Control of Structure Formation in Phase Separation Simulations

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1 Abstract

A homogeneous binary mixture becomes thermodynamically unstable when it is quenched below the demixing (critical) temperature. Subsequent evolution of the mixture is characterized by the emergence and growth of domains enriched in either component. We simulated this using the conserved-order-parameter (COP) Ising model with Kawasaki spin-exchange dynamics, on a two-dimensional lattice. The structure of such a system can be quantitatively described by the so-called structure factor $S(k)$; which can be found from a Fourier transform of the lattice. Usually, a COP Ising simulation is performed at a constant temperature after the quench. This results in a circularly-averaged structure factor $S(k)$ showing a single peak, the wave vector of which corresponds to the typical domain size.

We have confirmed the known fact that in a single-quench experiment, for large values of $k$, $S(k)$ follows Porod’s law: $S(k) \sim k^{-(d+1)}$, with $d = 2$; the dimension our system. Furthermore, we have confirmed with great accuracy the growth law $R(t) = a + bt^{\frac{1}{3}}$ for the domain size $R$.

The main goal of our study is to let the system evolve to arbitrarily predefined structure factors, by time variation of external parameters. This is a form of self-organized structure formation; there is no direct manipulation of the particles. We focussed on a specific case where we vary the temperature to create a circularly-averaged structure factor $S(k)$ that shows two equally high peaks, a so-called “camel bump”. The second peak is formed at a larger value of $k$ (corresponding to a smaller domain size) by the following double-quench process:

- The system starts with a homogeneous binary mixture and is evolved for a relatively long time at a constant temperature below the critical temperature (single quench).
  (e.g. we simulated for $10^5$ MCS at $T = 0.7T_c$.)

- The system is heated for a very short time at a constant temperature much larger than the critical temperature.
  (e.g. we heated for 150 MCS at infinite temperature, i.e. $\beta = \frac{1}{kT} = 0$.)

- The system is evolved again at a constant temperature below the critical temperature (second quench), but for a much shorter time than the first quench.
  (e.g. we simulated for $10^3$ MCS at $T = 0.7T_c$.)

During heating at infinite temperature, the particles in our simulation make a random walk. The average displacement vector of a particle accurately follows a two-dimensional isotropic Gaussian distribution with $\sigma = \sqrt{2t}$, where $t$ is the time measured in Monte Carlo Steps (MCS). The evolution of the connected correlation function $g(x, y)$ can be accurately predicted by convoluting it with this Gaussian (while manually correcting for $g(0, 0) = 1$), i.e. the heating process acts on $g(x, y)$ like a Gaussian low-pass filter that smoothes the function by reducing high frequencies in its Fourier transform $S(k_x, k_y)$. Therefore the prediction of the structure factor $S(k_x, k_y)$ can be efficiently calculated by multiplying it with the Fourier transform of this Gaussian, and adding a term to correct for the fact that we manually put $g(0, 0) = 1$.

During heating, particles on or near a domain border can easily leave their own domain and diffuse into another domain. On a sub-domain scale (large $k$) the density of the other type of particles changes to a nonzero value $\rho_{\text{eff}}$, which goes increasingly towards $\frac{1}{2}$ when the heating proceeds. When the heating does not last for a too long time, the larger-scale structure (small $k$) of the domains is still present in $S(k)$ (but the peak is lower), while the large-$k$ part of $S(k)$ looks exactly like the one of a randomly mixed system at density $\rho_{\text{eff}}$.

Indeed we have shown that, after the second quench, the new peak that emerges at large values of $k$ grows exactly like the one in a single-quench simulation that starts with a homogeneous mixture at density $\rho_{\text{eff}}$ and evolves at the same temperature. (This is true until its position gets close to the position of the first peak.)
2 Introduction

In this thesis we will study phase separation of a binary mixture. We are specifically interested in the shape of the structure factor (see section 3.6 for its definition). To study the phase separation, we will use computer simulations of the conserved-order-parameter (COP) Ising Model on a two-dimensional square lattice (with Kawasaki spin-exchange dynamics and Metropolis acceptance probability). The standard way to simulate this is to start with a randomly mixed state (high temperature) and then to proceed at a constant heat-bath temperature below the demixing (critical) temperature. This drop in temperature is often called a quench. Subsequent evolution of the mixture is characterized by the emergence and growth of domains enriched in either component, see Fig. 1. From these kinds of simulations performed by others, but also by experiments and theoretical work, the general form and time dependence of the structure factor for such a system is known. Fig. 2 shows a typical structure factor as seen in our simulations.

Our general goal is to be able to generate an arbitrary predefined structure factor, by time variation of external parameters. So contrary to most studies we will change external parameters such as the temperature during the course of a simulation to influence the shape of the structure factor. In a standard simulation at fixed temperature, the structure factor $S(k)$ will have one
Figure 3: The birth of a camel bump. The first snapshot is the same as the third in Fig. 1. From here the temperature is changed to $T = \infty$ ($\beta = \frac{1}{kT} = 0$) for only 150 MCS (second snapshot), after which the system is quenched at a temperature of $T = 0.7T_{\text{critical}}$ for $10^3$ MCS (third snapshot).

Figure 4: Structure factor corresponding to the third snapshot in Fig. 3.

peak (i.e. one local maximum away from $k = 0$). From the infinite number of possible structure factors, we concentrated on one specific case: a structure factor with two local maxima in $S(k)$, with arbitrary positions $k_{\text{max,1}}$ and $k_{\text{max,2}}$. We will refer to such a structure as “camel bump”. We have indeed succeeded in creating a camel bump by changing the temperature during the course of a simulation. Snapshots of some important times during the simulation are shown in Fig. 3. The structure factor corresponding to the final snapshot is shown in Fig. 4.

The forming of domains is a form of self-organized structure formation: the structure is built up by a thermodynamical process; without direct manipulating of the particles. Using an external parameter like temperature to control the formation of structure is much easier than direct manipulating on a small scale.
3 Theory

In this section, some theory about the (ordinary and conserved-order-parameter) Ising model will be presented. Readers who are already familiar with this topic can skip this section and go directly to the presentation of results in section 4.

3.1 Ising Model

The Ising model is a simple model of a magnet. It is composed of particles on a lattice. The lattice can be of any dimension and shape, but it is usually a square (two-dimensional) or cubic (three-dimensional) lattice. In our study we restrict ourselves to a square lattice, because this way our simulations will be faster to perform than by taking a three-dimensional lattice.

Each particle \(i\) on the lattice has a spin \(s_i\) which can only take the values \(s_i = \pm 1\). Spins interact, giving rise to products \(s_is_j\) in the Hamiltonian (energy function). In real magnets these interactions will be strongest for particles close to each other and weaker for particles further apart. We will simplify this by only taking into account interactions between nearest neighbors on the lattice, and assuming that all interactions have the same strength \(J\). If we also include an external magnetic field \(B\) coupling to the spins, the Hamiltonian is:

\[
H = -J \sum_{\langle ij \rangle} s_is_j - B \sum_i s_i. \tag{1}
\]

Here \(\langle ij \rangle\) denotes that the spins \(i\) and \(j\) are nearest neighbors. (In a square lattice, a spin \(i\) has 4 nearest neighbors; i.e. neighbors up, down, left and right.) Because we wish to avoid possibly complicating boundary effects we make use of periodic boundary conditions; spins on the border of the lattice are neighbors of spins on the opposing border.

With a positive value of \(J\), aligned spins lower the total energy and anti-aligned spins raise the total energy. Thus the spins will have the tendency to align. They will also want to be positive if the external field \(B\) is positive, and negative if \(B\) is negative.

The total magnetization \(M\) of our system is simply the sum of all spins \(s_i\). This is an extensive variable; it scales with the size of our system. It is therefore often more convenient to look at the mean magnetization per lattice site \(m = \frac{M}{N}\), where \(N\) is the number of sites. This value does not scale with the size of the system (intensive) and always takes values between \(-1\) (all spins oriented downwards) and 1 (all spins oriented upwards).

3.2 Equilibrium and Boltzmann factors

We assume that our system is surrounded by a thermal reservoir or heat-bath that has a temperature \(T\). This pushes the temperature of the system towards the temperature of the reservoir.

Our system can take several spin configurations or states. If our system reaches equilibrium, the probability to find the system in a particular state \(\mu\) is given by its Boltzmann factor

\[
p_{\mu} = \frac{1}{Z} e^{-E_\mu/kT} = \frac{1}{Z} e^{-\beta E_\mu}. \tag{2}
\]

We write \(\beta = \frac{1}{kT}\), where \(k\) is Boltzmann’s constant (1.38 \times 10^{-23} \text{ JK}^{-1})\. \(E_\mu\) is the energy of the system in state \(\mu\), which can be calculated from the Hamiltonian (as given in Eq. (1)). \(Z\) is the partition sum, it is given by

\[
Z = \sum_{\mu} e^{-\beta E_\mu}.
\]

It is a normalizing constant which ensures that \(\sum_{\mu} p_{\mu} = 1\). This is needed because the probability to find the system in one out of all the possible states must be equal to 1.
If a system is in state $\mu$, there is a transition probability $P(\mu \to \nu)$ that the system goes to another state $\nu$. A Monte Carlo Ising algorithm simulates this by selecting at random one of the possible next states $\nu$ and then accepting this state with a probability that is proportional to the probability $P(\mu \to \nu)$. In order to do this the computer program uses a random function, hence the name Monte Carlo algorithm.

The system typically starts in a state far from equilibrium by having a random configuration of spins (high temperature of the system itself), and then running the simulation at constant heat-bath temperature $T$. This sudden temperature drop is called a quench.

The transition probabilities are determined by the dynamics of the system. We are not entirely free to choose them, because the probabilities must correctly sample the Boltzmann distribution. This gives two restrictions on the transition probabilities, as explained below.

Since the probability to find a system in any state $\nu$ is nonzero, there must be a way to go from any given state $\mu$ to state $\nu$ through a chain of states $\mu_i$ so that all $P(\mu_i \to \mu_{i+1})$ are nonzero. This is called the condition of ergodicity.

When the system is at equilibrium, the rate at which transitions into and out of a given state $\mu$ are taking place must be equal, so

$$\sum_\nu p_\mu P(\mu \to \nu) = \sum_\nu p_\nu P(\nu \to \mu), \quad (3)$$

One way to satisfy this equation is to impose that

$$p_\mu P(\mu \to \nu) = p_\nu P(\nu \to \mu),$$

for all pairs of states $\mu$ and $\nu$.

The above condition is called detailed balance.

So in order for our system to reach an equilibrium with detailed balance and a Boltzmann distribution, the transition probabilities must satisfy

$$\frac{P(\mu \to \nu)}{P(\nu \to \mu)} = \frac{p_\nu}{p_\mu} = e^{-\beta(E_\nu - E_\mu)}. \quad (4)$$

This still leaves a lot of space on how to choose the transition probabilities, i.e. how we choose the dynamics of the system.

### 3.3 Metropolis Algorithm

We shall now describe the most commonly used algorithm to simulate the Ising model. For simplicity we shall take $B = 0$. A spin $s_k$ is selected at random from the lattice (that is with probability $\frac{1}{N}$), and is proposed to be flipped so that $s_k \to -s_k$. The difference $E_\nu - E_\mu$ in energy between the state $\mu$ before and the state $\nu$ after the spin flip can be calculated from Eq. (1). Because almost no spins change in going to the new state there is a more efficient way to calculate the energy difference. We write the Hamiltonian for $E_\mu$, where we explicitly take out the terms that involve the spin $k$:

$$E_\mu = -J \sum_{\langle ij \rangle \in \{i,j\}} s_\mu^i s_\mu^j - J s_\mu^k \sum_{\langle ik \rangle \in \{i,k\}} s_\mu^i.$$

The equation for $E_\nu$ is exactly the same if we replace $\mu$ by $\nu$. Furthermore we can use that $s_\nu^k = -s_\mu^k$ and $s_\nu^i = s_\mu^i$ for $i$ not equal to $k$, so

$$E_\nu = -J \sum_{\langle ij \rangle \in \{i,j\} \setminus \{k\}} s_\mu^i s_\mu^j + J s_\mu^k \sum_{\langle ik \rangle \in \{i,k\}} s_\mu^i.$$

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So the energy difference is
\[ E_\nu - E_\mu = 2J \sum_{\langle ik \rangle} s_i^\mu. \tag{5} \]

This sum runs only over the nearest neighbors of the spin \( s_k \) that is selected to be flipped. Note also that this expression only depends on the situation in our initial state \( \mu \) so we can calculate the energy difference without having to carry out the spin flip.

With these dynamics, only the transition between states that differ by the flip of a single spin will have a nonzero transition probability. Note though, that the condition of ergodicity is satisfied, because you can reach any state \( \nu \) from a given state \( \mu \) by subsequently flipping every spin that occurs in state \( \nu \) if it has an opposite sign to the spin in state \( \mu \).

We must now determine the probability \( A(\mu \to \nu) \) for which we accept the proposed move. The transition probabilities must satisfy the condition of detailed balance as given in Eq. (4), so we must have
\[ \frac{A(\mu \to \nu)}{A(\nu \to \mu)} = \frac{P(\mu \to \nu)}{P(\nu \to \mu)} = e^{-\beta (E_\nu - E_\mu)}. \tag{6} \]

Because we do not wish to waste more of our computer time than needed by rejecting proposed moves, we would like to maximize the acceptance probability while still satisfying the condition for detailed balance. Of course a probability cannot be larger than 1. Suppose that \( E_\nu - E_\mu \leq 0 \). In this case the right hand side of the equation is larger than or equal to 1, so we can take \( A(\mu \to \nu) = 1 \). Then we must have
\[ A(\nu \to \mu) = e^{-\beta (E_\mu - E_\nu)}. \tag{7} \]

Now if \( E_\nu - E_\mu > 0 \), the right hand side of Eq. (6) is smaller than 1, and the maximum value we can get for \( A(\mu \to \nu) \) is when \( A(\nu \to \mu) = 1 \), in which case we have to take \( A(\mu \to \nu) = e^{-\beta (E_\mu - E_\nu)} \). This is consistent with Eq. (7) where \( E_\mu - E_\nu \geq 0 \). This means that we have indeed found the maximum possible acceptance probability. We arrived at the so-called Metropolis acceptance probability:
\[ A(\mu \to \nu) = \begin{cases} 1 & \text{if } E_\nu - E_\mu > 0, \\ e^{-\beta (E_\mu - E_\nu)} & \text{otherwise}. \end{cases} \tag{8} \]

We see that, if the proposed spin flip would lower the total energy, the move is always accepted. If, however, it raises the energy, the move is accepted with a probability proportional to a Boltzmann factor. Note that this probability to accept a move that increases the energy is high at higher temperatures (more thermal fluctuations) and low at lower temperatures. So when we simulate the system at a low temperature, the spins will tend to align, causing domains of spins pointing in the same direction, that grow in size during the run of a simulation, until at equilibrium the absolute value of the total magnetization is (very close to) 1. However, at high temperatures, the acceptance probability gets close to 1, even if a proposed move increases the total energy in the system. This means that there is not much domain growth, and the total magnetization will be (close to) 0. Somewhere in between these low and high temperatures lies the critical temperature. Below this temperature, the size of the domains keeps increasing with time. Above the critical temperature there is a very limited domain formation; the domains are small and do not keep on growing with time.

### 3.4 The Conserved-Order-Parameter Ising Model

Mathematically, the conserved-order-parameter Ising Model (COP) is very similar to the ordinary Ising model, but both the dynamics and the physical interpretation differ. It is a model for either a gas or a mixture of two different kinds of particles (we will call them type A and
B) on a lattice. Every lattice site can have one of two values $\sigma_i$. A value of $\sigma_i = 1$ corresponds to a lattice site that is occupied by a particle (type A) and a value of $\sigma_i = 0$ corresponds to a vacant lattice site (or a particle of type B). The total number of particles (of type A) is given by

$$\sum_i \sigma_i = \rho N. \quad (9)$$

Here $N$ is the total number of sites and $\rho$ is the density of particles. We assume that no particles can leave the system (or change into another particle), so the density $\rho$ has a constant value. Also note that each lattice point can be occupied by at most one particle ($\sigma_i$ is either 0 or 1). This can be seen as a hard-sphere repulsion caused by Pauli exclusion.

We shall now determine the Hamiltonian. For this we only take into account the attractive force between two particles that are nearest neighbors. If they have an attraction with energy $\epsilon$ the Hamiltonian becomes:

$$H = -\epsilon \sum_{\langle ij \rangle} \sigma_i \sigma_j.$$

Now this Hamiltonian is closely related to the one of the Ising model, given in Eq. (1), in the absence of a magnetic field ($B = 0$). To see this, we connect the values 0 and 1 that $\sigma_i$ can take to the spin values $-1$ and $+1$ that $s_i$ can take by the relation:

$$s_i = 2\sigma_i - 1.$$

Now a “spin” of $s_i = -1$ corresponds to a vacant site (or particle of type B) and a value of $s_i = 1$ to an occupied site. We can rewrite the Hamiltonian with these variables:

$$H = -\frac{\epsilon}{4} \sum_{\langle ij \rangle} (s_i + 1) (s_j + 1)$$

$$= -\frac{1}{4} \epsilon \sum_{\langle ij \rangle} s_i s_j - \frac{1}{2} z \epsilon N,$$

where $z$ is the number of nearest neighbors of a site.

We can also rewrite Eq. (9) using the same change of variables. This gives

$$\sum_i s_i = (2\rho - 1)N. \quad (11)$$

So a density of $\rho = 1$ corresponds to a total “magnetization” per site of $m = 1$, a density of $\rho = 0$ corresponds to magnetization $m = -1$.

We can use this equation to simplify the Hamiltonian in terms of the variables $s_i$:

$$H = -\frac{1}{4} \epsilon \sum_{\langle ij \rangle} s_i s_j - z \epsilon N \rho.$$

So if we define $J = \frac{1}{4} \epsilon$ we get the form

$$H = -J \sum_{\langle ij \rangle} s_i s_j + \text{constant}. \quad (12)$$

The constant term does not affect the expectation values of observable quantities, so we see that in fact the Hamiltonian behind the COP Ising model is the same as the one behind the ordinary Ising model (Eq. (1)). The total magnetization $M = \sum_i s_i$ in the ordinary Ising model is called the order parameter. In the conserved-order-parameter Ising model this is conserved (Eq. (11)), hence the name.
3.5 Kawasaki Dynamics

Obviously the dynamics of the COP Ising model cannot consist of spin flips as this would break conservation of the total magnetization. A more logical dynamics would be to exchange the spins of two neighbors. A particle can move to an adjacent vacant spot (or in the two-component mixture point of view, particles of type \( A \) and \( B \) exchange their places). This dynamics is called Kawasaki dynamics.

We can still use the Metropolis acceptance probability as given in Eq. (8). The energy difference \( E_\nu - E_\mu \) can now take on different values than in the nonconserved case. We derive an equation for the energy difference which uses the fact that, for a single spin exchange, most spins remain unchanged, so that we do not have to sum over all spins to calculate it. Let \( k \) and \( k' \) be the two nearest neighbor spins we propose to exchange, then

\[
E_\nu - E_\mu = 2J \left( s_k^\mu \sum_{\langle ik \rangle} s_i^\mu s_k^\nu + s_k^\nu \sum_{\langle jk' \rangle} s_j^\mu \right), \tag{13}
\]

when the spins are of opposite sign, and \( E_\nu - E_\mu = 0 \) when the spins have the same sign.

In order to prove this, we first write the expression for \( E_\mu \); for this we use the Hamiltonian of Eq. (12):

\[
E_\mu = -J \sum_{\langle ij \rangle} s_i^\mu s_j^\mu + \text{constant}.
\]

This can be split into terms that do not change as the state goes from \( \mu \) to \( \nu \) and terms that do change, as follows:

\[
E_\mu = \text{constant} - J \sum_{\langle ij \rangle \cap \{k,k'\} = \emptyset} s_i^\mu s_j^\mu - J \sum_{\langle ik \rangle} s_i^\mu s_k^\nu + J \sum_{\langle jk' \rangle} s_j^\mu s_k^\nu - J s_i^\mu s_j^\nu.
\]

For \( E_\nu \) we can write exactly the same expression with \( \mu \) replaced by \( \nu \). Because the spins \( k \) and \( k' \) are switched to give the state \( \nu \) we must have \( s_k^\nu = s_k^\nu \) and \( s_{k'}^\nu = s_{k'}^\nu \). Furthermore, for all \( i \) not equal to \( k \) or \( k' \) we have \( s_i^\nu = s_i^\nu \). With this we can write \( E_\nu \) entirely in terms of the spins in state \( \mu \):

\[
E_\nu = \text{constant} - J \sum_{\langle ij \rangle \cap \{k,k'\} = \emptyset} s_i^\nu s_j^\nu - J \sum_{\langle ik \rangle} s_i^\nu s_k^\nu + J \sum_{\langle jk' \rangle} s_j^\nu s_k^\nu - J s_i^\nu s_j^\nu.
\]

so

\[
E_\nu - E_\mu = J \left[ s_k^\mu \left( -\sum_{\langle jk' \rangle} s_j^\mu + \sum_{\langle ik \rangle} s_i^\mu \right) + s_k^\nu \left( -\sum_{\langle ik \rangle} s_i^\mu + \sum_{\langle jk' \rangle} s_j^\mu \right) \right].
\]

The two terms between parentheses are exactly each others opposite, so when \( s_k = s_{k'} \) the two terms cancel each other and the energy difference is 0. In case \( s_k \) and \( s_{k'} \) have different signs, the terms sum up to give exactly Eq. (13).

Note that the right-hand side of Eq. (13) only depends on the initial state \( \mu \) and not on the final state \( \nu \). (As is the case for the ordinary Ising model, see Eq. (5).) This means we can calculate the energy difference in advance; without making the actual move.

In a single step, the algorithm selects a pair of nearest-neighbor spins and then accepts or rejects this move with Metropolis acceptance probability. This process is repeated for many steps. A good time scale should satisfy that after a certain amount of time, on average each spin
pair is proposed to be exchanged the same amount of times, regardless of the number of spins \( N \) in the system. So a larger system requires more steps to reach the same time. The standard simulation time is the number of Monte Carlo steps (MCS). One Monte Carlo step is reached after every step in the algorithm is performed \( N \) times, regardless whether the next move is accepted or rejected. So, statistically, in one Monte Carlo step, each spin pair is proposed to be exchanged once.

### 3.6 Structure Factor

A quantity which gives a lot of information about the structure of the system is the two-point connected correlation function. It can be calculated for both the conserved-order-parameter model and the ordinary Ising model.

\[
G^{(2)}_c (i, j) \equiv \langle s_i s_j \rangle - \langle s_i \rangle \langle s_j \rangle = \langle s_i s_j \rangle - m^2. \tag{14}
\]

This is an average over multiple systems, or for systems that have reached equilibrium it can also mean an average over time. The average value of the spin at a site is equal to \( m \), the magnetization per spin. Because the systems we are modeling are translationally invariant, the correlation function \( G^{(2)}_c (i, j) \) depends only on the displacement vector \( r \) between lattice points \( i \) and \( j \), but not on the exact position of the points. So if \( r_i \) is the position vector of spin \( i \), we have

\[
G^{(2)}_c (r) \equiv G^{(2)}_c (r_i, r_i + r),
\]

which does not depend on the position \( r_i \). If we wish to calculate \( G^{(2)}_c (r) \) we can get a better estimate by averaging over all \( i \) and \( j \) with displacement \( r \), instead of taking only one pair of spins:

\[
G^{(2)}_c (r) = \frac{1}{N} \sum_{i,j \text{ with } r_j - r_i = r} \left( \langle s_i s_j \rangle - m^2 \right).
\]

Note that this function is periodic if we take a lattice with periodic boundary conditions. It is possible to calculate directly the function \( G^{(2)}_c (r) \) for all possible \( r \) on our lattice during a simulation, but this can take a considerable amount of time, because the number of steps to calculate it scales with the number of lattice points \( N \) as \( N^2 \). There is, however, a quicker way to calculate the correlation function by using the spatial discrete Fourier transform. First note that

\[
\langle s_i s_j \rangle - m^2 = \langle s_i s_j \rangle - \langle ms_i \rangle - \langle ms_j \rangle + m^2 = \langle (s_i - m)(s_j - m) \rangle,
\]

so

\[
\tilde{G}^{(2)}_c (k) = \sum_r e^{ik \cdot r} G^{(2)}_c (r)
\]

\[
= \frac{1}{N} \sum_r \sum_{i,j \text{ with } r_j - r_i = r} e^{ik \cdot r} \left( \langle s_i s_j \rangle - m^2 \right)
\]

\[
= \frac{1}{N} \left\langle \sum_{r_i} e^{-ik \cdot r_i} (s_i - m) \sum_{r_j} e^{-ik \cdot r_j} (s_j - m) \right\rangle
\]

\[
= \frac{1}{N} \left\langle \left| \tilde{s}' (k) \right|^2 \right\rangle,
\]

where \( \tilde{s}' (k) \) is the Fourier transform of \( s'_i \equiv s_i - m \). So the Fourier transform of the correlation function can be calculated by taking the absolute value squared of the Fourier transform of
the spins of the lattice and averaging this. Now to get the correlation function \( G^{(2)}_c(\mathbf{r}) \) itself, we simply perform an inverse Fourier transform. Because discrete Fourier transforms can be calculated using the Fast Fourier Transform (FFT) algorithm, which has a computational cost of order \( N \log(N) \), this method of calculating the correlation function is faster (provided the lattice is not very small).

The Fourier transform of the correlation function, the quantity

\[
S(k) \equiv \tilde{G}^{(2)}_c(k) = \frac{1}{N} \left\langle |\tilde{s}'(k)|^2 \right\rangle,
\]

is also called the structure factor. It is in itself an interesting function and it is the quantity that is directly measured in scattering experiments.

### 3.7 Measuring Domain Size

In simulations of the ordinary Ising model as well as the conserved-order-parameter model, spins with the same orientation tend to stick together to form domains where all spins are oriented the same way (see Fig. 1). This is a result of the Hamiltonian; neighbors with aligned spins lower the total energy of the system; anti-aligned spins increase it. It would be interesting to have a way to calculate the average size of such a domain. It turns out that an estimate for the domain size can be directly obtained from the structure factor. It is a function of the displacement vector \( k \); but our system is essentially isotropic: it looks the same in all directions, so we can average \( S(k) \) in all directions, to get the circularly-averaged structure factor \( S(k) \). We shall often simply write “structure factor” to refer to the circularly-averaged structure factor; the meaning should be clear from the context.

In plots of the circularly-averaged structure factor of a system which was simulated at a constant temperature, there is always one peak (see Fig. 2). The wave vector of this peak corresponds to the typical domain size. A good way to calculate the position of the peak is by using the first moment \( k^* \):

\[
k^* = \frac{\int_0^{\infty} kS(k)dk}{\int_0^{\infty} S(k)dk}.
\]

The domain size \( R \) is then given by converting this wave vector to the spatial domain:

\[
R = \frac{2\pi}{k^*}.
\]

Another definition for the domain size uses the radial distribution function \( g(r) \) (or rdf for short). This is the circularly-averaged connected correlation function; it is the radial average of \( G^{(2)}_c(\mathbf{r}) \), which can be found by inverse Fourier transforming \( S(k) \). For the COP Ising model, the rdf starts at 1, crosses the zero line and then keeps oscillating around it. (See Fig. 5 for the rdf corresponding to the structure factor that was plotted in Fig. 2 in the introduction.) The typical domain size \( R \) can then be defined as the point where the rdf first crosses the zero line.
4 Results

All our simulations have been performed on a square lattice with \( N = 256 \times 256 \) sites for the COP Ising model with spin-exchange (Kawasaki) dynamics and Metropolis acceptance probability. Except for the prediction of the second peak in subsection 4.3, we always used an equal amount of up-pointing and down-pointing spins, i.e. we have a density of \( \rho = 0.5 \), which corresponds to a mean magnetization of \( m = 0 \).

4.1 Single Quench

In a single-quench experiment the simulation starts with a randomly mixed state of up- and down-pointing spins, with a certain density \( \rho \) of up-pointing spins. Usually a density \( \rho = 0.5 \) is used. The simulation then runs at a constant temperature below the critical temperature \( T_c \), so we get a single quench. During the simulation, quantities we are interested in (such as the structure factor) can be recorded.

We ran such a simulation at temperature \( T = 0.7T_c \), with density \( \rho = 0.5 \). The structure factor we found is plotted in figure 6 for three different times. Snapshots of the corresponding lattices can be seen in the introduction in figure 1. We compare our results to what is already known about the COP Ising model at constant temperature after a single quench.

We compare the large \( k \) behavior of our structure factor to the theoretical behavior. According to Porod’s law, at large \( k \) the structure factor \( S(k) \) behaves as \( k^{-(d+1)} \), where \( d \) is the dimension of the system. Hence, we fitted the data for \( t = 1.6 \cdot 10^4 \) MCS to \( S = a + bk^{-3} \), for values of \( k \) between 25 and 128. We found that \( a = 1.5 \times 10^{-1} \), and \( b = 5.3 \times 10^5 \). This function is also plotted in Fig. 6, and agrees reasonably well.

The growth of the domain size as a function of the time is given in figure 7. This size was calculated from the structure factor every \( 10^3 \) MCS using Eq. (16). The domain size \( R \) grows with time as

\[
R(t) = a + bt^{1/3}
\]

This growth law was derived theoretically by Lifshitz and Slyozov (Lifshitz & Slyozov (1961)), and at a later time confirmed by COP Ising simulations (see for example Amar et
Figure 6: Structure factor for three different times during a single-quench simulation of the COP Ising model at temperature $T = 0.7T_c$ and $\rho = 0.5$. For $t = 1.6 \cdot 10^4$ MCS a fit to Porod’s law is also drawn.

al. (1988) and Huse (1986)). The domain size in our simulation is in good agreement with this growth law.
Figure 7: Domain size $R$ (calculated using Eq. (16)) as a function of the time $t$ in MCS during a single-quench simulation of the COP Ising model at temperature $T = 0.7T_c$ and $\rho = 0.5$. The small crosses are data from our simulation, the solid line is a fit to $R(t) = a + bt^{1/3}$ ($a = 2.39, b = 0.2489$).

4.2 Heating

4.2.1 Underlying Process

For high temperatures the factor $e^{-\beta(E_\nu - E_\mu)}$ in the acceptance probability (Eq. (8)) becomes very close to 1, because $\beta = \frac{1}{kT}$ gets close to 0. In this section we shall assume that $\beta = 0$, i.e. an infinite temperature. This means that all proposed spin exchanges will be accepted. On average each spin pair is exchanged once during one Monte Carlo step. This means that every particle makes a random walk on a two-dimensional lattice. This means that in a few MCS the domain walls get fuzzier and the domains get less and less distinctive; inside a domain the concentration of particles with opposite spin increases.

Let us now consider a random walk along a $d$-dimensional lattice. We wish to determine the average squared distance traveled by a particle. Suppose a particle starts at the origin of the lattice. Every step consists of the random selection of one of the axes and the particle moves along this axis over a distance 1 either up or down. So a single step can be characterized by the axis number $k$, with $0 \leq k \leq d$ and a direction $s = \pm 1$. Suppose that a particle is at position $r(n)$ after a time of $n$ steps. If axis $k$ and direction $s$ were chosen, then at time $n + 1$, the particle is at a position $r(n + 1) = r(n) + s\hat{r}_k$, where $\hat{r}_k$ is the unit vector of the axis $k$.

$$\langle |r(n + 1)|^2 \rangle = \frac{1}{2d} \sum_{k \in \{1,...,d\}} \sum_{s = \pm 1} |r(n) + s\hat{r}_k|^2 = \frac{1}{2d} \sum_{k \in \{1,...,d\}} \left( |r(n) + \hat{r}_k|^2 + |r(n) - \hat{r}_k|^2 \right)$$

$$= \frac{1}{2d} \sum_{k \in \{1,...,d\}} \left( 2|r(n)|^2 + r(n) \cdot \hat{r}_k - r(n) \cdot \hat{r}_k + 2|r(\hat{r}_k)|^2 \right)$$

$$= \frac{1}{d} \sum_{k \in \{1,...,d\}} \left( |r(n)|^2 + 1 \right) = |r(n)|^2 + 1$$

So every single step the average squared distance to the origin increases with 1. This means
that it satisfies
\[ \langle |r(n)|^2 \rangle = n. \quad (17) \]

If we look at the random walk as a diffusion process, we can calculate the diffusion constant \( D \), from
\[ \langle |r(t) - r_0|^2 \rangle = 2dDt, \quad (18) \]

Because on average each spin takes two steps of a random walk, we can connect our simulation to a random walk by setting \( n = 2t \), with \( t \) the simulation time of heating in MCS. Combining Eqs. (17) and (18) yields
\[ D = \frac{1}{d}, \]
so for our system we expect a diffusion constant of \( D = \frac{1}{2} \).

We have adapted our simulation so that after a certain time of domain formation at a temperature below the critical temperature, we start to keep track of the particles at the edges of a domain wall; these particles get numbered and their initial coordinates are stored. We shall call this time \( t = 0 \). Subsequently, the simulation is run at infinite temperature (\( \beta = 0 \)). At different times we calculate the squared distance of each numbered particle to its original position (remembering the boundary conditions), and save the average over all numbered particles. The results of such a simulation that started with \( 5 \cdot 10^3 \) MCS at temperature \( T = 0.7T_c \) are shown in Fig. 8. A fit to \( (\Delta r)^2 = at \) yields a value for \( a \) of \( a = 2.028 \). Because our simulation is two-dimensional, it follows from Eq. (18) that the diffusion constant \( D \) is approximately
\[ D = \frac{2.028}{4} = 0.507, \quad (19) \]
so the diffusion constant is in good agreement with the theoretical diffusion constant \( \frac{1}{2} \) that we calculated above.

So far we have only considered the average squared distance the particles have traveled, we now consider how the possible displacements along the \( x \)-axis and \( y \)-axis are distributed. For a single particle making a one-dimensional random walk starting at the origin, the probability distribution \( P_n(d) \) of finding the particle at coordinate \( d \) after \( n \) steps have been taken is given by a binomial function
\[ P_n(d) = \begin{cases} \frac{1}{2^n} \binom{n}{\frac{d+n}{2}} & \text{if } d+n \text{ is even.} \\ 0 & \text{if } d+n \text{ is uneven.} \end{cases} \]

For higher values of the number of steps \( n \), this probability distribution can be approximated with great accuracy by a Gaussian curve:
\[ h_\sigma(x) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{x^2}{2\sigma^2}}. \quad (20) \]

(We use the letter \( h \) instead of \( g \) to denote the Gaussian function, because the letter \( g \) is already used for the radial distribution function.) Its total area is one, and its width (the standard deviation of the distribution) \( \sigma \) will increase with the number of random walk steps. The square of \( \sigma \) is equal to the average squared distance traveled by a random walking particle. We found before that this squared distance is equal to the number of steps \( n \) (Eq. (17)), so \( \sigma = \sqrt{n} \).

Every Monte Carlo step, each particle makes two random walk steps in one of four random directions along the axes on average. Because spin pairs are chosen at random, some particles may have taken no steps at all, and some may have taken a lot more than two steps. This means that, as opposed to the binomial distribution where particles cannot travel further than
the number of random walk steps, a particle in our simulation can in principle be found at a much larger distance from its original position. Still the average squared distance the particles travel, very accurately follows the theoretical value of a random walk as can be seen in Fig. 8. Furthermore, in the binomial distribution a particle can never be found at position $d$ when $d + n$ is uneven. This will not be so in our case because it is very probable for the number of random walk steps that a single particle has taken to differ from the average number of steps ($2t$) the particles have taken.

All this gives reason to believe that for our system the Gaussian distribution (where there is a small probability to find a particle very far from its original position) is in fact more accurate than the binomial distribution. (Of course this Gaussian distribution is only valid for integer displacements.)

We have seen before that the number of random walk steps is exactly twice the number of MCS. These steps are in one of four two-dimensional random directions. This means that each MCS, on average one step will be taken along the $x$-axis, and one along the $y$-axis. So if we only consider a single coordinate, the width of its one-dimensional Gaussian distribution must be:

$$\sigma = \sqrt{t}.$$

Fig. 9 shows the distribution along the $x$-axis (the distribution along the $y$-axis is similar) as found in our simulation. This simulation was performed on a $256 \times 256$ lattice and started with $5 \cdot 10^3$ MCS at temperature $T = 0.7T_c$. We then started to keep track of the position of particles at the boundary of the domains (approximately $7 \cdot 10^3$). The distribution is found by counting how many of these particles have reached the displacement $\Delta x$. The Gaussian function with $\sigma = \sqrt{t}$ is also drawn. It is clear from this picture that indeed the displacement distribution of particles in our system quite accurately follows a Gaussian distribution, which is already apparent after only 10 MCS.
4.2.2 Structure Factor

We would like to know how the heating process influences the structure factor of the system. In other words; if $S_0(k)$ is the structure factor as obtained by an Ising simulation at temperature under the critical temperature, is it possible to predict the structure factor $S_t(k)$ after $t$ MCS of heating at infinite temperature; without having to perform the actual simulation?

Our hypothesis is that the existing correlation function changes over time by integrating over the Gaussian distribution. Of course our system is two-dimensional, so we should integrate $g(x, y)$ at time $t = 0$, just before heating, over a two-dimensional isotropic Gaussian curve. (i.e. it looks the same in both the $x$ and $y$ directions.) This two-dimensional Gaussian function is given by

$$h_{\sigma}(x, y) = \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{x^2}{2\sigma^2}} \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{y^2}{2\sigma^2}} = \frac{1}{2\pi\sigma^2} e^{-\frac{(x^2+y^2)}{2\sigma^2}}$$  \hspace{1em} (21)

The squared distance in a two-dimensional random walk on a lattice is the number of random walk steps $n$ (Eq. (17)), so we take a value of $\sigma_t = \sqrt{n} = \sqrt{2t}$, with $t$ the time in MCS. So our predicted correlation function at time $t$ MCS of heating is:

$$g_t(x, y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} g_0(x, y) e^{-\frac{(x-x')^2+(y-y')^2}{2\sigma_t^2}} \, dx' \, dy'$$  \hspace{1em} (22)

We have directly calculated this convolution at several times during another simulation that started with $5 \cdot 10^3$ MCS at a temperature of $T = 0.7T_c$, and for a simulation that started with $10^5$ MCS at the same temperature. The measured correlation functions and the predicted correlation functions at two different times are shown in Figs. 10 and 11. The measured results from our simulation in this figure are very close to the predicted results. However they do differ at the point $r = 0$. Because a particle’s spin is identical to itself, the correlation between a
spin and itself is 1. This means that \( g(r = 0) = g(0, 0) = 1 \); this is why we “manually” put \( g_t(0, 0) = 1 \), before radial averaging.

Because we are interested in the structure factor, we have performed the same simulations, but calculated the structure factor by applying a Fourier transform to the \( g(x, y) \) of Eq. (22). \( g(x, y) \) was corrected at \( r = 0 \) by putting \( g(0, 0) = 1 \). The results are shown in Figs. 12 and 13. The predicted structure factors are in good agreement with the structure factors found from the simulation.

Because the structure factor \( S(k_x, k_y) \) is by definition the Fourier transform of \( g(x, y) \), the predicted structure factor \( S_t(k_x, k_y) \) after \( t \) MCS of heating can be found by Fourier transforming \( g_t(x, y) \). To find the correct prediction of \( S \) we cannot ignore the correction \( g(0, 0) = 1 \) that we made above. This correction can be written as:

\[
g'_t(x, y) = g_t(x, y) + (1 - g_t(0, 0))\delta_{(0,0)}(x, y),
\]

where \( g_t(x, y) \) is the correlation function as given in Eq. (22), and

\[
\delta_{(0,0)}(x, y) = \begin{cases} 1 & \text{if } x = y = 0, \\ 0 & \text{for all other values of } x \text{ and } y. \end{cases}
\]

The discrete Fourier transform of \( \delta_{(0,0)} \) is the constant function 1, so the discrete Fourier transform of \( g'_t(x, y) \) is:

\[
S_t(k_x, k_y) = \mathcal{F} [g_0(x, y) * h_{\sigma_t}(x) * h_{\sigma_t}(y)](k_x, k_y) + \mathcal{F} [(1 - g_t(0, 0))\delta_{(0,0)}](k_x, k_y)
= S_0(k_x, k_y)\mathcal{F}[h_{\sigma_t}(x)](k_x)\mathcal{F}[h_{\sigma_t}(y)](k_y) + 1 - g_t(0, 0),
\]

where we used that the Fourier transform of the convolution between two functions is the product of their Fourier transforms.

The discrete Fourier transform of the Gaussian function \( h_{\sigma_t}(x) \) is again a Gaussian function:

\[
\mathcal{F}[h_{\sigma_t}(x)](k_x) = e^{-2\pi^2\sigma^2(k_x^2 + k_y^2)}.
\]

Here \( L \) is the width of the lattice. Since we have a square lattice, the number of lattice sites \( N \) satisfies \( N = L^2 \). Substituting the above equation in Eq. (24) yields

\[
S_t(k_x, k_y) = S_0(k_x, k_y)e^{-2\pi^2\sigma^2 \frac{k_x^2 + k_y^2}{L^2}} + 1 - g_t(0, 0).
\]

\( S_0 \) can be calculated by from a Fourier transform of the spins in the lattice (see Eq. (15)).

This is a much more efficient way to calculate the structure factor, because instead of performing a two-dimensional integration for all our lattice points, we can directly multiply the structure factor \( S_0(k_x, k_y) \) by the Fourier transform of the two-dimensional Gaussian function and add \( 1 - g_t(0, 0) \). The value for \( g_t(0, 0) \) can be calculated from Eq. (22). So using this method to find the structure factor \( S \), we still need to calculate the connected correlation function \( g(x, y) \) at \( t = 0 \) (by inverse Fourier transforming \( S_0 \)) and perform a two-dimensional (numerical) integration for the point \( (0, 0) \). However all this does not take much time compared to directly calculating this convolution at every point of the \( L \times L \) lattice. Note that Eq. (25) depends only on \( k_x^2 + k_y^2 = k^2 \), where \( k \) is the distance of \( (k_x, k_y) \) to the origin. So the prediction of the circularly-averaged structure factor \( S_t(k) \) can be calculated by

\[
S_t(k) = S_0(k)e^{-2\pi^2\sigma^2 \frac{k^2}{4}} + 1 - g_t(0, 0).
\]

This calculation requires much less multiplications than the two-dimensional case of \( S_t(k_x, k_y) \).

We have performed two simulations under the same conditions as before (starting with systems simulated for \( 5 \times 10^5 \) MCS and \( 10^5 \) MCS at \( T = 0.7T_c \)) and determined the simulated
Figure 10: Circularly-averaged connected correlation function $g(r)$ at different times during heating. The simulation started with a lattice found by a single-quench simulation of the COP Ising model at temperature $T = 0.7T_c$ after $5 \cdot 10^3$ MCS. The solid lines are data from our simulation, the dashed lines are predictions of $g(r)$ found by convoluting $g(x, y)$ just before heating ($t = 0$) with a two-dimensional Gaussian curve with $\sigma = \sqrt{2t}$. Note that for all curves $g(0) = 1$.

and predicted structure factors. The results can be seen in Figs. 14 and 15. As can be seen in these figures, the predicted structure factors are in good agreement with the simulated ones (and also with the ones in Figs. 12 and 13 found by directly calculating the Gaussian convolution of $g(x, y)$).
Figure 11: Circularly-averaged connected correlation function $g(r)$ at different times during heating. The simulation started with a lattice found by a single-quench simulation of the COP Ising model at temperature $T = 0.7T_c$ after $10^5$ MCS. The solid lines are data from our simulation, the dashed lines are predictions of $g(r)$ found by convoluting $g(x, y)$ just before heating ($t = 0$) with a two-dimensional Gaussian curve with $\sigma = \sqrt{2t}$. Note that for all curves $g(0) = 1$.

Figure 12: Circularly-averaged structure factor $S(k)$ corresponding to the correlation functions as plotted in Fig. 10. The different $S(k_x, k_y)$ were calculated by performing a fourier transform on the corresponding $g(x, y)$. 
Figure 13: Circularly-averaged structure factor $S(k)$ corresponding to the correlation functions as plotted in Fig. 11. The different $S(k_x, k_y)$ were calculated by performing a fourier transform on the corresponding $g(x, y)$.

Figure 14: Circularly-averaged structure factor $S(k)$ corresponding to the correlation functions as plotted in Fig. 10. The predicted $S(k_x, k_y)$ at different times were calculated by multiplying $S(k_x, k_y)$ at $t = 0$ (just before heating) with the fourier transform of the Gaussian curve that was used in the Gaussian convolution of Fig. 10, and adding $1 - g_t(0,0)$ to correct for the correlation between a particle and itself.
Figure 15: Circularly-averaged structure factor $S(k)$ corresponding to the correlation functions as plotted in Fig. 11. The predicted $S(k_x, k_y)$ at different times were calculated by multiplying $S(k_x, k_y)$ at $t = 0$ (just before heating) with the fourier transform of the Gaussian curve that was used in the Gaussian convolution of Fig. 11, and adding $1 - g_t(0,0)$ to correct for the correlation between a particle and itself.

4.3 Camel Bump

4.3.1 Creation: Double Quench

Let us consider a system that has been simulated with a standard single-quench simulation for a significant amount of time at temperature below the critical temperature, followed by some time of heating at infinite temperature (for example $10^5$ MCS at $T = 0.7T_c$, see Fig. 16).

After heating for a few dozen MCS the structure factor still has a peak at small values of $k$. This peak gets lower when the heating proceeds. The large values of $k$ correspond to the small-scale structure. We see that, after heating, the large-$k$ part of the structure factor resembles the structure factor of a randomly mixed lattice, like the one we use to start a single-quench Ising simulation. This structure is also drawn in the figure. Apart from some noise, a randomly mixed system of spins with density $\rho = 0.5$ has a structure factor equal to the constant function 1. The corresponding correlation function is the inverse discrete Fourier transform of this function. We have seen before that this is the function $\delta_{(0,0)}$, which is 1 at $(0,0)$ and 0 elsewhere. So the correlation function $g(r)$ is 1 for $r = 0$ and 0 everywhere else, meaning there is no correlation between different spins.

In a single-quench experiment, the small distance scale corresponding to the large-$k$ part of the structure factor is where domain formation begins: the single peak starts at large $k$ and moves to smaller $k$ when time progresses. Because the large-$k$ region after heating resembles the begin situation of a single-quench experiment (which is also apparent in a snapshot of the situation, see Fig. 17), we expect that we can grow a second peak just as in the single-quench experiment by continuing the simulation of the system at temperature below the critical temperature. So this is a double quench. Because the speed with which the domains grow decrease with simulation time ($R(t) = a + bt^3$), we expect the peak at smaller $k$ to remain almost intact while the second peak at larger $k$ grows and shifts to smaller $k$.

We have performed this double-quench simulation starting with the system corresponding to 100 MCS of heating in Fig. 16, and letting the simulation proceed at a temperature of $T = 0.7T_c$, so the temperature has dropped instantly from infinite to below the critical temperature (the
Figure 16: Circularly-averaged structure factor \( S(k) \) at different times \( t \) (MCS) of heating. The simulation started with a lattice found by a single-quench simulation of the COP Ising model at temperature \( T = 0.7T_c \) after \( 10^5 \) MCS. The structure factor of a randomly mixed state of particles with \( \rho = 0.5 \) is also drawn.

Figure 17: Snapshot of the lattice corresponding to the structure factor at time \( t = 0 \) and \( t = 100 \) in Fig. 16.
Figure 18: Circularly-averaged structure factor $S(k)$. The simulation started with a randomly mixed state with $\rho = 0.5$ which was simulated for $10^5$ MCS at $T = 0.7T_c$ and subsequently heated for 100 MCS at $T = \infty (\beta = \frac{1}{kT} = 0)$. Then the temperature was changed back to $T = 0.7T_c$. The structure factor is shown for different times after this double quench. For comparison, for $t = 5 \cdot 10^3$ the structure factor of a single-quench experiment at the same temperature and $\rho = 0.5$ is also drawn.

second quench). The results are shown in Fig. 18. For comparison, we also performed a single-quench simulation starting with a randomly mixed state at the same temperature. The structure factor of this system is shown for $t = 5 \cdot 10^3$. It is clear from these two figures that the peak at larger $k$ grows approximately like the way it does in a single-quench experiment, but the peak in the latter experiment is slightly higher.

4.3.2 Prediction of the Second Peak

It is clear from Fig. 16, that it takes some time of heating before the large-$k$ part of the structure factor reaches the constant value 1. In fact, at $t = 100$, it has not yet reached 1 but a value of 0.95. At $t = 20$ for example, it is at $S = 0.64$. This difference from 1 could explain the fact that the second peak was slightly lower than the peak we get in a single-quench experiment (Fig. 18).

A structure factor that is constant, but smaller than 1 corresponds to a randomly mixed system with a total magnetization per spin that differs from 0; in other words a system with an unequal amount of up-pointing and down-pointing spins. To show this we consider a system with an arbitrary magnetization $m$, with $-1 \leq m \leq 1$, in which all spins are randomly mixed. This means that $\langle s_i s_j \rangle = 1$ for $i = j$ as it is in the case of $m = 0$. But when $i \neq j$ we have:

$$\langle s_i s_j \rangle = \frac{1}{N^2} \sum_{i=0}^{N} \sum_{j=0}^{N} s_i s_j = \frac{1}{N} \sum_{i=0}^{N} s_i \frac{1}{N} \sum_{j=0}^{N} s_j = m^2.$$ 

So by using the definition of the connected correlation function (Eq. (14)), we see that

$$g(i, j) = \begin{cases} 1 - m^2 & \text{if } i = j, \\ 0 & \text{otherwise}. \end{cases}$$

Instead of a jump from 1 to 0 there is a jump from $1 - m^2$ to 0. So instead of the constant
function 1, the structure factor \( S(k) \) of a randomly mixed system with magnetization \( m \) is given by

\[
S(k) = 1 - m^2. \tag{26}
\]

In the previous section, we made a prediction of the structure factor during heating, given in Eq. (25), where we could successfully get the right large-\( k \) behavior of \( S \) by making a correction for the fact that \( g(0,0) \) should be 1. This means for \( S \) that we add the constant term \( 1 - g_t(0,0) \), where \( g_t(0,0) \) is the value of the uncorrected prediction for \( g \) at \((0,0)\). (This is a convolution of \( g \) at the point \((0,0)\) with a Gaussian as given in equation (22)). If, for a moment, we ignore the first peak at small \( k \) of the structure factor during heating, we see that it has exactly the structure as given in Eq. (26), so we have for the difference between 1 of the correction term

\[
g_t(0,0) = m_{\text{eff}}^2,
\]

where we wrote \( m_{\text{eff}} \) instead of just \( m \), because the system we are looking at has another mean magnetization (in fact the systems we simulated have a mean magnetization of \( m = 0 \)). This suggests that the small scale structure of the system has an effective magnetization, which differs from the mean magnetization \( m \), and goes towards magnetization 0 as the heating progresses. Intuitively this is quite clear because in a domain (almost) all spins have the same direction, so that inside the domain \( m^2 = 1 \). When the heating progresses, the spins near the domain border move further and further into the adjacent domain, so that the local concentration goes closer and closer towards \( \rho = 0 \).

We suppose now, that we can get a better prediction of the growth of the second peak after heating, by performing a single-quench simulation at a magnetization that satisfies Eq. (26) for the large-\( k \) value of the structure factor during heating. We tested this for the structure factor as shown in Fig. 16 that has been heated for 50 MCS. The large-\( k \) value of this structure factor is clearly below 1. We fitted this structure factor for the values \( 25 \leq k \leq 128 \) to the constant function. This yields a value of \( S = 0.840 \). Equating this value to \( 1 - m_{\text{eff}}^2 \) gives a value of \( m_{\text{eff}} = 0.400 \). We know from Eq. (11) that

\[
\rho = \frac{1}{2} m + \frac{1}{2} 
\]

so this value of \( m \) corresponds to a particle density \( \rho_{\text{eff}} = 0.700 \), which differs significantly from the density \( \rho = 0.5 \). We have performed two single-quench experiments, both at \( T = 0.7T_c \): one with \( \rho = 0.5 \) and one with \( \rho = 0.700 \). These are shown for \( t = 100 \) MCS and \( t = 10^3 \) MCS in Figs. 19 and 20, together with the double-quench experiment that starts with the lattice found after 50 MCS of heating. We see that, for \( t = 100 \) MCS, the structure factor for \( \rho = 0.700 \) matches exactly the structure of the second peak in our double-quench experiment, whereas the structure factor for \( \rho = 0.5 \) has too large values. For \( t = 10^3 \) MCS, the structure factor for \( \rho = 0.5 \) still has too large values, but now the same is also true for \( \rho = 0.5 \), nevertheless, the difference with the double-quench peak is much less.

From Fig. 20 it is clear that the first peak also changes during the second-quench part of our simulation; it starts growing again. While we can reasonably “predict” the structure of the second peak by the structure of a single-quench experiment, we do not really know how the first peak of the camel bump changes with time. This first peak cannot be directly compared to the one in a single-quench experiment since it is lowered considerably by the heating process (in this example from a value around \( S = 110 \) to a value around \( S = 20 \)), while the position is at a much smaller value of \( k \) than can be expected for a peak of this height.

We would now like to get two peaks of equal height. We do not want the second peak to get too close to (or worse, to be swallowed up by) the first peak before they reach an equal
Figure 19: Circularly-averaged structure factor $S(k)$ at time $t = 0$ and $t = 100$ MCS of a double quench. The simulation started with a lattice found by a single-quench simulation of the COP Ising model at temperature $T = 0.7T_c$ after $10^5$ MCS, followed by 50 MCS of heating at infinite temperature. For comparison the structure factor of a single quench experiment at time $t = 100$ and same temperature with $\rho = 0.5$ and $\rho = 0.700$ is also drawn.

Figure 20: Circularly-averaged structure factor $S(k)$ at time $t = 0$ and $t = 10^3$ MCS of a double quench. The simulation started with a lattice found by a single-quench simulation of the COP Ising model at temperature $T = 0.7T_c$ after $10^5$ MCS, followed by 50 MCS of heating at infinite temperature. For comparison, the structure factor of a single-quench experiment at time $t = 10^3$ and same temperature, with $\rho = 0.5$ and $\rho = 0.700$ is also drawn.
Figure 21: Circularly-averaged structure factor $S(k)$. The simulation started with a randomly mixed state with $\rho = 0.5$ which was simulated for $10^5$ MCS at $T = 0.7T_c$ and subsequently heated for 150 MCS at infinite temperature. Then the temperature was changed back to $T = 0.7T_c$. The structure factor is shown for different times after this double quench. For comparison, for $t = 10^3$ MCS, the structure factor of a single-quench experiment at the same temperature and $\rho = 0.5$ is also drawn.

height. This is what happened in Fig. 18, which had 100 MCS of heating. In order to prevent this we simply lower the first peak a bit more by heating for a longer time. We choose 150 MCS of heating, and then switch back the temperature to $0.7T_c$ and look at how much MCS we need for the peaks to reach equal height. The structure factor is shown in Fig. 21 for some different running times of the double-quench experiment. The structure factor for $t = 10^3$ MCS corresponds to the one shown in the introduction. Snapshots of the lattice for the camel bump, and before and after heating can be seen in Fig. 3 of the introduction.

After 150 MCS of heating, the large-k part of $S$ equals the constant 0.988, very close to 1. Fig. 21 also shows the structure factor in a single quench experiment with $\rho = 0.5$ at $t = 10^3$ MCS. It is clear that the second peak at $t = 10^3$ corresponds very well to this peak.
5 Conclusions

Our main focus has been on using external parameters to influence the shape of the circularly-averaged structure factor $S(k)$ in a model of a demixing binary mixture; which was simulated by a conserved-order-parameter Ising model. By varying only the temperature, we have indeed been able to create structure factors with two peaks (including a “camel bump”) as opposed to the single peak one gets in the usual single-quench experiment. The two peaks correspond to both a smaller and a larger scale structure of the lattice (see Fig. 3 for a snapshot). In order to get two peaks, we used the following double-quench process:

- The system starts with a homogeneous binary mixture and is evolved for a relatively long time at a constant temperature below the critical temperature (single quench). (e.g. to get the $S(k)$ of Fig. 4 we simulated for $10^5$ MCS at $T = 0.7T_c$.)

- The system is heated for a very short time at a constant temperature much larger than the critical temperature. (e.g. we heated for 150 MCS at infinite temperature, i.e. $\beta = \frac{1}{kT} = 0$.)

- The system is evolved again at a constant temperature below the critical temperature (second quench), but for a much shorter time than the first quench. (e.g. we simulated for $10^3$ MCS at $T = 0.7T_c$.)

By varying the simulation time we can influence the position of the two peaks; by varying the duration of heating we can influence the height of the first peak. We suspect that we can also make three (or even more) peaks in the same way we made the second peak, provided we take much longer simulation times to prevent a new peak merging into the last one.

Besides this, we have also confirmed the known fact that in a single-quench experiment, for large values of $k$ the circularly-averaged structure factor $S(k)$ follows Porod’s law: $S(k) \sim k^{-(d+1)}$ with $d = 2$; the dimension our system (see Fig. 6). Furthermore, we have confirmed with great accuracy the growth law $R(t) = a + bt^{\frac{1}{3}}$ for the domain size $R$ (see Fig. 7).

We are able to predict the exact shape of the structure factor during heating, without having to perform a simulation. Apart from a small correction, the way we calculate this is exactly equivalent to the operation of Gaussian smoothing on the radial distribution function $g(x, y)$. The smoothed function can be calculated by convoluting the function over a two-dimensional isotropic Gaussian curve with $\sigma = \sqrt{2t}$. In terms of signal processing, this is equivalent to the application of a Gaussian low-pass filter. A low-pass filter removes or reduces high frequencies and keeps low frequencies intact. Such a filter can be applied by multiplying the Fourier transform of $g(x, y)$, namely the structure factor $S(k_x, k_y)$, with the Fourier transform of the Gaussian (which is again a Gaussian). Indeed, apart from the correction, this is what we found in Eq. (25). This is a very efficient way to calculate the predicted structure factor.

We cannot give a good prediction for the evolution of the first peak, when the system evolves at a constant temperature after the double quench. This peak starts growing again, but its growth cannot be directly compared to the growth of a peak in a single-quench experiment. This is because the peak was lowered considerably by the heating process, while its position is at a much smaller value of $k$ than can be expected for a peak of this height. Also we do not know of an analytical way (like we had for heating) of predicting the growth of this peak.

We do have been able to “predict” the shape of the second (large-$k$) peak in the structure factor of a doubly-quenched system by comparison to a single-quench experiment. The large-$k$ part of $S(k)$ after heating, which is a constant between 0 and 1, is the same as the structure of a randomly mixed system at a magnetization that satisfies $S(k) = 1 - m^2$ (Eq. (26)). Hence we can calculate an effective magnetization by equating the constant value after heating to $1 - m^2_{eff}$. Now the new peak grows exactly like the one in a single-quench simulation that starts with a homogeneous mixture at magnetization $m_{eff}$, and which evolves at the same temperature. This is true until its position gets close to the position of the first peak.
References


