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The role of long relaxation times in a simple model with massless modes

R. Timmons

Department of Physics, St. Francis Xavier University,

Antigonish, Nova Scotia, Canada. B2G 2W5

K. De'Bell*

Department of Mathematics, Statistics and Computer Science,

St. Francis Xavier University, Antigonish, Nova Scotia, Canada. B2G 2W5

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Abstract

The next-nearest-neighbour Ising model with competing nearest-neighbour (nn) and next - nearest-neighbour (nnn) interactions, provides an example of a system in which massless modes destroy order at any finite temperature. This occurs only at a critical ratio, K_c , of the nnn and nn interactions. In this paper we investigate the role of long relaxation times in determining the behaviour of the system when the ratio of the nnn and nn interactions, K , is at and close to this critical ratio. Despite the absence of an order-disorder transition in systems with $K = K_c$, in Monte Carlo simulations reported here the temperature dependent behaviour of the relaxation times indicates the existence of a glass transition with a glass transition temperature of $T_g \approx 0.26$ in a model with nearest neighbour interaction $J = k_B = 1$.

keywords next-nearest-neighbour interactions, Ising model, phase diagram, glass-like transition

* Communicating Author, email: kdebell@stfx.ca

I. INTRODUCTION

Simple models of ultra-thin film systems that exhibit pattern formation due to competing interactions remain of considerable interest despite the long history of these models. This continued interest is, in part, due to the range of physical systems that exhibit pattern formation. Such systems include garnet films[1], ultra-thin magnetic systems [2], and fluid layers exhibiting nano-phase separation[3]. The interest in these systems is further driven by the continued evolution of experimental methods that allow the microscopic characterization of both equilibrium and non-equilibrium properties of such systems, and offer the possibility of manipulation of the pattern structures[3–7].

If a system is in thermal equilibrium and ordered, regions of the other potential ordered ground state patterns are dynamic structures that exist in a background of the ordering ground state pattern and are generally unstable as they dissolve into the background and/or break-up due to the formation and growth of internal voids. These dynamic processes maintain the equilibrium distribution of regions of these minority patterns. However, if relatively stable regions of the potential ground state patterns occur these may move the system from the equilibrium distribution of the phases[8]. For example, in ultra-thin magnetic films with uniaxial out-of-plane moments and both nearest neighbour (exchange) interactions and long range, dipolar interactions, the ground-state ideal patterns are stripes of “up”spins alternating with stripes of “down”spins[2]; however if such a system is initially saturated in an all up or all down (i.e. ferromagnetic) ideal pattern then allowed to relax it will exhibit the formation of both regions with vertical stripes and regions with horizontal stripes [9]. The coexistence of these regions may be very long lived and is associated with locked in topological structures where the two types of region meet. More generally, relatively stable structures in which two (or more) of the possible ordered patterns coexist may occur when this corresponds to the “locking-in” of topological structures associated with the symmetry operations that connect the possible ideal ground state patterns. (In what follows, the formation of these relatively stable coexisting regions and their dynamics are referred to as “non-equilibrium processes”).

In quasi-two-dimensional physical systems that have ground states of (ideal) alternating stripe patterns, pattern formation is often due to competition between short-range interactions and longer-range interactions that have a power law dependence on distance

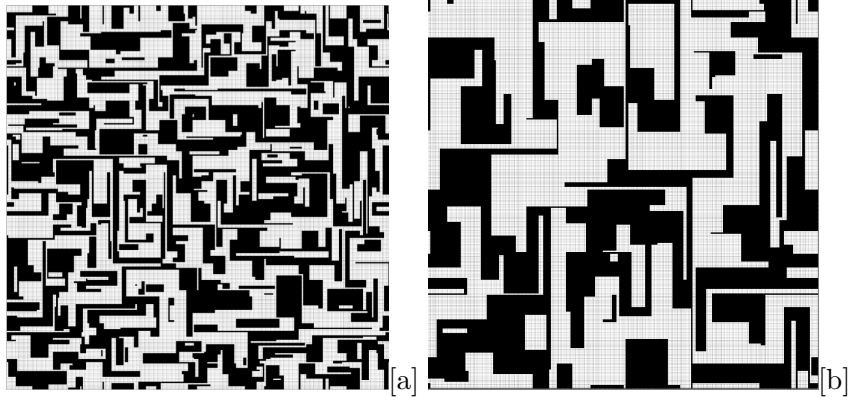


FIG. 1. Typical spin configurations observed during Monte Carlo simulations of a system with $L = 256$, $K = -0.5$, at temperatures $T = 0.36$ [a] and $T = 0.26$ [b].

[2, 10, 11]. However, striped pattern formation may be observed in even simpler models such as the next-nearest neighbour Ising model with competing nearest-neighbour and next-nearest-neighbour interactions[12]. In experimental realizations of pattern forming quasi-two dimensional systems observed structures are often labyrinth like (rather than perfectly ordered striped phases). Monte Carlo studies of the nnn-Ising model also exhibit labyrinth-like structures (e.g. Fig 1). In the work described here, the nnn-Ising model is used to illustrate the role that long relaxation times and the associated locking-in of topological defects play in such systems.

In the following section the model is defined and some key aspects of its history reviewed. Sect. II also contains a summary of some symmetry considerations regarding this model. The main results of this study, derived from Monte Carlo simulation of the model, are presented in Sect III. Sect. IV concludes this study with some commentary on the possible generality of these results.

II. MODEL DEFINITION AND BACKGROUND

The two-dimensional, nnn-Ising model is constructed by associating with each unique point, i , on a lattice, a local variable, $s_i = \pm 1$. Hereafter the local lattice variables are referred to as spins and the language of magnetic systems is used to describe the macroscopic properties of the model. In this study, the spin variables are distributed on a square lattice

with cyclic boundary conditions. The Hamiltonian of the system is then.

$$-\mathcal{H} = J \sum_{nn} s_i s_j + K \sum_{nnn} s_i s_j \quad (1)$$

The first term on the right-hand-side of this equation is a sum over lattice points such that i and j are nearest neighbours on the lattice. Throughout we take the parameter J to be positive and in the numerical work described below it is set to equal the Boltzmann constant, k_B , except where stated otherwise. The second sum on the right-hand-side of the equation is the corresponding sum over next-nearest-neighbour pairs. Thus the parameters of the model are the nnn interaction parameter K and the temperature T . (It is not necessary to consider the $J < 0$ case as the well known gauge transform

$$\sigma_i = (-1)^{x_i+y_i} s_i; \quad J' = -J \quad (2)$$

transforms a checkerboard antiferromagnetic ground state to a ferromagnetic state but does not affect the value of K and transforms striped ground states to striped ground states. Therefore the properties of the model with negative J are trivially related to those with the corresponding positive value of J .)

In 1969, Fan and Wu[12] performed a detailed analysis of the ground state properties of this model. They showed that the ground states of the system are ferromagnetic (all spins =+1 or all spins= -1) if the next-nearest-neighbour parameter, K , exceeds a critical value; i.e., for $K > K_c = -0.5J$; however, for $K < K_c$ the ground states consist of stripes of spins aligned in the +1 direction alternating with stripes of spins aligned in the -1 direction. Each stripe of aligned spins is one spin wide. Thus there is a change from two ferromagnetic ground states to four striped ground states (two with stripes in the x (horizontal) direction and two with stripes in the y (vertical) direction) at $K = K_c$. Further Fan and Wu noted that the degeneracy of the striped and ferromagnetic ground states at $K = K_c$ means that the system does not have a non-zero order-disorder critical temperature at this value of K . The mechanism that destroys order in an infinite system at any finite temperature if $K = K_c$, can easily be visualized by considering the system initially in a striped phase with a single spin flipped in the opposite direction to the stripe it is in. Spin flips can then propagate along the stripe without energy cost (i.e. these dynamical modes are massless). In an infinite system the probability of a single flipped spin in each stripe is non-zero at any non-zero temperature and hence the striped phases are unstable. Similarly the ferromagnetic

states are destroyed by the zero-energy propagation of spin flip excitations in the infinite system at any non-zero temperature.

Subsequent studies of the finite temperature phase diagram of this model involved some controversy. In particular, different studies resulted in different conclusions about the nature of the transition for values of K close to K_c . Early work[13, 14] took the order-disorder transition to be continuous; however later work indicated that a discontinuous transition could occur for parameter values in the range $-1.2J < K < -0.5J$ [15–18]. More recently, Murtazaev et al. [19] have applied histogram Monte Carlo methods with Binder cumulant analysis, and concluded that the transition is continuous in the range $-1.0J < K < 0.1$. Also recently, Jin et al.[20, 21] have considered the nature of the order-disorder transition in this model. These authors also used Monte-Carlo combined with Binder cumulant based methods as well as cluster-mean-field theory and transfer matrix methods. Jin et al. concluded that there is evidence for a weak discontinuous transition in a narrow range of values $K < K_c$ but point out that a continuous transition can not be fully ruled out[21].

In this article, we set aside the question of the order of the transition when $K \neq K_c$ and focus primarily on the role of slow dynamics in the absence of an order-disorder phase transition at $K = K_c$. However we also present some evidence that these slow dynamics play a role in the behaviour of the system when K is close to but not equal to K_c .

In the simulations described in the next section, the specific heat, magnetization, and (initial) susceptibility, were calculated directly using:

$$C_v = \frac{\langle E^2 \rangle - \langle E \rangle^2}{NT^2}; \quad (3)$$

$$M = \sum_{i=1}^N s_i; \quad \chi = \frac{\langle M^2 \rangle - \langle M \rangle^2}{NT} \quad (4)$$

in the above equations, the Boltzmann constant $k_B = 1$, E is the configuration energy and $\langle \rangle$ denotes a configurational average.

To calculate the components of the staggered magnetization and staggered susceptibility corresponding to the ordered phases with striped ground states we perform two gauge transformations

$$\sigma_i^h = (-1)^{x_i} s_i; \quad \sigma_i^v = (-1)^{y_i} s_i \quad (5)$$

The first of these converts the horizontal stripe ground states to ferromagnetic ideal patterns (and converts the vertical stripe ground states to chequerboard antiferromagnetic ideal pat-

terns). The second converts the vertical stripe ground states to ferromagnetic ideal patterns (and converts the horizontal stripe ground states to checkerboard antiferromagnetic ideal patterns). The staggered magnetization, $\vec{M}_s = (M^h, M^v)$, is then calculated from

$$M^h = \sum_{i=1}^N \sigma_i^h; \quad M^v = \sum_{i=1}^N \sigma_i^v \quad (6)$$

and the staggered susceptibility, $\vec{\chi}_s = (\chi^h, \chi^v)$ is calculated using

$$\chi^h = \frac{\langle M^{h2} \rangle - \langle M^h \rangle^2}{NT}; \quad \chi^v = \frac{\langle M^{v2} \rangle - \langle M^v \rangle^2}{NT} \quad (7)$$

It is convenient to calculate the autocorrelation function for $t > 0$:-

$$\gamma(t) = \left\langle \frac{\sum_{i=1}^N s_i(0)s_i(t)/N - m(0)m(t)}{1 - m(0)m(t)} \right\rangle \quad (8)$$

where t is a time measured in Monte Carlo cycles, and $m(t)$ is the magnetization per lattice site at time t . The definition above is appropriate for systems with ferromagnetic ground states as the subtraction of the second term in each of the numerator and denominator ensures that the autocorrelation function is zero if t exceeds the system relaxation time in the ferromagnetic state. The staggered form of the autocorrelation functions for the two types of stripe phase is a straightforward generalization of the ferromagnetic form for the autocorrelation function:-

$$\gamma^h(t) = \left\langle \frac{\sum_{i=1}^N s_i(0)s_i(t)/N - m^h(0)m^h(t)}{1 - m^h(0)m^h(t)} \right\rangle \quad (9)$$

$$\gamma^v(t) = \left\langle \frac{\sum_{i=1}^N s_i(0)s_i(t)/N - m^v(0)m^v(t)}{1 - m^v(0)m^v(t)} \right\rangle \quad (10)$$

(The usual initial condition, $\gamma(0) = \gamma^h(0) = \gamma^v(0) = 1$, is understood).

As the autocorrelation function is used, rather than the more general two point correlation function that involves both a spatial displacement and a time displacement of the two spins, Eqs. 8, 9 and 10 differ only in the order parameter that is used in the subtracted terms in the numerator and denominator. These subtracted terms ensure that the autocorrelation function corresponding to the ordered state of the system, will be zero if t is large compared with the relaxation time, τ . Moreover, if the system is ordered either in one of the ferromagnetic states or one of the striped states the autocorrelation functions corresponding to the other two types of possible ordered states satisfy

$$\gamma_d \approx m_o^2 \quad t > \tau \quad (11)$$

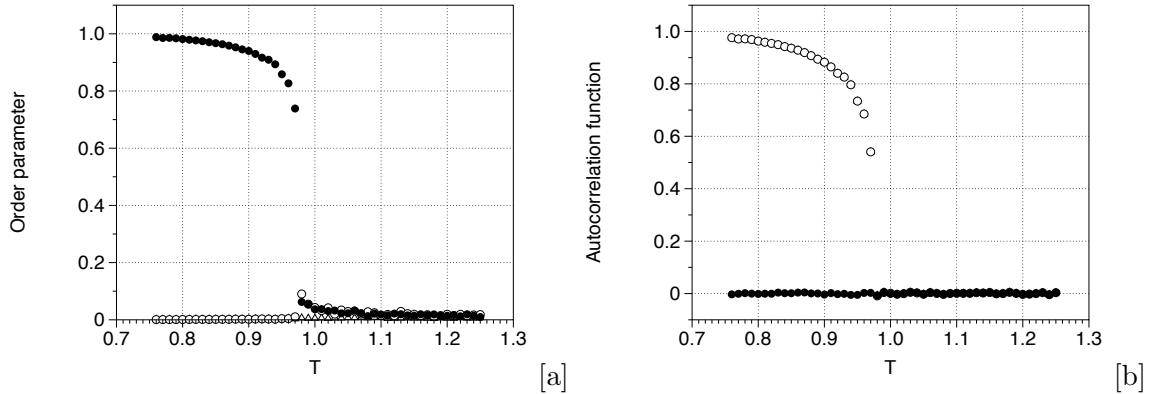


FIG. 2. Variation of the vertical striped (closed circle), horizontal striped (open circle) ferromagnetic (triangle) order parameters [a] and the corresponding autocorrelation functions [b] as a function of temperature from a typical warming simulation run with $K = -0.6$ and $L = 128$

where γ_d is the autocorrelation function for one of the non-ordering types of state and m_o is the order parameter of the ordering state. This relation is approximate in any finite system since the autocorrelation function of the ordered state is not perfectly zero in a finite system.

Consequently, if the system is in equilibrium and orders in one of the possible ordered states below the critical temperature, the temperature at which the autocorrelation functions of the non-ordering states becomes (effectively) zero provides an estimate of T_c which must be consistent with the values obtained from other observables in the usual way (Fig. 2). Thus, in a coordinated examination of the low temperature phase, involving the autocorrelation functions corresponding to all of the possible ordered ground states, deviation from this expected behaviour for the autocorrelation functions of the ordering and non-ordering ground states may indicate that the relaxation time of the system exceeds the simulation time.

III. MONTE CARLO SIMULATIONS

A. Equilibrium properties $K \neq K_c$

The phase lines separating the disordered phase from the ordered phase at given values of the nnn interaction parameter, K , were determined using conventional Monte Carlo methods based on a Metropolis algorithm with Glauber dynamics. Typically linear system sizes of $L = 128$ and $L = 256$ were used; however systems with $L = 512$ were used for some

consistency checks. Simulation runs included warming runs in which the system began in a fully ordered striped phase ($K < K_c$) or fully ordered ferromagnetic phase ($K > K_c$) at a low temperature and the temperature was slowly increased, and cooling runs in which the system began in a disordered state ($s_i = \pm 1$ with equal probability) at a high temperature and the temperature was slowly decreased. In addition, a small number of simulations in which temperature, T , was fixed and K was slowly varied were also performed.

For all values of $K \neq K_c$ considered the behaviour of the system observed in the simulations was consistent between heating and cooling and indicated a continuous order phase transition, providing that the number of Monte Carlo steps after each temperature change was sufficiently large and providing that the number of Monte Carlo steps used for data collection was sufficiently large. Moreover, providing that the number of Monte Carlo steps used was sufficiently large, at all values of $K \neq K_c$ common to this study and that of Murtazaev et al., the results obtained for T_c in this study were consistent with those of Murtazaev et al.[19, Fig.10]. (Values of K used in this study include values both more distant from K_c and closer to K_c than those used by Murtazaev et al.). Likewise, if the number of Monte Carlo steps used was sufficient, no significant size effects in T_c were observed for systems with $L \geq 128$. However, if the number of Monte Carlo steps was not sufficient (or if the system size was $L \leq 64$) consistent results were not obtained and the nature of the transition became difficult to discern.

As is well known, systems in equilibrium approaching a continuous phase transition exhibit a diverging correlation length and a diverging relaxation time. Consequently, for systems in equilibrium, attention must be paid to the number of Monte Carlo steps and the system size used in the simulation. However, as will be shown below, the behaviour of the nnn Ising model at and close to K_c indicates that the system behaviour is influenced by non-equilibrium processes. To exhibit the role played by non-equilibrium processes in determining the properties of the system, in the remainder of this section we focus on the ferromagnetic autocorrelation function, γ , and the components of the staggered autocorrelation function for the striped phases, $\vec{\gamma}_s = (\gamma^h, \gamma^v)$, and relaxation times that can be defined in terms of these autocorrelation functions.

B. Non equilibrium properties

As noted in Sect. II, if o denotes the majority phase, ordered state (ferromagnetic, vertical stripes or horizontal stripes) and d denotes either of the minority ordered phases, if the system is in equilibrium then γ^d is related to the order parameter of the majority phase, M^o by Eq. 11.

For example, for a system with $L = 128$ and $K = -0.6$ if $\tau = 5000$ is chosen the observed behaviour of the autocorrelation function is consistent with Eq. 11 and the value of T_c obtained from γ^d in either a cooling or warming run (and either choice of d) is consistent with the value of T_c obtained from the initial susceptibilities of the same system (Fig. 2). More generally, for any $K \neq K_c$ considered it was possible to make τ sufficiently large that the expected equilibrium behaviour was observed and consistent values of T_c obtained.

However, simulations of the system at $K = K_c = -0.5$ do not show results that are consistent with either a simple order-disorder transition or a smooth reduction in disorder at very low temperatures even at the largest values of τ considered. In particular, if the system is warmed from either a striped ordered state or a ferromagnetic state at low temperature, the autocorrelation function corresponding to the ordered state, quickly reaches a value close to one; then all three autocorrelation functions decline until they reach a value of approximately zero at $T_a = 0.39 \pm 0.01$ (Fig. 3). However there is no discernible evidence of a well defined transition in the specific heat data. The susceptibility data shows peak values at a temperature of $T = 0.31 \pm 0.02$ and the order parameters effectively vanish at this value of T . (Typically two of the three initial susceptibilities (χ, χ_h, χ_v) exhibit discernable peaks but at slightly different values of T within this range). The behaviour is similar if the system is initiated in a disordered state and cooled from a high temperature except that there is in this case no discernible evidence for a phase transition in the order parameter data. This behaviour is observed even in systems of linear size up to $L = 256$ and there is no significant variation in the behaviour with system size when results for systems with $L = 128$ and $L = 256$ are compared.

The reason for this anomalous behaviour when $K = -0.5$ becomes apparent if we consider the temperature behaviour of the relaxation time τ^* defined as the value of τ at which any of the three autocorrelation functions (Eqs. 8, 9 and 10) reaches a value ≤ 0.01 . Initializing the system in a random state then cooling from a high temperature, for both $K = -0.55$

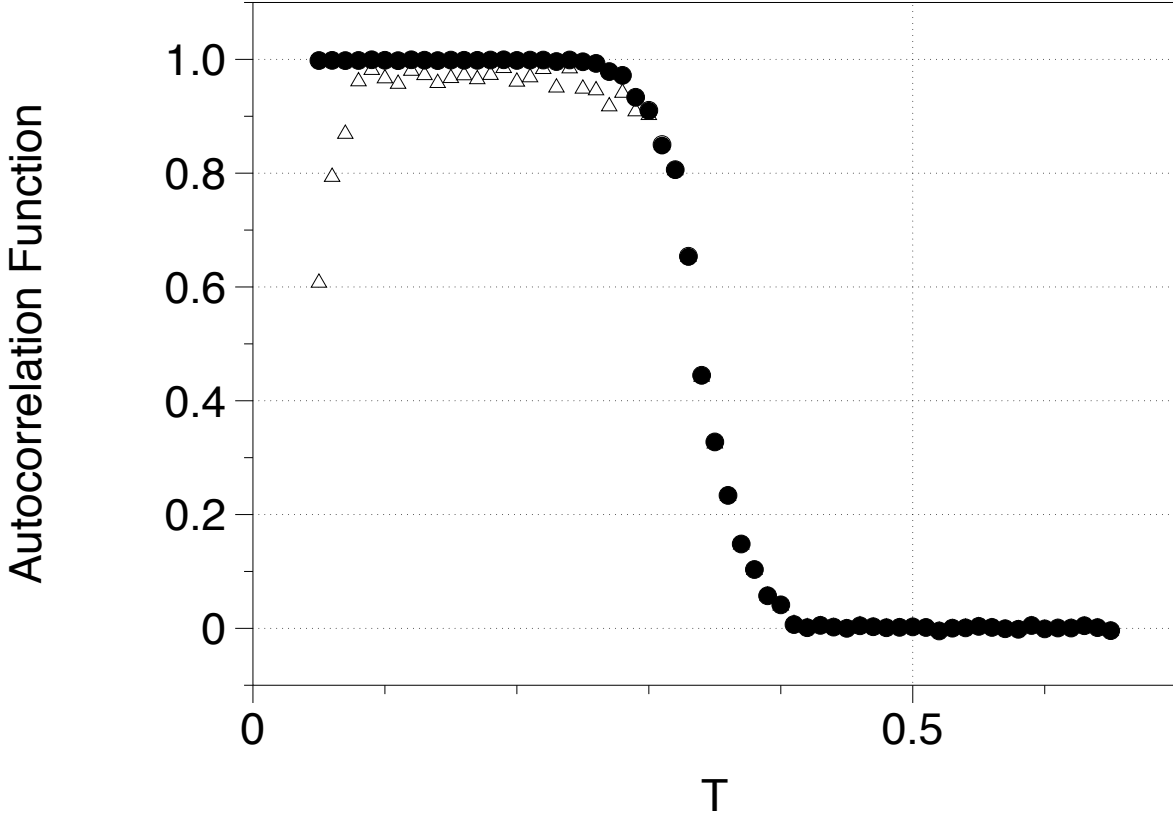


FIG. 3. Variation of the vertical striped (closed circle), horizontal striped (open circle) ferromagnetic (triangle) autocorrelation functions as a function of temperature from a typical warming simulation run with $K = -0.5$ and $L = 128$. The system was initially prepared in a ferromagnetic ground state but similar results are observed if the system is initially prepared in a striped ground state.

and $K = -0.45$ we find that the value of τ^* has a high sharp peak at T_c but on either side of T_c rapidly declines as a function of temperature as $|T - T_c|$ increases. However, if the system with $K = -0.50$ is cooled from a high temperature state the value of τ^* appears to diverge but as the temperature continues to decrease the value of τ^* does not show a peak and subsequent decline but rather remains in excess of the number of Monte Carlo cycles in the simulation. The data can be fitted by assuming

$$\tau^* = A|T_g - T|^{-\rho} \quad T > T_g \quad (12)$$

and the resulting best fit values are $T_g = 0.261 \pm 0.001$ and $\rho = 3.38 \pm 0.02$ (Fig. 4).

Further, if the system with $K = -0.50$ is cooled from a high temperature state to a

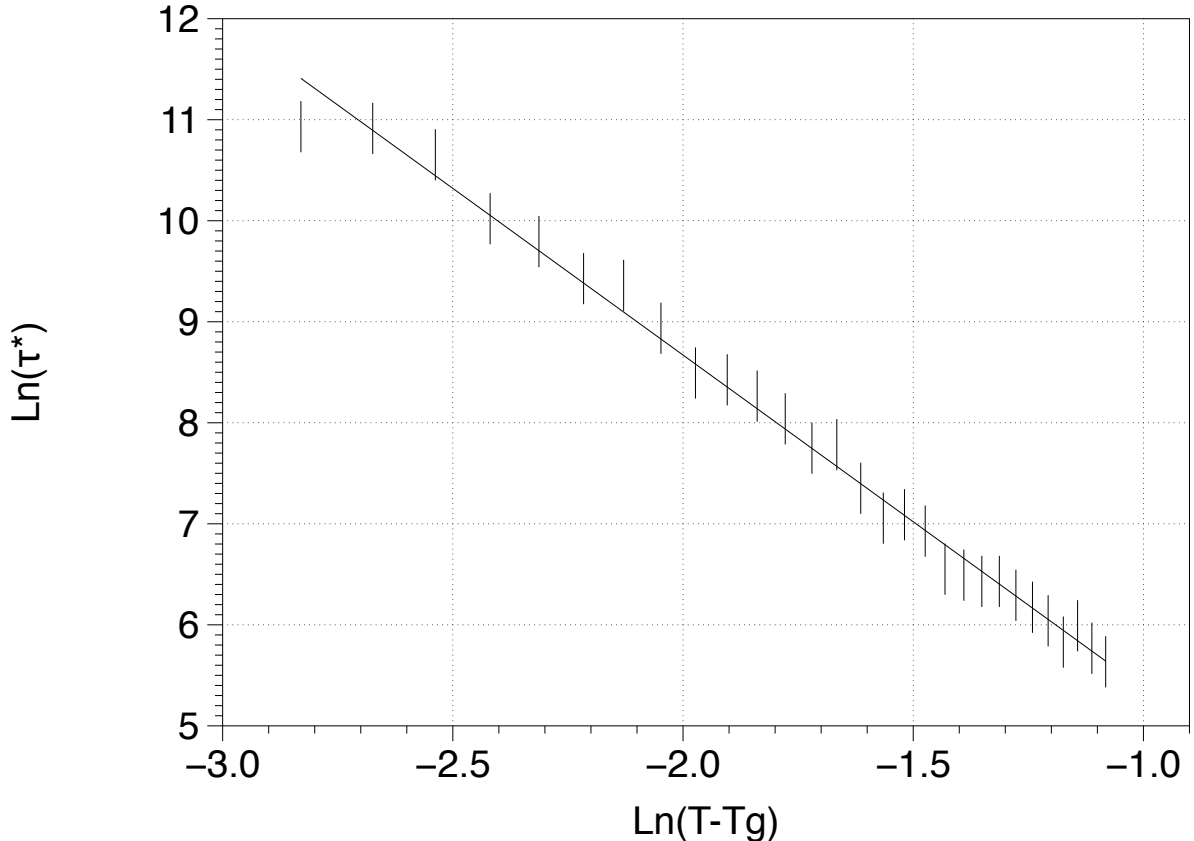


FIG. 4. Log-Log plot of the variation of the relaxation time τ^* with temperature, in a typical run for a system with $K = -0.5$ and $L = 128$

temperature between T_a and T_g , the autocorrelation function at that temperature can be fitted by assuming a stretched exponential dependence on t [22, 23]. I.e.

$$\gamma = B e^{(-t/\tau)^\beta} \quad (13)$$

where τ is a function of the temperature, T , at which the autocorrelation function is calculated. Assuming the same power law behaviour as in Eq. 12, an analysis of the increase in τ as T_g is approached from above, is consistent with the values of T_g and ρ found in the analysis of τ^* (Fig. 5).

Typical spin configurations for $K = K_c$ at $T_g < T < T_a$ and $T \approx T_g$ are shown in Fig. 1. Observation of video clips formed from individual spin configurations in this temperature range show that the dynamics are extremely slow when $T_g < T < T_a$ and are essentially frozen when $T < T_g$. The implications of this observed behaviour are discussed in Sect. IV.

The observation of the divergence of τ^* as the temperature approaches $T_g > 0$ from above

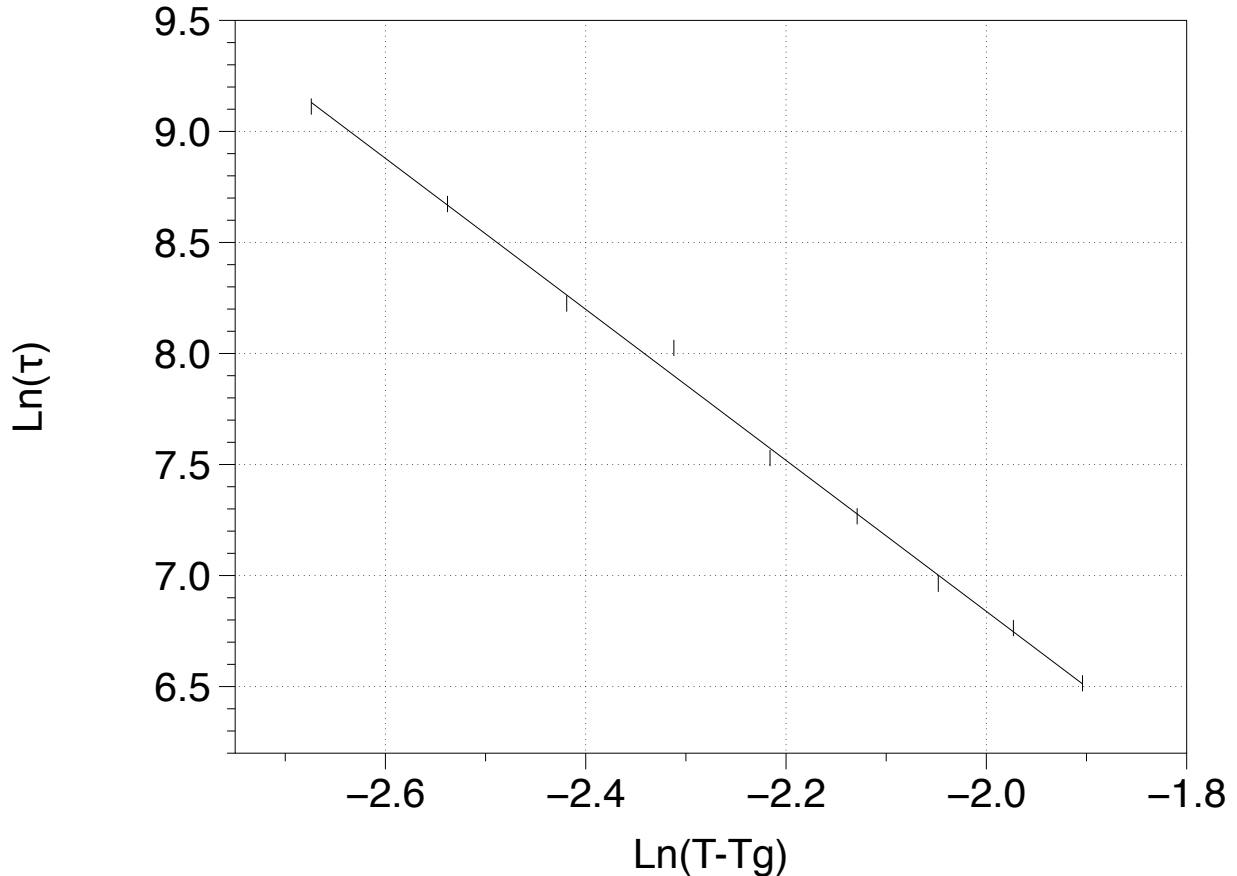


FIG. 5. Log-Log plot of the variation of the relaxation time τ with temperature, in a typical run for a system with $K = -0.5$ and $L = 256$

when $K = -0.5$ indicates that while no order-disorder transition exists at finite temperature, as the system is cooled the slowing of dynamical processes give rise to a significant change in the properties of the system as T_g is approached.

To determine if there is any evidence that this dynamical behaviour affects the properties of the system when $K \neq K_c$, a similar analysis of τ^* was performed for systems with $K = -0.55$ and $K = -0.45$. For $K = -0.55$ this results in a value of $T_g = 0.7695 \pm 0.0005$ which is indistinguishable from the estimate of T_c obtained from the order parameter, specific heat, and staggered susceptibility ($T_c = 0.77 \pm 0.01$). I.e. for a system with $K_c = -0.55$ there is no evidence that the existence of the minority phases result in any deviation from the normal equilibrium behaviour in the vicinity of a second order transition, including critical slowing down. However, for systems with $K = -0.45$ a value of $T_g = 0.611 \pm 0.001$ is obtained. This value of T_g is significantly lower than the estimate of T_c obtained from the

order parameter, autocorrelation function and susceptibility ($T_c = 0.66 \pm 0.02$). We take this to indicate that there is some effect due to the existence of minority (striped) phase regions as T approaches T_c from above but these effects are essentially removed once T_c is reached and the system orders ferromagnetically. The observation that the specific heat data shows an unusual high level of noise in the range $0.62 < T < 0.70$ appears to further support this view.

Previously Westfahl et al.[10] have discussed glass formation in the context of striped systems with competing short-range and long-range (Coulomb) interactions. In particular, Westfahl et al. noted that the glass transition is associated with the occurrence of a large number of possible metastable states with regions of stripes with different orientations and pattern defects, and that entropy-driven transitions between different states result in dynamical processes with a relaxation time determined by the Vogel-Fulcher law[10, Eq.6]. Accordingly, the Monte Carlo data obtained with $K = -0.5$, for the relaxation time τ^* was fitted to the form of the Vogel-Fulcher law; i.e.

$$\tau^* = A e^{BT_0/(T-T_0)} \quad (14)$$

The values for τ^* are equally well fitted by this form as by the power law form of Eq. 12 and a value of

$$T_0 = 0.091 \pm 0.005 \quad (15)$$

is obtained. This is consistent with the expected result $T_0 < T_g < T_a$ where T_g represents the observed glass transition temperature, which may depend on factors such as cooling rate, and T_0 represents a theoretical limit at which the system freezes into a glass like state even if the cooling rate is infinitely slow. (For a fuller discussion of these distinct characteristic temperatures see Ref. [10]). However, it should be noted that here both the value of T_g and the value of T_0 are obtained by fitting a single data set obtained at $T > T_g$, to different assumed fitting functions. From a practical point of view, this reflects the difficulty in the fitting of data determined by underlying processes with a mixture of time scales.

IV. SUMMARY AND DISCUSSION

While the occurrence of a finite temperature, order-disorder phase transition in the nnn Ising model with $K = K_c$ is excluded by the existence of massless modes as outlined in

Sect. II, the analysis of the relaxation times presented in Sect. III indicates that this system undergoes a dynamical, glass-like transition as the system is cooled from high temperatures. Both the analysis of the relaxation time τ^* and the stretched exponential analysis of the autocorrelation function result in a “glass transition temperature” of $T_g = 0.261 \pm 0.01$. For a detailed discussion of the relation between stretched exponentials and glass-like states, the reader is referred to the review by Phillips[22]. (The possibility of glass like phases in the nnn Ising model with competing interactions has previously been discussed by O’Hare et al[24, 25] whose results were based on small plaquette calculations and Monte Carlo simulations for systems of $L = 100$. These authors also discussed the energetics of interfaces between regions corresponding to different ground states and showed examples of labyrinth like structures obtained in the $L = 100$ Monte Carlo simulations[24, Fig.20] . Based on a 5-site plaquette calculation, O’Hare et al concluded that in a range of approximately $-0.67 < K < -0.33$ (in the units of the current article), a phase exists with (possibly) complex ordering and an order-disorder transition temperature higher than the critical temperature of the striped ($K < K_c$) or ferromagnetic ($K > K_c$) phase[25, Fig.2].)

The results in Sect. III indicate that the nnn Ising model exhibits a glass-like transition ($\tau^* \rightarrow \infty$ when $T \leq T_g$) only for a special choice of the model parameters, i.e. for $K = K_c$. Nonetheless, the occurrence of glassy states in physical systems is common[22]. Rapid progress in the preparation and manipulation of quasi two-dimensional systems experimentally, continues to draw focus on the pattern structures observed in such systems. The structures observed are often labyrinth-like and display two or more phases each existing in mesoscopic or macroscopic regions[1, 3, 5]. That is, they are more like the glass-like structures generated by the dynamical transition that occurs at $K = -0.5$ than like the equilibrium ordered phase patterns corresponding to the ground states. Further, simulation studies of a two dimensional model of ultra-thin films with both short ranged and long ranged interactions have also observed the “locking-in” in states with coexisting regions of multiple phases that are stable over long-time periods[9]. In the course of this work, we have also observed that for systems with K not equal to K_c but close to its value, these types of states can be formed by sufficiently rapidly cooling the system to below its order-disorder transition temperature. Once formed, these states are stable over relatively long periods of time[26].

As noted at the end of Sect. III, the present study provides some evidence that if $K > K_c$

but is close to K_c in value, even if the system is cooled sufficiently slowly to observe the order-disorder transition, the slow dynamics of the minority phase regions play some role in determining the properties of the system.

In summary, the nnn Ising model is a simple toy model that may be used to investigate non-equilibrium glass-like behaviours typical of those that occur in a wide variety of quasi two-dimensional systems. Particularly, this system offers the possibility of further investigating the interface between equilibrium behaviour and non-equilibrium behaviour by changing two parameters, K and T , and by varying the rate of change of these variables. Or to state this another way, to fully understand the properties of this system when $K \approx K_c$ it may be necessary to include the dependence on the time regime ($t \ll \tau, t \approx \tau, t \gg \tau$) through appropriate scaling laws.

In the context of time dependence in pattern forming systems, a recent study by Horowitz et al.[27] of short-time dynamics of the transition from the striped phase to the tetragonal phase in a square lattice Ising model with exchange and dipolar interactions is of interest. The study reported here is limited to the square lattice model and therefore the degeneracy that destroys the ordered state when $K = K_c$ arises only from the competition between the interactions. However, in view of recent advances in methods for fabricating ultra-thin films, studies of dynamics in systems with structural frustration will also be of interest (see, e.g., the work by Holden et al.[28] and Maksymenko et al.[29] and references therein).

Despite the conceptual simplicity of the next-nearest-neighbour Ising model, it exhibits complex behaviour which demonstrates how topological defects affect the behaviour of pattern forming systems. The order of the transition when K is not equal to K_c but close to it, is not the primary focus of the work reported here, however, the existence of these slow dynamical modes suggests that methods that explicitly take into account non-equilibrium effects may be necessary to answer that question. Work by Shirakura et al.[30] on a simple model with competing interactions has also identified possible discrepancies between simulation results from equilibrium studies and those from non-equilibrium studies.

A remaining question is the precise behaviour of the order-disorder phase transition lines as the critical value of K is approached. Some previous authors provide a phase diagram that show the order-disorder transition lines continuously approaching $T = 0$ with a finite slope[19, 25]. However, at the smallest value of $|K - K_c|$ considered in this work, the values of T_c for both the striped phase and the ferromagnetic phase are well above $T = 0$ and

simple extrapolation of the phase transition lines to $K = K_c$ result in non-zero estimates of the limits $\lim_{K \rightarrow K_c^+} T_c(K)$ and $\lim_{K \rightarrow K_c^-} T_c(K)$. For example, fitting the data points for each of the phase lines by a simple quadratic function for each $K > K_c$ and $K < K_c$:

$$T_c = m_1|K - K_c| + m_2|K - K_c|^2 + b \quad (16)$$

results in a R-squared value of approximately $R^2 = 0.9995$ in each case but with a non-zero intersection with the line $K = K_c$ (Fig. 6). Nonetheless, given the difficulties of numerical studies close to $K = K_c$ outlined in this paper we can not rule out the possibility that the phase transition lines approach the line $K = K_c$ asymptotically thus giving the result $\lim_{K \rightarrow K_c^+} T_c(K) = \lim_{K \rightarrow K_c^-} T_c(K) = 0$. In the results of the current study, there is some (albeit weak) support for this speculation of an asymptotic behaviour of the phase lines. For $y = |K - K_c| > 0.2$, $T_c(y)$ is higher in the ferromagnetic case than in the striped phase case. However for $y < 0.2$ (the last two data points for each ordered state type), this is reversed; i.e. the rate of increase of the absolute value of the slope of the T_c vs. $|K - K_c|$ line is greater for the phase line separating the disordered phase from the ferromagnetic phase than for the phase line separating the disordered phase from the striped phase (Fig. 6). Assuming this trend continues, either the two one-sided limits are unequal (as suggested by simple extrapolation) or the limit $\lim_{K \rightarrow K_c^+} T_c(K) = \lim_{K \rightarrow K_c^-} T_c(K) = 0$ is approached asymptotically. For the purpose of illustration, the asymptotic regression function

$$T_c = C(e^{|K-K_c|} - 1)^D \quad 0 < D < 1 \quad (17)$$

is plotted on the phase diagram in Fig. 7. (R^2 values are 0.9985 and 0.9916 for $K > K_c$ and $K < K_c$ respectively).

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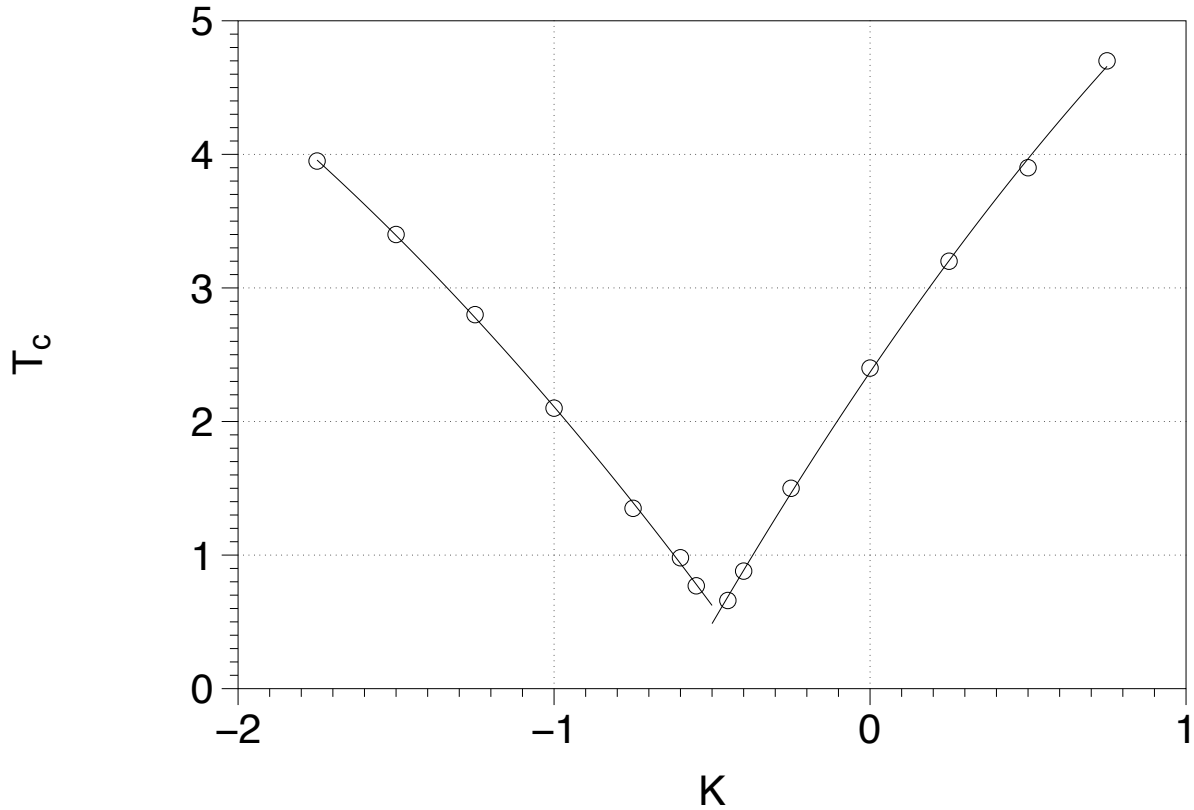


FIG. 6. Phase diagram of the nnn-Ising model. Open circles show the critical temperature for the order disorder transition for the striped phase ($K < -0.5$) and the order disorder transition for the ferromagnetic phase ($K > -0.5$). Solid lines show the quadratic regression function (Eq. 16)

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- [1] D. Andelman and R. E. Rosensweig, *J. Phys. Chem.* **113**, 3785 (2009).
 - [2] K. De’Bell, A. B. MacIsaac, and J. P. Whitehead, *Rev. Mod. Phys.* **72**, 225 (2000).
 - [3] N. Severin, J. Gienger, V. Scenev, P. Lange, I. M. Sokolov, and J. P. Rabe, *Nano Letters* **15**, 1171 (2015).
 - [4] M. Kronseder, T. N. G. Meier, M. Zimmermann, M. Buchner, M. Vogel, and C. Back, *Nature Communications* **6**, 6832 1 (2015).
 - [5] J. Kemmer, S. Wilfert, J. Kugel, T. Maurer, P. J. Hsu, and M. Bode, *Phys. Rev. B* **91**, 184412 (2015).

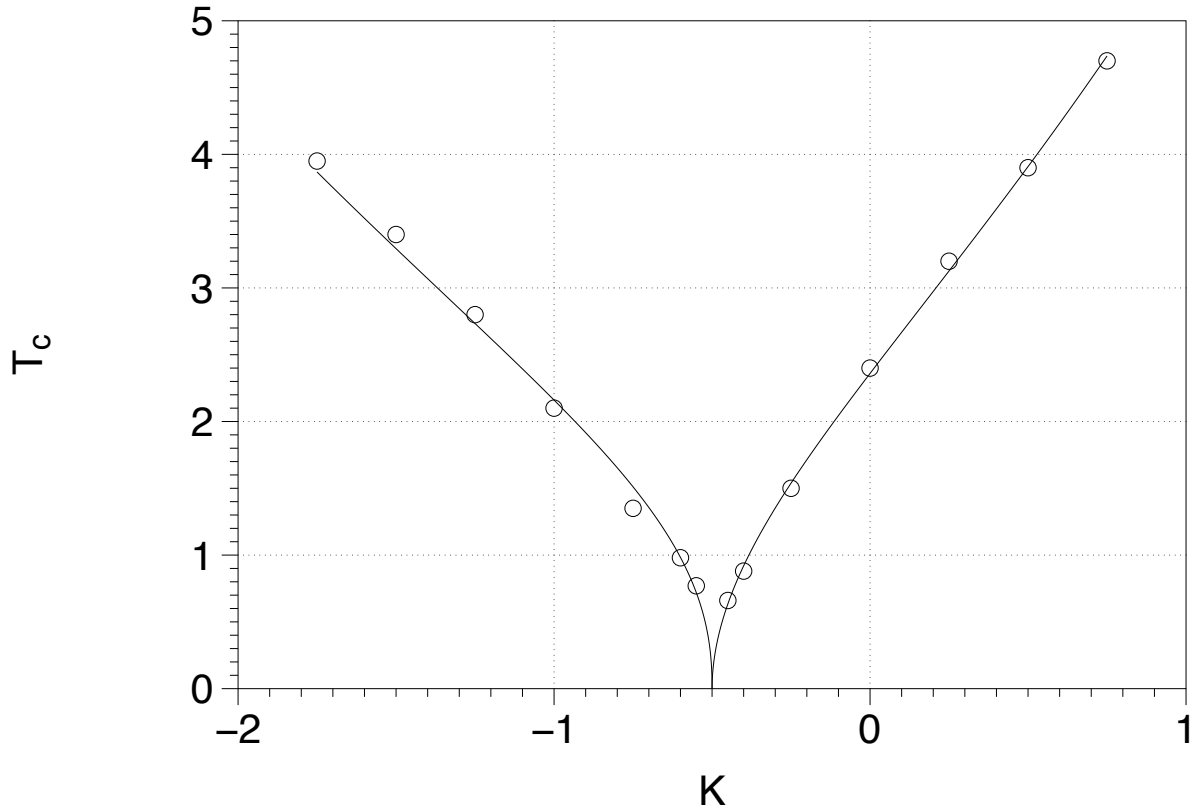


FIG. 7. Phase diagram of the nnn-Ising model. Open circles show the critical temperature for the order disorder transition for the striped phase ($K < -0.5$) and the order disorder transition for the ferromagnetic phase ($K > -0.5$). Solid lines show the asymptotic regression function (Eq. 17)

- [6] G. M. Genkin, Y. N. Nozdrin, A. V. Okomel'kov, and I. D. Tokman, Phys. Rev. B **86**, 024405 1 (2012).
- [7] K. Chesnel, J. Nelson, B. Wilcken, and S. D. Kevan, Journal of Synchrotron Radiation **19**, 293 (2012).
- [8] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 851 (1993).
- [9] S. P. Bromley, J. P. Whitehead, K. De'Bell, and A. B. MacIsaac, J. Magn. Magn. Mater. **264**, 14 (2003).
- [10] H. Westfahl, Jr., J. Schmalian, and P. G. Wolynes, Phys. Rev. B **64**, 174203 (2001).
- [11] A. Giuliani, H. Lieb, and R. Seiringer, Phys. Rev. B **88**, 064401 1 (2013).
- [12] C. Fan and F. Y. Wu, Phys. Rev. **179**, 560 (1969).
- [13] M. P. Nightingale, Phys. Lett. A **59**, 486 (1977).

- [14] J. Oitmaa, *J. Phys. A* **14**, 1159 (1981).
- [15] J. L. Moran-Lopez, F. Aguilera-Granja, and J. M. Sanchez, *Phys. Rev. B* **48**, 3519 (1993).
- [16] J. L. Moran-Lopez, F. Aguilera-Granja, and J. M. Sanchez, *J. Phys. Condens. Matter* **6**, 9759 (1994).
- [17] E. Lopez-Sandoval, J. L. Moran-Lopez, and F. Aguilera-Granja, *Solid State Commun.* **112**, 437 (1999).
- [18] C. Buzano and M. Pretti, *Phys. Rev. B* **56**, 636 (1997).
- [19] A. K. Murtazaev, M. K. Ramazanov, F. A. Kassan-Ogly, and M. K. Badiev, *Journal of Experimental and Theoretical Physics* **117**, 1091 (2013).
- [20] S. Jin, A. Sen, and A. W. Sandvik, *Phys. Rev. Lett.* **108**, 045702 (2012).
- [21] S. Jin, A. Sen, W. Guo, and A. W. Sandvik, *Phys. Rev. B* **87**, 144406 (2013).
- [22] J. C. Phillips, *Rep. Prog. Phys.* **59**, 1133 (1996).
- [23] S. C. Glotzer, N. Jan, T. Lookman, A. B. MacIsaac, and P. H. Poole, *Phys. Rev. E* **57**, 7350 (1998).
- [24] A. O'Hare, F. V. Kusmartsev, K. I. Kugel, and M. S. Laad, *Phys. Rev. B* **76**, 064528 (2007).
- [25] A. O'Hare, F. V. Kusmartsev, and K. I. Kugel, *Acta Phys. Pol. A* **115**, 150 (2009).
- [26] R. Timmons, unpublished (2015).
- [27] C. M. Horowitz, M. A. Bab, M. Mazzini, M. L. R. Puzzo, and G. P. Saracco, *Phys. Rev. E* **92**, 042127 (2015).
- [28] M. S. Holden, M. L. Plummer, I. Saika-Voivod, and B. W. Southern, *Phys. Rev. B* **91**, 224425 (2015).
- [29] M. Maksymenko, V. R. Chandra, and R. Moessner, *Phys. Rev. B* **91**, 184407 (2015).
- [30] T. Shirakura, F. Matsubara, and N. Suzuki, *Phys. Rev. B* **90**, 144410 1 (2014).