Polytype Kinetics and Quenching of Spin Chains with Competing Interactions using Trimer-flip Dynamics

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Abstract

We consider the effects of a quench to $T = 0$ on a spin system with axial next-next nearest neighbour Ising interactions, evolving under a conserved 3-spin flip dynamics. Such a model is motivated by the kinetics of stacking layers in polytypes near the $3C - 6H$ transition. We find that the system generically gets arrested in interesting metastable states which have inhomogeneously distributed quiescent and active regions. In such arrested states, the autocorrelation function decays as a stretched exponential $\sim \exp(-t/\tau_o)^{\frac{1}{3}}$. The latter feature can be understood in terms of a mapping of the dynamics within active stretches to the well known simple exclusion process of particles on a line, and bounds can be put on $\tau_o$.

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1 Introduction

When a statistical system is quenched by suddenly reducing the temperature $T$, we encounter an interesting situation, namely the nonequilibrium evolution of a high-$T$ state in a low-$T$ environment. Two outcomes are possible. The system may approach the equilibrium ground state, perhaps through a coarsening route. Alternatively, it may get arrested in a long-lived nonequilibrium state whose characteristics depend both on the interactions in the system and on gross features of the dynamics, such as conservation laws. In this paper, we examine the consequences of quenching a 1-d Ising spin system with competing interactions, which evolves by a multispin-flip dynamics which conserves an infinite number of quantities. Both the interactions and the kinetics are motivated by the use of the extended axial next nearest neighbour Ising (ANNNI) model to describe polytypism in close-packed crystals, as explained below.

Within this model, we find that the nature of the arrested state obtained by a quench to $T = 0$ depends on the degree of competition in the interactions. In several cases, the state is one in which there are dynamically active and quiescent regions interspersed randomly through the system. Such inhomogeneously active and quiescent (IQA) states have interesting dynamical properties, and are the focus of this paper. In particular, we show that in this state, the autocorrelation function follows a stretched exponential decay law, and that this can be related to the dynamics of the simple exclusion process in finite regions.

2 Polytype Kinetics and the ANNNI Model

Materials such as silicon carbide and zinc sulphide are known to undergo a number of transitions from one close-packed arrangement to another when external conditions such as the temperature and pressure are changed [1]. Each crystalline structure is associated with a particular sequence of stacking close-packed triangular layers; transitions between different polytypes correspond to rearrangements of the stacking sequence.

Let us denote the three possible arrangements of the triangular lattice in a layer by A, B and C [1]. Stacking of layers are then described by sequences such as ABCABC · · · (3C, face-centered cubic), ABCACB · · · (6H, hexagonal close-packed) and ABAB · · · (2H, hexagonal close-packed), each a different polytype. The polytypic constraint of close-packing implies that no two successive layers can be of the same type.

The extended ANNNI model with third neighbour interactions plays an important role in understanding the occurrence of polytypes [2, 3]. The Hamiltonian is

$$\mathcal{H} = -J_1 \sum_i S_i S_{i+1} - J_2 \sum_i S_i S_{i+2} - J_3 \sum_i S_i S_{i+3}$$

(1)

where $J_1$, $J_2$ and $J_3$ are coupling constants, competition between which can lead to the formation of long-period structures. The Ising spins $S_i = \pm 1$ in Eq.(1) are related to successive pairs in the stacking sequence of A’s, B’s and C’s as follows: Pairs AB, BC, CA correspond to $S_i = 1$, while pairs BA, CB, AC correspond to $S_i = -1$. Thus for instance the 3C structure is translated to the spin sequence \textup{↑↑↑ · · ·}, the 6H structure is \textup{↑↑↓↓↓ · · ·} while the 2H structure is \textup{↑↓↑↓ · · ·}. A portion of the ground state phase diagram is shown in
Figure 1: A portion of the ground state phase diagram of the third neighbour ANNNI model. The lines are first order boundaries; thick lines denote multiphase boundaries with an exponentially large number of degenerate ground states.

Fig. 1. The phase boundaries shown all correspond to first order transitions; the heavy lines denote boundaries on which an exponentially large number of ground state configurations are degenerate [4].

Besides providing a description of polytype phases, the extended ANNNI model can also be used to model polytype kinetics. The first such study was concerned with the kinetic effects of quenching across the \( T = 0 \) 2H-6H phase boundary [5], and the nature of the resulting arrested states. Here we study the effects of quenching the model from \( T = \infty \) to \( T = 0 \) in the vicinity of the 3C-6H phase boundary. While modeling kinetics, it is important to allow for only those spin re-arrangements which correspond to physically admissible moves of the stacked layers. While local changes in the stacking sequence correspond to local changes in the spin configuration, the reverse is not necessarily true. For instance, a single spin flip corresponds to a deformation fault which involves the simultaneous movement of a macroscopic number of layers.

Near the 3C-6H transition, there is experimental evidence [6, 7] that the predominant kinetic move is a double-layer displacement which involves the simultaneous movement of two adjacent layers, for example, \( \cdots CABC \cdots \rightarrow \cdots CBAC \cdots \); two successive layers in the sequence can interchange provided that such an interchange does not violate the polytypic constraint of no successive occurrences of the same letter. In spin language, the double-layer-displacement move corresponds to flipping triplets of successive spins, e.g. \( \cdots \uparrow\uparrow\downarrow \downarrow \downarrow \uparrow \rightarrow \uparrow\uparrow\uparrow\uparrow\uparrow\uparrow \rightarrow \uparrow\downarrow\downarrow\uparrow\uparrow \cdots \). This is a generalization of the familiar single-spin-flip Glauber dynamics, but there is an important qualitative difference between the two. While Glauber dynamics in nonconserving, it turns out that triplet flip dynamics leaves invariant an infinite number of conserved quantities [8]. These conservation laws have important consequences for time-dependent properties.
3 Quenching from $T = \infty$ to $T = 0$: the IQA State

Our problem involves studying the effect of quenching to $T = 0$ a system with competing interactions and conservation laws. The dynamics consists of stochastic flips of triplets, and the $T = 0$ environment allows only moves with energy $\Delta E \leq 0$.

Figure 2 is a schematic depiction of the available phase space (i) at $T = \infty$, and (ii) along the $3C - 6H$ multiphase phase boundary at $T = 0$, where any configuration with parallel spin stretches with 3 or more spins is an allowed ground state. At $T = \infty$, it is known \cite{8, 9} that the infinity of conservation laws leads to a partitioning of the $2^L$-large phase space into a large number $N_L$ of sectors; $N_L$ grows as $\sim \mu^L$ where $\mu \simeq 1.618$ is the golden mean. In case (ii), by contrast, using a transfer matrix method we find that the total number of configurations is reduced to $\sim \kappa^L$ with $\kappa \simeq 1.466$. This ground state subspace is further partitioned into $\tilde{N}_L \sim \tilde{\mu}^L$ sectors, with $\tilde{\mu} \simeq 1.167$ as a consequence of the infinitely many conservation laws.

Evolving the system from a random initial configuration, with the $\Delta E \leq 0$ condition enforced, corresponds to a rapid quench from $T = \infty$ to $T = 0$. The energy non-raising condition imposes local constraints on whether or not a pair of chosen spins can actually be flipped; these constraints are a function of $J_1$, $J_2$ and $J_3$. The normalized energy changes $\Delta e \equiv \Delta E/J_1$ involved in flipping a triplet $\uparrow\uparrow\uparrow$ to $\downarrow\downarrow\downarrow$ depend on the environments of the triplet, and are given in Table 1. There are 20 distinct local environments; the other unlisted environments (out of a total of 64 possibilities) are related to these by reflection symmetries.
Table 1: Energy changes for triplet flips in various environments

| Dynamical moves | $\Delta e$ |
|-----------------|------------|
| $\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\uparrow\uparrow\uparrow$ | $4(1 + 2j_2 + 3j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | 0 |
| $\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(1 + 2j_2 + 2j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(1 + j_2 + 2j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(j_2 + 2j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\downarrow\uparrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(j_2 + j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\uparrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4j_3$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(j_2 + j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4j_3$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | 0 |
| $\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(-1 + j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | -4 |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $-4(1 + j_2)$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\downarrow\uparrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(1 + j_2 + j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $4(1 + j_3)$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | 0 |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $-4j_2$ |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | 0 |
| $\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$ $\rightarrow$ $\uparrow\uparrow\downarrow\downarrow\downarrow\uparrow\uparrow$ | $-4(1 + 2j_2 + j_3)$ |

For all values of $j_2 = J_2/J_1$ and $j_3 = J_3/J_1$ for which the $\Delta e$’s listed in Table 1 have the same sign, the system will reach an arrested state of the same character.

We used numerical simulations to investigate the properties of arrested states obtained by quenching to points on the $3C - 6H$ boundary, and in the $3C$ phase (points $P$ and $Q$ in Fig. 1, respectively). In both cases, the quench does not lead to the ground state. For instance, at point $P$, the system does not approach the subspace depicted in Fig. 2, but rather an arrested state which is infinitely long-lived at $T = 0$. Figure 3 gives an idea of the nature of this arrested state. The state has alternating quiescent and active stretches of random lengths. The lengths of active stretches have an exponential distribution, $A_P(\ell) \sim \exp(-\lambda_P\ell)$ with $\lambda_P \simeq 0.22$. We monitored the decay of the spin autocorrelation function $C_P(t)$ in this state (Fig. 4), and found that it decays as a stretched exponential $\sim \exp(-(t/\tau_P)^{1/3})$. We repeated the quench for a point $Q$ (Fig. 1) in the $3C$ phase, away from the multiphase line. The equilibrium ground state is $\uparrow\uparrow\uparrow\uparrow\cdots$ in the case $Q$, in contrast to the large degeneracy of ground states at $P$. Nevertheless, on quenching towards $Q$, we found again an IQA state with an exponentially distributed number of active stretches, $A_Q(\ell) \sim \exp(-\lambda_Q\ell)$ with $\lambda_Q \simeq 0.13$. Further, the autocorrelation function $C_Q(t)$ decays as $\sim \exp(-(t/\tau_Q)^{1/3})$, but with $\tau_Q > \tau_P$ (Fig. 4). A study of the nature of the excitations in the active stretches in the two cases $P$ and $Q$ shows that they are quite different in character. Despite this, as argued below, they show the common feature of diffusing excitations confined to randomly distributed stretches of finite lengths, and this leads to the same form of stretched exponential decay of $C(t)$ in both cases.
Figure 3: The space-time diagram of an IQA arrested state (left) with activity depicted in white. The activity pattern in a single active stretch of the IQA state (right) shows the diffusive motion of active walkers, with hard core interactions.

Quench to P ($j_2 = 0, j_3 = -1/3$, on $3C - 6H$ boundary): In this case, the active regions consist of parallel spin patches (PSPs) in a background which is one of two types: (a) antiparallel doublets $\uparrow\uparrow\uparrow\downarrow\downarrow \cdots$ (b) antiparallel doublets with some $\uparrow\downarrow$ pairs interspersed between doublets, e.g. $\uparrow\uparrow\downarrow\uparrow\uparrow \cdots$. Flipping a triplet at the edge of a PSP is an allowed ($\Delta E = 0$) move, and results in a translation of the PSP by either one or two lattice spacings, e.g. $\uparrow\uparrow\downarrow\uparrow\uparrow\uparrow\uparrow\downarrow\downarrow \rightarrow \uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow\downarrow$. In the process, the size of the PSP may fluctuate between 4 and 5, depending on the surroundings, e.g. $\uparrow\uparrow\uparrow\downarrow\downarrow\downarrow\downarrow\uparrow\uparrow \rightarrow \uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow\downarrow$. Backgrounds of type (b) can support only a single PSP, as two PSPs around an $\uparrow\downarrow$ pair lead to a frozen structure. By contrast, backgrounds of type (a) can support several mobile PSPs. In this case, two PSPs have a hard core repulsion and cannot pass through each other. These statements can be easily checked using Table 1, since $\Delta E > 0$ moves are disallowed. Thus we have a gas of extended objects with hard core interactions, diffusing in a confined region. The boundaries of quiescent regions have configurations only of a few types — for example, one such left-boundary is $\uparrow\uparrow\downarrow\downarrow\downarrow \cdots$, which is stable with respect to the adjacent active stretches described above (again specific moves can be checked using Table 1).

Quench to Q ($j_2 = 1, j_3 = 0$, in $3C$ phase): In this case, active regions consist of stretches of ground state configurations, separated by domain walls, e.g. $\cdots \uparrow\uparrow\uparrow\uparrow \downarrow\downarrow\downarrow \downarrow \uparrow\uparrow\uparrow \cdots$. The spacing between successive domain walls is $(3m + 2)$ with $m = 0, 1, 2, \ldots$. A triplet adjacent to a wall can flip, whereupon the wall moves by three lattice spacings. Two diffusing walls exhibit a short range repulsive interaction, as they can approach no closer than two lattice spacings. Once again, the dynamics is tantamount to a gas of objects with hard core repulsion diffusing in a confined region. The quiescent boundaries in this case are also of a
few types, e.g. a stable left-boundary of a quiescent region is \( \uparrow \uparrow \downarrow \uparrow \uparrow \) \( \ldots \); it can be checked that any attempt to flip its edge spins leads to energy raising.

In the active stretches in both cases \( P \) and \( Q \), we have diffusing walkers with hard core repulsion confined to finite regions between impenetrable walls. Let the number density of such walkers in any active stretch be \( \tilde{\rho} \). The autocorrelation function \( C^X_{\ell}(\tilde{\rho}, t) \) (where \( X \) stands for either \( P \) or \( Q \)), of such a stretch is expected to decay at large times as \( \exp(-t/\tau_X) \). Here \( \tau_X \) is the relaxation time which depends on the size \( \ell \) of the finite stretch. A crucial point to note is that the diffusing walkers are extended objects — the PSPs in case \( P \) have finite size, and the domain walls in case \( Q \) have finite range of repulsion between them. In spite of this, in both cases the dynamics can be mapped exactly to the well known simple exclusion process (SEP) \(^{10}\) of particles on a line of a reduced length \( \ell_X = r_X \ell' \). Here \( \ell' \) differs slightly from \( \ell \), and is defined below.

In case \( P \), for a background of type (a), every PSP is mapped onto a single particle, while a doublet is mapped onto a hole (\( \uparrow \uparrow \downarrow \uparrow \uparrow \uparrow \uparrow \uparrow \downarrow \uparrow \uparrow \downarrow \uparrow \rightarrow \circ \circ \bullet \circ \bullet \bullet \circ \bullet \) leading to \( r_X = (1 - 3\rho)/2 \). In the case of backgrounds of type (b), which have only a single PSP, \( \ell_X \) depends
on the number of $\uparrow \downarrow$ pairs present; if the maximum number of such pairs are allowed (e.g. $\uparrow \uparrow \uparrow \downarrow \downarrow \uparrow \uparrow \uparrow \uparrow \uparrow \cdots$), $r_X = 3/4$, as every doublet and a single spin gets mapped to a hole. In case $Q$, every parallel triplet is replaced by a hole and an $\uparrow \downarrow$ pair associated with a domain wall is replaced by a particle ($\uparrow \uparrow \uparrow \uparrow \downarrow \downarrow \downarrow \downarrow \uparrow \uparrow \uparrow \rightarrow \circ \circ \circ \circ \circ \circ \circ$) leading to $r_X = (1+\rho)/3$. The modified length $\ell'$ is $(\ell + 4)$ and $(\ell + 2)$ in the cases $P$ and $Q$ respectively. The difference between $\ell$ and $\ell'$ arises from consideration of the dynamics of the walkers near the active-quiescent boundaries; since $\ell' \simeq \ell$ for large $\ell$, we have $\rho = \tilde{\rho} / \ell' \simeq \tilde{\rho}$.

For the mapped SEP with free boundary conditions, one knows that the relaxation time $= \ell_X^2/\pi^2 D$ where $D = 1/2$ is the diffusion constant of the SEP. Hence in our problem $\tau_X = 2r^*_X \ell^2/\pi^2$. The full autocorrelation function $C^X(t)$ is then given by averaging over the distribution $A(\ell, \rho)$ of stretches of length $\ell$ and walker density $\rho$.

$$C^X(t) = \sum_{\ell, \rho} A(\ell, \rho) C^X(\rho, t)$$  \hspace{1cm} (2)

To make further progress, note that the largest and the smallest values of $\tau_X$ (as $\rho$ is varied from $1/\ell'$ to its maximum possible value) provide bounds on $C^X(\rho, t)$, and thereby on $C^X(t)$. Using $A(\ell) \sim \exp(-\lambda_X \ell)$ and $\tau_X = 2r^*_X \ell^2/\pi^2$, (where $r^*_X$ is a different constant for the minimum and maximum densities), we see that the above sum is dominated by a saddle point value $\ell'^* = (\pi^2 t/\lambda_X r^*_X)^\frac{1}{2}$. This leads to a stretched exponential form for both the upper and lower bounds, implying that $C^X(t) \sim \exp(-(t/\tau^X_o)^\frac{1}{2})$. In case $P$, the lower and upper bounds on $\tau^P_o$ are $(1/5)^2 (8/(27\pi^2 \lambda^2_P))$ (corresponding to $\rho = 1/5$) and $8/(27\pi^2 \lambda^2_P)$ (corresponding to $\rho = 0$). In case $Q$, they are $(1/2)^2 (8/(27\pi^2 \lambda^2_Q))$ (corresponding to $\rho = 1/2$) and $8/(27\pi^2 \lambda^2_Q)$ (corresponding to $\rho = 0$). Inserting numerical values $\lambda_P = 0.22$ and $\lambda_Q = 0.13$, we have $0.024 \leq \tau^P_o \leq 0.61$ and $0.44 \leq \tau^Q_o \leq 1.75$. This helps us to understand the reason for the numerically observed slower decay ($\tau^P_o = 0.37$ and $\tau^Q_o = 0.56$ in Fig. 4) of $C^Q(t)$ compared to $C^P(t)$.

4 Conclusion

In our study of $T = \infty \rightarrow T = 0$ quenches of the extended ANNNI model evolving under triplet flip dynamics, we found interesting arrested states with interspersed quiescent and active regions. Although the character of excitations is quite different in the two cases studied, the autocorrelation function shows similar stretched exponential decays. This was traced to the fact that active regions in both cases carry diffusing excitations, and the dynamics in both cases can be mapped on to the simple exclusion process.

We have found and studied IQA states in quenches of other models including the ANNNI model with Kawasaki dynamics. In this case, in addition to the stretched exponential decay of $C(t)$ in steady state, we found an interesting 2-step relaxation towards equilibrium when the temperature is raised slightly \((\uparrow \uparrow \downarrow \uparrow \downarrow \downarrow \uparrow \downarrow \uparrow \uparrow \uparrow \cdots)\).

It would be interesting to try to identify signatures of IQA states in rapid quenches of polytypic materials, for instance near the $3C - 6H$ phase boundary.

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