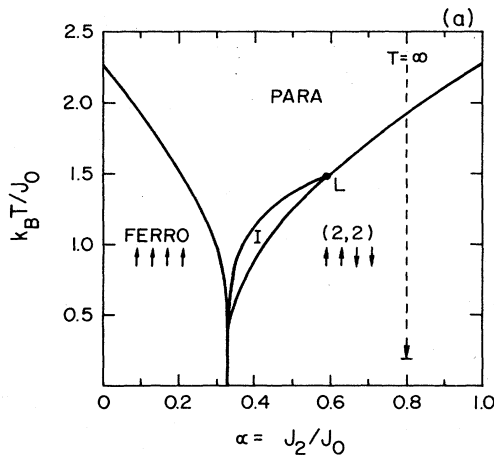


FIG. 1. (a) Schematic phase diagram of $d=2$ ANNNI model based on the free-fermion approximation by Villain and Bak (Ref. 19). Ferro, para, (2,2), and I refer to the ferromagnetic, paramagnetic, antiphase, and incommensurate regions of the phase diagram, respectively. The location and temperature of the quench is indicated. (b) Some of the different types of domain walls using lattice-gas terminology; (i) heavy-light wall, (ii) superheavy-superlight wall, (iii) soft superheavy-soft superlight wall, (iv) soft heavy-soft light wall; (v) and (vi) are the only two possible antiphase boundaries in the y direction. Open and solid circles correspond to up and down spins, respectively.

netic layers of up and down spins in the x direction. This is described by a two-component order parameter²⁰

$$\psi_\alpha = \frac{1}{(NM)^{1/2}} \left\langle \sum_{n,m} s_{nm} e^{i(\vec{Q}_\alpha \cdot \vec{r}_{nm})} \right\rangle, \quad \alpha = 1, 2 \quad (4)$$

where $\vec{Q}_1 = (2\pi/a)(\frac{1}{4}, 0)$, $\vec{Q}_2 = (2\pi/a)(-\frac{1}{4}, 0)$, and M and N are the number of sites in the x and y directions, respectively. After a deep quench to the (2,2) antiphase region we expect to see the structure factor

$$S_\alpha(\vec{k}, t) = \left\langle \left| \sum_{n,m} s_{nm} e^{i(\vec{Q}_\alpha + \vec{k}) \cdot \vec{r}_{nm}} \right|^2 \right\rangle, \quad \alpha = 1, 2 \quad (5)$$

evolving at these Bragg positions ($\vec{Q}_\alpha$ ($\vec{k}$ is the deviation from the Bragg position)). In this paper we study $S_1(\vec{k}, t)$ and drop the subscript hereafter.²⁸ Due to the anisotropy of the Hamiltonian, Eq. (3), we would expect to see an anisotropic structure factor. Thus the growth rates in the x and y directions should differ. Indeed we find that the growth laws in the x and y directions have the same time dependence but with different amplitude factors. The growth laws are found to be most consistent with an effective power-law behavior given by the exponent $n \approx 0.50$. Further theoretical studies of the growth laws for this system seem necessary, however, in view of the complexity of the domain walls shown in Fig. 1(b). The role of these domain walls will be discussed in qualitative terms later. We also propose a generalized dynamical scaling form which takes into account the anisotropy of the structure factor.

II. RESULTS

We have studied the kinetics of domain growth of the ANNNI model^{19,20} by preparing a system of $M \times N$ spins in an unstable state by instantaneous quenching from an infinite temperature to the low temperature $k_B T/J_0 = 0.2$. With the choice of anisotropy parameters, $\alpha = 0.8$ ($K = 4$), this is a deep quench within the (2,2) antiphase region [Fig. 1(a)]. The dynamics of the system is chosen to be a simple "Glauber spin-flip" process, which for an adsorbate system would correspond to random evaporation and condensation events of adatoms on a surface. We have considered a square lattice with periodic boundary conditions primarily of size 120×60 . Test runs on a larger (200×100) lattice yielded the same results as were obtained on the smaller lattice. This we interpret as indicating that the system size is large enough to encompass the physics of the phenomena of interest. Other more subtle finite-size effects are possible,^{4,6} however.

A standard requirement of Monte Carlo simulation is to obtain good statistics. In nonequilibrium phenomena this becomes even more important, because time averages are not ensemble averages. It should also be mentioned that a larger system cannot be "broken up" into many smaller independent systems to effectively increase the number of quenches. This is due to a long-range instability at $t=0$ which connects all regions of the system.¹³ Our results involve 120 and 300 runs for the structure-factor and perimeter-length studies, respectively. We believe this provides reasonable statistics for this model.

The evolution of the system after the quench is rapid, as is demonstrated by a typical sequence of diagrams in Fig. 2. The time region of our study is limited to ≤ 200 Monte Carlo steps (MCS) per spin, i.e., to the regime before percolation effects become important. The domain growth proceeds anisotropically. This is to be expected since the Hamiltonian is anisotropic. It should be mentioned that in Fig. 2 we have not made any distinction between the various domain walls, shown in Fig. 1(b). Later in this section we will discuss their role in determining the rate of growth of domains. We also find three- and four-rayed vertices occurring in the morphological structure, due to the fourfold degeneracy of the ground state. These do not appear to play a significant role in the growth.

The anisotropy of domain growth is apparent in the shape of the structure factor. This can be made quantitative by determining a generalized second moment in the (k_x, k_y) plane,²¹

$$k_2(\theta) = \frac{\sum k^2(\theta) S(k(\theta), t)}{\sum S(k(\theta), t)}, \quad (6)$$

where θ is the angle between $k(\theta)$ and the k_x direction. Thus $\theta=0$ and $\theta=\pi/2$ correspond to second moments in the x [$k_2^{(x)} = k_2(\theta=0)$] and y [$k_2^{(y)} = k_2(\theta=\pi/2)$] directions, respectively. We find that the anisotropy of the structure factor²² as measured by the quantity $(k_2^{(y)}/k_2^{(x)})$ remains roughly constant over the time region 0–200 MCS/spin. This feature of the anisotropy is an indication of scaling of the structure factor $S(\vec{k}, t)$ with a time-dependent length. We have tested the scaling in various cross sections in the (k_x, k_y) plane by calculating the following scaling functions,

$$F(u) = k_2(\theta, t) S(k(\theta), t), \quad u = k(\theta) / \sqrt{k_2(\theta, t)} \quad (7)$$

for various values of θ . In Fig. 3(a) we show this scaling along the line $2k_x = k_y$. For all the θ values considered, the scaling seems to hold to a good approximation.²³ As an alternative time-dependent length scale we have used $S(0, t)$ (which is proportional to the square of a length) in the following way:⁶

$$\tilde{F}(\tilde{u}) = S(k(\theta), t) / S(0, t), \quad \tilde{u} = \sqrt{S(0, t)} k(\theta). \quad (8a)$$

In Figs. 3(b) and 3(c) we show the scaling along the lines $2k_x = k_y$ and $k_x = 0$, respectively. Scaling also holds in these cases. In addition, we have tested scaling using the second moment of the “full” anisotropic structure factor,

$$k_2(t) = \frac{\sum \vec{k}^2 S(\vec{k}, t)}{\sum S(\vec{k}, t)}. \quad (8b)$$

All these differently defined length scales seem to lead to scaling of $S(\vec{k}, t)$. Thus these length scales are essentially equivalent.

In order to analyze the growth rate of the average domain size, we have used three different definitions of a length scale. The first is due to Sadiq and Binder,⁶ who define a length through

$$S(0, t) = MN(\psi_1^2 + \psi_2^2), \quad (9)$$

so that

$$R_M^2(t) = S(0, t) / \psi_T^2 \quad (10)$$

corresponds to an effective domain area. ψ_T denotes the equilibrium value of the order parameter, which for the low temperature considered in the present study has a value close to unity. The second quantity we use is the perimeter length per unit area (i.e., the number of broken

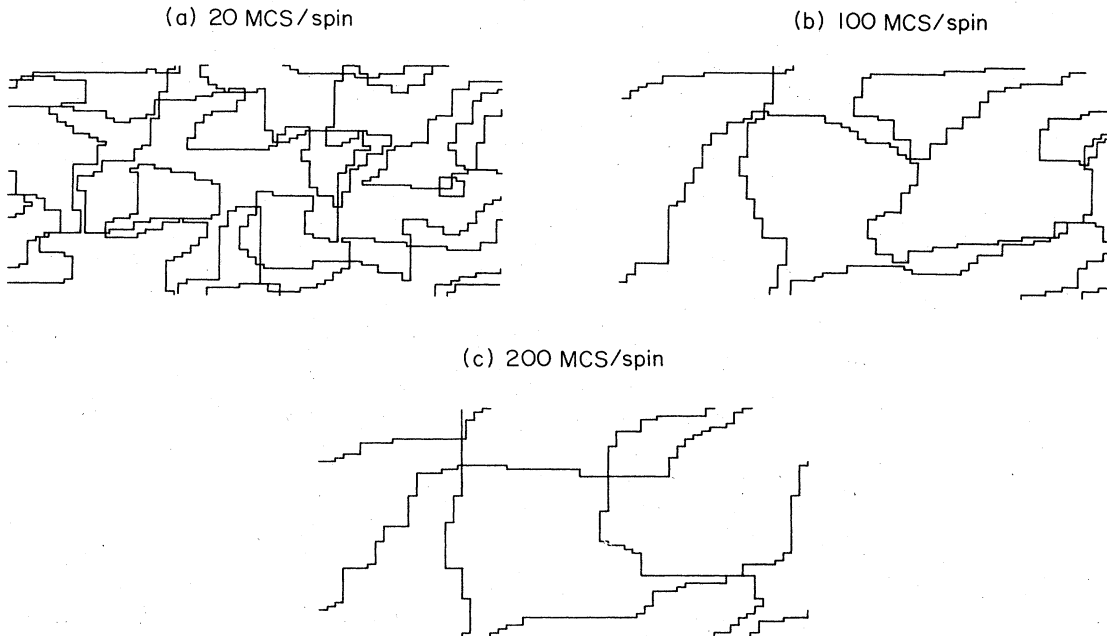


FIG. 2. Typical time evolution of domains after an instantaneous quench. No distinction is made between different types of domain walls of Fig. 1(b).

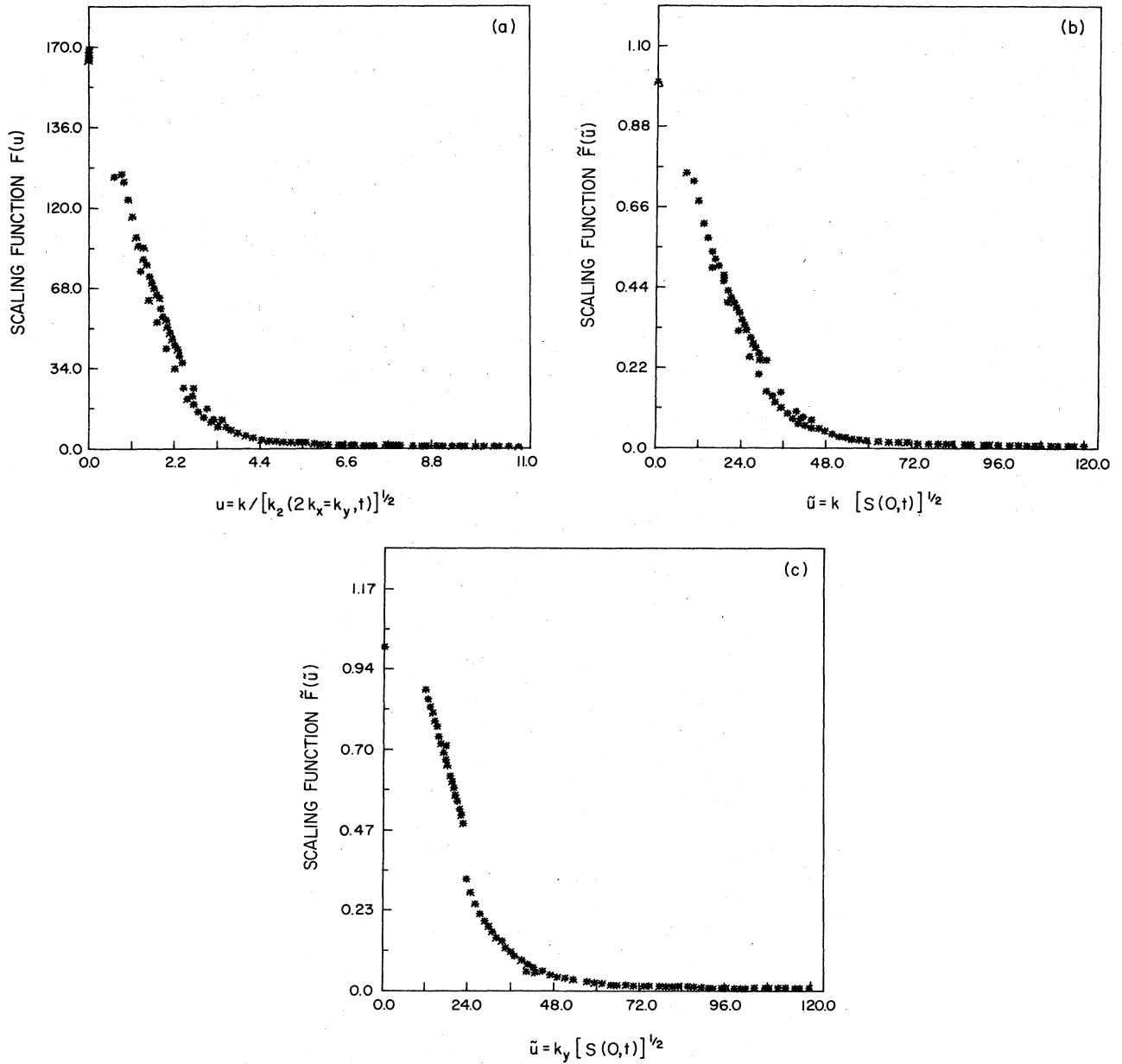


FIG. 3. Dynamical scaling function of the structure factor. (a) Scaling with the second moment $k_2(\theta)$ along the line $2k_x=k_y$. (b) Scaling with the peak height $S(0, t)$ along the line $2k_x=k_y$. (c) Scaling with the $S(0, t)$ along the line $k_x=0$.

bonds)¹³ in the x and y directions, respectively. We will quote results for the inverse perimeter density squared, $A_i(t)$, for the x and y directions, and also for the total perimeter. The third length scale chosen involves the inverse second moment as defined in Eqs. (6) and (8). It should be emphasized that although these length scales all have units of the square of a length, they do differ in their ways of measuring order. $S(0, t)$ measures the long-wavelength correlations in the system $S(0, t) = NM \langle \psi^2 \rangle$. The squares of the inverse perimeter densities, $A_i(t)$ ($i=x, y, \text{tot}$), are sensitive to short-range correlations and can often be related to the average energy in the evolving system.⁶ The inverse second moments probe the intermediate-range correlation.¹³ We consider $S(0, t)$ and

$A_i(t)$ to yield more reliable results than the inverse second moments due to the sensitivity of the latter quantities to the choice of ultraviolet cutoff in Eqs. (6) and (8). It is well known that for scattering from a sharp interface $S(k, t) \sim k^{-d-1}$ for large k .²⁴ Thus the second moment will have an additional, probably weak, time dependence involving the ultraviolet cutoff.

In Fig. 4 we present our data for the $S(0, t)$ and $A_i(t)$.²⁵ It is clear from this figure that finite-size effects start to play an increasingly important role in the growth rates for times greater than about 150 MCS/spin.²⁶ We have also analyzed the behavior of various choices of the inverse second moment. Apart from an initial transient region (< 20 MCS/spin) these quantities have a time dependence

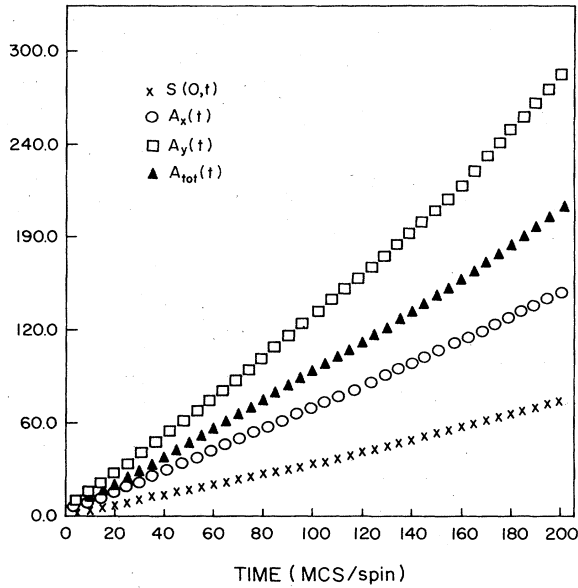


FIG. 4. Growth rate of the long-range order parameter $S(0,t)$ (crosses) and inverse perimeter densities squared, $A_i(t)$; $A_x(t)$ refers to the x perimeter (circles), $A_y(t)$ refers to the y perimeter (squares), and $A_{\text{tot}}(t)$ refers to the total perimeter (triangles) of domains. $S(0,t)$ and $A_i(t)$ have units of area. The vertical scale is arbitrary. Error bars are discussed in the text.

quite similar to that of $S(0,t)$ and $A_i(t)$. It should also be noted that at fixed time the half width of the structure factor in the x direction is considerably narrower than that in the y direction. This reflects the fact, noted in the Discussion, that the overall growth rate is greater in the x direction than in the y direction.

We have analyzed the growth laws of $S(0,t)$ and $A_i(t)$ by performing two different types of least-squares fits: (a) $y = Dt + B$, where B takes into account the initial transient region; (b) $y = Dt^{2n}$. In Table I we present the results for the least-squares analysis together with the correlation factor to indicate the goodness of a fit. The error bars quoted in this table refer to the pointlike errors given by the inverse square root of the number of quenches. The estimates, which were based on the central-limit theorem, can be considered to be conservative. Our results are most consistent with the effective power-law exponent $n \approx 0.50$.

We now briefly discuss the role of different types of domain walls which can appear in the x direction during growth. For the given anisotropy parameter α , the "heavy" and "light" domain walls, Fig. 1(b)(i), are energetically as favorable as the "soft superheavy" and "soft superlight" domain walls shown in Fig. 1(b)(iii). The two

other types of walls, shown in Fig. 1(b), are energetically very unfavorable. Such walls would not occur (beyond a short transient period) for $K_B T/J_0 = 0$. At finite temperatures, however, this picture could change completely due to the role of entropy. To test the density of these various domain walls we have followed the density of the energetically least favorable domain walls, i.e., "superheavy" and "superlight" Fig. 1(b)(ii), over the time region 0–200 MCS/spin for the 300 quenches to $K_B T/J_0 = 0.2$. After an initial transient time, the density of these walls is about 5% of the total density of walls. Thus we expect to find all these different types of walls occurring during domain growth, although heavy-light walls dominate for $t \gtrsim 20$ MCS. Of the two types of anti-phase boundaries which can occur in the y direction, the wall shown in Fig. 1(b)(v) is energetically more favorable. We expect the ground-state degeneracy and the intrinsic softness of domain walls to play a role in determining the growth rates. Within the precision (and time domain) of our study, these effects seem to result in different amplitudes for growth in the x and y directions.

III. DISCUSSION

The scaling property of the anisotropic structure factor yields valuable information about the average morphology of the domain growth. Indeed, we found scaling with a time-dependent length scale. Thus the anisotropic domain growth proceeds via self-similar pattern formation. However, the anisotropy does not seem to introduce a new length scale in the system. All the length scales we have studied are essentially equivalent.

We have also extensively studied the growth law using various choices of length scales. The best estimate for the exponent n which characterizes the average domain size ($n \approx 0.50$) in the present study of a fourfold degenerate modulated system is the same as the corresponding exponent for the twofold degenerate nonconserved (Ising) system.²⁷ We find the time dependence of the growth rates to be the same in both the x and y directions. The overall growth rate, however, is faster in the x direction (i.e., the amplitude is larger in the x direction). Indeed, we find that the ordering "percolates" in the x direction, but not in the y direction, during several quenches in the study. This more rapid evolution in the x direction is at first sight surprising, but presumably is related to the local free energies involved in the kinetics of the domain walls. Clearly, theoretical work on the roles of the various domain walls in the kinetics of ordering in the ANNNI model is necessary, even though the dominant mechanism for growth appears to be the interface curvature.

TABLE I. Analysis of the time dependence of the structure factor and the squares of inverse perimeter densities, as defined in the text. A measure of the goodness of fit is given by the correlation factor ρ .

Quantity	Linear fit $y = Dt + B$			Effective power-law fit $y - y_0 = D(t - t_0)^{2n}$		
	D	B	ρ	D	$2n \pm \Delta$	ρ
$S(0,t)$	3.39	0.17	0.999 86	3.38	1.0 ± 0.1	0.999 87
$A_x(t)$	6.90	11.27	0.999 91	7.29	0.99 ± 0.06	0.999 82
$A_y(t)$	3.24	1.52	0.999 98	3.51	0.98 ± 0.06	0.999 97
$A_{\text{tot}}(t)$	1.14	1.14	0.999 97	1.24	0.98 ± 0.06	0.999 92

ACKNOWLEDGMENTS

We would like to thank Dr. Martin Grant and Mr. Jacques Amar for useful discussions and comments. This work was supported by National Science Foundation

Grant No. DMR-8312958. One of us (T.A.-N.) was supported by a grant from the Neste Corporation of Finland. Two of us (K.K. and T.A.-N.) were also supported by the Finnish Academy.

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- ²¹In calculating a second moment we have used the following cutoff in k space, $k_c = 8 \times 2\pi / N_i a$, where $N_i = M, N$ and a (equal to unity in our units) is the lattice constant.
- ²²At very low temperatures $S(\vec{k}, t)$ for the Ising model could be anisotropic [G. F. Mazenko and O. T. Valls (unpublished)].
- ²³The scaling function for the anisotropic structure has more fluctuations than in the isotropic case. This is due to data smoothing achieved by circular averaging of the isotropic structure factor (see Refs. 4, 10, and 11).
- ²⁴See, for example, *Small Angle X-Ray Scattering*, edited by O. Glatter and O. Kratky (Academic, New York, 1982); this behavior is called Porod's law.
- ²⁵The curve for $A_y(t)$ in Fig. 4 has been calculated taking into account only the antiferromagnetic domain boundaries. Since after a short initial time the type-(v) domain walls [Fig. 1(b)] are very dominant, our approximation only introduces an additional constant, whose value is close to 2. This has been taken into account in the results given in Table I.
- ²⁶Indeed, the curve for $A_y(t)$ increases more rapidly than linearly for $200 \leq t \leq 500$ MCS/spin. We have not included this data in our analysis, since the system often has percolated in the x direction within 200 MCS. Nevertheless, this behavior for $A_y(t)$ may qualitatively reflect the role of domain walls in this regime.
- ²⁷We should note that since the time evolution of this system is so rapid, it is possible that the problem of relating Monte Carlo time to real time might not be trivial. See, for example, the discussion by M. Y. Choi and B. A. Huberman, *Phys. Rev. B* **28**, 2547 (1983).
- ²⁸We have restricted ourselves to where $k_x, k_y \geq 0$. The infinite system $S(\vec{k}, t)$ is anisotropic around the origin of the peak, but in the infinite limit it is expected to become symmetric along the k_x and k_y axes.