Quenched Dynamics of Artificial Spin Ice: Coarsening versus Kibble-Zurek

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Artificial spin ices are ideal frustrated model systems in which to explore or design emergent phenomena with unprecedented characterization of the constituent degrees of freedom. In square spin ice, violations of the ice rule are topological excitations essential to the kinetics of the system, providing an ideal testbed for studying the dynamics of such defects under varied quench rates. In this work we describe the first test of the Kibble-Zurek mechanism and critical coarsening in colloidal square and colloidal hexagonal ice under quenches from a weakly interacting liquid state into a strongly interacting regime. As expected, for infinitely slow quenches, the system is defect free, while for increasing quench rate, an increasing number of defects remain in the sample. For square ice, we find regimes in which the defect population decreases as a power law with decreasing quench rate. A detailed scaling analysis shows that for a wide range of parameters, including quench rates that are accessible by experiments, the behavior is described by critical coarsening rather than by the Kibble-Zurek mechanism, since the defect-defect interactions are long ranged. For quenches closer to the critical point, however, there can be a competition between the two mechanisms.

INTRODUCTION

The term artificial spin ice (ASI) describes a variety of systems that can be modeled by frustrated, interacting effective binary degrees of freedom which obey the ice rule. The ASI size scales are much larger than those of real spin ice systems, allowing the individual spin degrees of freedom to be imaged directly [1–3]. ASI can be realized using arrays of nanomagnets [1–9], colloids in ordered trap arrays [10–14], and vortices in nanostructured superconductors [15–19]. Of the wide variety of different ASI geometries, the first and most studied are square [1–3, 8, 20] and hexagonal ices [2, 4–7, 11, 18, 21]. While both obey the ice rule in their low energy states, the square geometry produces an antiferromagnetic ground state, while the ice-manifold of hexagonal ice can remain disordered.

A particularly appealing feature of ASI systems is that they contain well defined defects that take the form of non-ice rule obeying vertices. The system can be characterized by its different vertex types, which can be labeled according to the number of spins pointing toward each vertex. In the square ice, the vertices are named Nn where n is the number of spins pointing toward the vertex. Here, N0 and N4 are called double monopoles, N1 and N3 are monopoles, and the N2 biased and N2 gs are ice rule obeying vertices, where the latter is the ground state vertex configuration [2]. In Figure 1(a) we highlight the different vertex types for the square ice, while Figure 1(b) shows the same for the honeycomb ice.

At high temperatures or when the interactions between neighboring effective spins are weak, the two-dimensional square ice forms a liquid state with finite non-ice rule obeying vertex populations. As the temperature decreases or the interaction strength increases, there is a phase transition to a long range ordered state in which only N2 gs vertices are present [2]. In the square ice system there is a underlying second-order phase transition from the ordered state to the disordered state [22] while for the honeycomb ice this is only a crossover [2].

Previous work on ASI has generally focused on equilibrium states; however, ASI are ideal systems in which to address current issues in nonequilibrium statistical mechanics. For example, the density of topological defects in a system after it is quenched at different rates through a second order phase transition has implications for defect formation in the early universe [23, 24], vortex formation at normal to superconducting transitions [25], liquid
crystal systems [26], Bose-Einstein condensates [27–29], ion crystals [30, 31], and manganites [32].

One scenario describing the behavior of the defects for varied quenched rates is the Kibble-Zurek (KZ) mechanism [23, 24, 33], in which the defect density $\rho_d$ increases with higher quench rate according to a universal power law, $\rho_d \propto \tau_Q^{-\beta}$, where $\tau_Q$ is the the time duration of the quench or inverse quench rate. Thus for large $\tau_Q$ or slow quench rates, $\rho_d$ is expected to be small. In the KZ mechanism, $\beta$ is related to the critical exponents associated with the underlying equilibrium second order phase transition through which the system is quenched. The KZ mechanism relies on the adiabatic-impulse approximation according to which defects are produced when the system falls out of equilibrium (the freeze-out time scale). In addition, it assumes that the density of defects arises exclusively from the nonadiabatic crossing of the critical point, in the absence of any dynamics in the ordered phase that may alter the defect population. Other scenarios for how the defect density could behave include a coarsening process produced by the motion and annihilation of defects on the ordered side of the phase transition due to strong defect-defect interactions [34]. An ASI system is ideal for testing these different scenarios since excitations such as monopoles are very well defined and the universality class of the phase transition in many types of ASI, including the square ice, is known. In addition, since the square ice exhibits a phase transition but the honeycomb ice does not, the two types of ice should have very different behaviors during a quench.

Here we consider simulations and scaling analysis of a magnetically interacting colloidal artificial spin ice. The advantage of colloidal ice is that the strength of the colloid-colloid interactions can be tuned experimentally as a function of time, bringing the system from a non-interacting regime to a strongly interacting regime as a function of magnetic field and giving access to a range of different quench rates. We start the system in the weakly interacting disordered regime, increase the magnetic field through the phase transition, and measure the population of the different vertex types as well as the spatial configurations of the defects. We consider both square ice, where there is a second order phase transition to an ordered ground state, and hexagonal ice, where there is only a crossover to a disordered ice rule obeying state. Our simulation faithfully mimics the experimental set up as described in Refs. [11, 12, 14, 35]. An advantage to studying a particle based model is that the time-dependent dynamics during the quench can be directly accessed using molecular dynamics techniques, avoiding the issues that arise in using Monte Carlo (MC) methods to examine KZ scaling. Different MC methods produce different results [36], while the MD approach faithfully represents the dynamics that actually occur.
FIG. 4: Transition from the ordered to the disordered state as a function of quenching speed. a) Fraction of non-ground state vertex types $N_{ngs}/N$ in the system vs magnetic field values $B$. From dark blue to dark red, the curves represent total run times of $\tau_Q = 10$, 20, 40, 80, 150, 300, 600, 1200, 2500, and 6000 s. b) Rescaling of $N_{ngs}/N$ vs $t/\tau_Q$, the time divided by the total quench time raised to the power $\alpha = 0.75$. c) Scaling of $N_{ngs}/N$ as a function of quenching time $\tau_Q$ for different magnetic field values. From dark blue to dark red, the curves represent constant magnetic fields of $B = 4$ mT (dark blue), $B = 6$ mT (light blue) $B = 8$ mT (light green), and subsequently, $B = 10, 12, 16, 20, 24, 28, 32, 36$, and $40$ mT. d) Power law exponents $\beta$ obtained from the data in panel c) vs the magnetic field value $B$. Inset: The fraction of the larger ground state cluster $n_{gs1} = N_{gs1}/N$ (upper pink line) and the fraction of the smaller ground state cluster $n_{gs2} = N_{gs2}/N$ (lower purple line) as a function of $B$, showing a bifurcation at the critical field (dashed line), corresponding to the spontaneous symmetry breaking.

RESULTS

In Figure 2 we show a schematic of the magnetically interacting colloids in a square ASI of double well traps. Each elongated trap holds a single colloid which can sit on either end of the trap, determining the direction of the effective spin. The colloid-colloid interaction force is given by $F_{pp}(r) = A_c/r^4$ with $A_c = 3 \times 10^6 \chi_m^2V^2B^2/(2\pi\mu)$ for particles a distance $r$ apart, where $B$ is the magnetic field. For our parameters, the critical magnetic field at which the equilibrium system orders into a defect free ground state is $B_c = 9$ mT. We start the system at $B = 0$ and increase the field to $B = 40$ mT at different sweep rates. Figure 3 shows the vertex populations with the same color scheme from Figure 2 in a simulation with a quench time duration of $\tau_Q = 80$ s at several values of $B$. The defects form closed loop grain boundaries similar to those observed in square ice systems with varied amounts of quenched disorder [8, 15, 37, 38]. For faster quench rates or smaller $\tau_Q$, the number of non-ground state vertices increases and the grain boundaries are smaller.

In Fig. 4(a) we plot the fraction of non-ground state vertices $N_{ngs}/N$ versus $B$ at different sweep rates. The fastest transition with $t = 10$ s is denoted by the rightmost blue line, and the quench rate decreases for curves that are further to the left. The systems are initialized in a completely random configuration at $B = 0.0$ with $N_{ngs}/N = 7/8$. As the quench rate decreases, the value of $N_{ngs}/N$ decreases. In Figure 4(b) we show that the $N_{ngs}/N$ curves from Figure 4(a) can be collapsed by dividing the time by $\tau_Q^{1/z}$, where $\alpha = 3/4$.

In Figure 4(c) we plot $N_{ngs}/N$ versus the quench time $\tau_Q$ at different fixed values of the magnetic field from $B = 4$ mT (top) to $B = 40$ mT (bottom). The runs were performed over the experimentally accessible range of $t = 10$ s to $t = 6000$ s. We fit each curve to a power law with $N_{ngs}/N \propto \tau_Q^{-\beta}$, where $N_{ngs}/N = \rho_d$, and we plot the resulting exponents $\beta$ versus $B$ in Figure 4(d). For $B > 9$ mT, the system does not order at all, while for $B > 12$ mT, the exponent saturates at $\beta = 0.45$. This indicates that we have two different regimes of behavior. For smaller magnetic fields between the values of $B = 10$ mT and $B = 12$ mT, we find a slower decay rate with an exponent between $\beta = 0.2$ and $\beta = 0.3$.

Kibble-Zurek Mechanism

Now that we have established that our system has both a critical point and power law scaling of the defect density for different quench rates, we can test whether our results are consistent with the KZ mechanism [23, 24, 33]. In particular, the lag time between the nonequilibrium and equilibrium value scale is expected to be set by the so-called freeze-out time $t \sim (\tau_Q^{1/z})^{1/z} \sim \tau_Q^{1+z}$. To investigate whether the transition obeys the KZ mechanism, we collapse the runs with different quench times together by rescaling the time axis. In Figure 4(b) we show the evolution of $N_{ngs}/N$ versus time where the time has been divided by a power of the quench time $\tau_Q$. The collapse is achieved with $\alpha = 3/4$. The KZ mechanism prediction then implies that $\tau_Q^{1+z} = \alpha = 3/4$; however, the coarse ice falls into the Ising universality class with $\nu = 1$ and $z = 2$ [39], which gives $\tau_Q^{1+z} = 2/3$.

Another prediction of the KZ mechanism is that the total number of defects scales as $\rho_d \sim \tau_Q^{-2/z}$, where $D$ is the dimension of the system. In our case, $D = 2$ and $\rho_d = N_{ngs}/N$. The 2D Ising model gives a prediction of $\rho_d \sim \tau_Q^{-2}$, but in Fig. 4(c) we find $\rho_d \sim \tau_Q^{-1/2}$ or $\rho_d \sim \tau_Q^{-1/2}$. indicating that the scaling of the defects that we obtain is not a result of the KZ mechanism. For quenches out to higher values of $B$, the defects such as +1 and −1 monopoles are strongly interacting and undergo a nonnegligible amount of dynamical motion via their effective Coulomb interactions, as has been observed in colloidal experiments [12] and simulations [35]. The presence of defect dynamics during the part of the quench
in the ordered state violates one of the assumptions for the KZ scenario. We note that for coarsening dynamics near a critical point, the ordered regions of radius R grow as \( R(t) \propto t^{1/2} \) \cite{40}, which for the Ising model gives \( R(t) \propto t^{1/2} \), where t is time. If the size of the ordered regions grows, the number of defects could be proportional to \( 1/R(t) \), in agreement with our observations. In other types of ASI, such as nanomagnetic systems, it is possible that the KZ regime could be accessed more easily since the motion of the defects is slower. Alternatively, there could be a regime of KZ behavior at much faster quench rates than those we considered, for which the defects simply do not have time to move.

We note that although, up to a nearest neighbor approximation, a magnetic square ice can be mapped into a \( J_1, J_2 \) Ising system \cite{41}, the colloidal square ice differs greatly from the magnetic square ice, both in energetics and in the nature of its frustration \cite{14, 42}. The colloidal square ice can only be mapped exactly into a magnetic square ice at equilibrium \cite{22, 43, 44}. This is because the colloidal ice contains many more states, corresponding to colloids in between preferential positions, which might make its out-of-equilibrium kinetics much different from those of its magnetic analogue. An example of the difference between these two ice systems appears in Ref. \cite{35}.

**Hexagonal system**

In the hexagonal ASI, each vertex is surrounded by only three elongated pinning sites. Unlike the square ice, the hexagonal ASI has no phase transition from a disordered to an ordered phase, so we would not expect the KZ scenario to apply. We conduct the same type of simulation from a zero field state to a maximum field of \( B = 40 \) mT, where the equilibrium configurations at higher \( B \) do not contain any monopoles. In Figure 5 we show snapshots of the transition in the hexagonal ASI as a function of increasing interaction strength \( B = 8, 10, 12, \) and 14 mT for a quench time of \( \tau_Q = 80 \) s. In this case, non-favorable vertex types disappear during the crossover to the disordered ice-rule obeying state without forming any spatially correlated structures or grain boundaries of the type observed in the square ice system. Therefore, the defect dynamics and coarsening should be different between the two systems. In the initial random configuration, the ground state vertices in the hexagonal ice already occupy \( N_{gs}/N = 3/4 \) of the system, in contrast to the square system where \( N_{gs}/N = 1/8 \) at initialization. As a result, the hexagonal ice does not need to nucleate and grow clusters of ice rule obeying vertices.

In Figure 6 we plot the number of non-ground state vertices as a function of the applied field for different \( \tau_Q \) values ranging from \( \tau_Q = 10 \) s to \( \tau_Q = 6000 \) s for the hexagonal ice. The transition happens over a narrower range of \( B \) than in the square ice since there are no kinetic barriers to overcome in the process of eliminating the non-ground state defects. This also gives a higher exponent of \( \alpha = 0.88 \) in the rescaling of \( N_{ngs}/N \) versus \( t/\tau_Q^2 \) shown in the inset of Fig. 6, indicating that the defect annihilation mechanism differs from what is found in the square ice system. In the square ice, the monopoles are located along grain boundaries and annihilate as the grain boundaries shrink. In contrast, the monopoles in the hexagonal ice are not on grain boundaries and can move toward each other along straight paths, as found in experiments \cite{6}. More relevantly, monopoles in hexagonal ice are not topologically protected. While charges ±2 of square ice cannot be reabsorbed but can only be annihilated and created in pairs, ±3 charged violations of the ice rule in hexagonal ice can appear and disappear individually. That is because even ice rule vertices are charged (±1) and thus each monopole in hexagonal ice can transfer charge to the surrounding plasma.

In Fig. 6(b), the plot of defect density \( N_{ngs}/N \) versus the quench duration \( \tau_Q \) for the hexagonal ice at varied \( B \) shows that there is no power law behavior in the density of defects, which is consistent with the lack of a phase transition in the system. As a result, the KZ mechanism scenario does not apply, and critical coarsening cannot occur due to the lack of a critical point.
the coarsening dynamics in the magnetic AFI could differ from that found in the particle-based AFI since the particle based system minimizes the global energy rather than the vertex energy, making the resulting ice state more fragile [14, 43]. The kinetics of annihilation and spin flipping are also likely to depend on the microscopic details of the particular ASI realization. Many ASI systems have Coulomb interactions between the monopoles, and in these it is possible that the KZ mechanism always competes with coarsening. One possible experiment to test this would be to create magnetic nanoislands that are sufficiently far apart to reduce the strength of the defect-defect interactions and minimize the coarsening. Other future directions are to consider alternative ASI geometries [2, 9, 14, 47–49], including geometries in which the monopoles are not as strongly bound [50, 51]. It would also be interesting to study the effect of disorder to see if the exponents change or whether glassy dynamics arise such as a crossover to a logarithmic rather than a power law decay. It may also be possible that a small amount of disorder could slow down the dynamics of the defects enough that the KZ mechanism regime could be accessed.

Conclusion

In conclusion, we have examined the defect density populations for varied quench rates from a disordered to an ordered state in square and hexagonal magnetically interacting colloidal spin ice systems. In the square ice, we find that when the quench into the ordered state is sufficiently deep, there is a power law decay of the defect density with \( \rho_d \propto \tau_Q^{-1/2} \). Based on scaling arguments for the university class of the square ice, we find that the behavior of the quenched square ice is governed by coarsening rather than the Kibble-Zurek mechanism. The lack of KZ behavior is likely due to the strong Coulomb interactions between the monopoles. This causes a considerable amount of defect dynamics to occur during the quench, while the KZ mechanism assumes that no dynamics occurs in the ordered phase. In the case of the hexagonal ice, which has no second order phase transition to an ordered state, we find a very different type of defect configurations as well as a lack of power law scaling of the defect density with varied quench rates.

Our results could be compared with quenches of different types of AFI in magnetic, colloidal, and superconducting systems. Each of these systems could exhibit different interactions between the defects or different kinetics, and it is possible that one or more of the systems could have a regime in which the KZ mechanism is observable.

Discussion

Our results can be compared directly to current experimental colloidal ASI systems. The experimental samples are smaller than what is considered in our simulations; however, the experiments could be repeated many times to improve the statistics. In magnetic ASI, there is now a system in which thermal transitions from a liquid to an ordered state can be realized [2, 45, 46]. In such samples, quenches can be performed by varying the rate at which the temperature is swept across the transition from the liquid to ordered state. In superconducting systems, where artificial ices can be realized using magnetic flux lines, a similar temperature control could be used at finite fields in passing from a normal to a superconducting state as a function of temperature. It is possible that...
METHODS

We simulate a system of colloidal superparamagnetic particles with a radius of \( r = 1\mu m \). The particles are placed in a square 100 \times 100 lattice containing 20,000 particles and 10,000 pinning sites or in a hexagonal 38 \times 66 lattice containing 15,048 particles and 10,032 pinning sites. Each pinning site is a double-well trap and the lattice containing 15,048, 048 particles and 10,032 pinning sites. Each pinning site is a double-well trap and the lattice containing 15,048, 048 particles and 10,032 pinning sites. Each pinning site is a double-well trap and the lattice containing 15,048, 048 particles and 10,032 pinning sites.

The elongated gravitational double-well traps are modeled as two spherical quarters connected by an elongated half-cylindrical trough of length 2µm in the square ice and 1.4µm in the hexagonal ice that has a repulsive bump in the middle. Each minimum of the double well is located at the end of the elongated trough, coinciding with the minimum in the spherical quarter. When the particle is in either of the spherical ends, an unbreakable harmonic spring force tethers the particle to the minimum with a spring constant of \( k = 0.222 \) pN/µm for the square ice and \( k = 2.22 \) pN/µm for the hexagonal ice. When the particle is in the elongated part of the pin, the same unbreakable harmonic spring force acts on it in the direction perpendicular to the elongated trough, and an additional force is exerted by the bump in the middle of the trough which has a maximum value of \( F_{sm} = 0.011 \) pN for the square ice and \( F_{sm} = 0.211 \) pN for the hexagonal ice. This force decays to zero linearly in each half of the elongated trough as the intersection with the spherical quarters is approached. These forces together compose the substrate force denoted as \( F^s_i \).

We use a smaller lattice constant for the less densely packed hexagonal ice because stronger inter-particle interactions are required to induce the spin ice ordering compared to the square ice system. We also increase the pinning strength significantly for the hexagonal ice to prevent the particles from ordering into a triangular lattice with each particle sitting at the center of the elongated trough, which destroys the spin ice nature of the particle based system. With the chosen values, which are within the experimentally realizable regime, the spin ice manifold is preserved.

Magnetization of the particles in the \( z \) direction by an external magnetic field produces a repulsive particle-particle interaction force \( F_{pp}(r) = A_c/r^4 \) with \( A_c = 3 \times 10^9 \chi_m^2 V^2 B^2/(2\pi\mu) \) for particles a distance \( r \) apart. Here \( \chi_m \) is the magnetic susceptibility, \( V \) is the particle volume, \( B \) is the magnetic field in mT, and all distances are measured in µm. At \( B = 40 \) mT, the maximum field we consider, this gives \( F_{pp} \approx 0.49 \) pN for \( r = 3 \) µm, which is a typical distance for the square ice, and \( F_{pp} \approx 6.05 \) pN for \( r = 1.6 \) µm, which is a typical distance for the hexagonal ice. The dynamics of colloid \( i \) are obtained using the following discretized overdamped equation of motion:

\[
\frac{1}{\mu} \frac{\Delta r_i}{\Delta t} = \sqrt{\frac{2}{D\Delta t}} k_B T N[0, 1] + F_{pp}^i + F_s^i \tag{1}
\]

Here the diffusion constant \( D = 3600 \text{ nm}^2/\text{s} \), the mobility \( \mu = 8.894 \mu\text{m/s/pN} \) and the simulation time step \( \Delta t = 1\mu\text{s} \). The first term on the right is a thermal force consisting of Langevin kicks of magnitude \( F_T = 0.954 \) pN corresponding to a temperature of \( t = 20^\circ\text{C} \). Here, \( N[0, 1] \) denotes a random number drawn from a normal (Gaussian) distribution with a mean of 0 and a standard deviation of 1. Each trap is filled with a single particle which is randomly placed in one of the two minima. We increase \( B \) linearly from \( B = 0 \) mT to \( B = 40 \) mT, following a procedure that is feasible to achieve experimentally. We average the results over 100 simulations performed with different random seeds.

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