

# Time reversal symmetry breakdown in normal and superconducting states in frustrated three-band systems

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We discuss the phase diagram and phase transitions in  $U(1) \times \mathbb{Z}_2$  three-band superconductors with broken time reversal symmetry. We find that beyond mean field approximation and for sufficiently strong frustration of interband interactions there appears an unusual metallic state precursory to a superconducting phase transition. In that state, the system is not superconducting. Nonetheless, it features a spontaneously broken  $\mathbb{Z}_2$  time reversal symmetry. By contrast, for weak frustration of interband coupling the energy of a domain wall between different  $\mathbb{Z}_2$  states is low and thus fluctuations restore broken time reversal symmetry in the superconducting state at low temperatures.

In recent years, the discovery of multiband superconductors such as the Iron Pnictides [1], has generated much interest for multiband superconducting systems. From a theoretical viewpoint, one of the main reasons for the strong interest is that in contrast to previously known two-band materials, iron-based superconductors may exhibit dramatically different physics due to the possibility of *frustrated* inter-band Josephson coupling originating with more than two bands crossing the Fermi-surface.[2–5]. In two-band superconductors the Josephson coupling locks the phase differences between the bands to 0 or  $\pi$ . By contrast, if one has three bands and the frustration of interband coupling is sufficiently strong, the ground state interband phase difference can be different from 0 or  $\pi$ . This leads to a superconducting state which breaks time reversal symmetry (BTRS) [2, 3]. From a symmetry viewpoint such a ground state breaks  $U(1) \times \mathbb{Z}_2$  [4]. Recently, such a scenario has received solid theoretical support [5] in connection with hole-doped  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ . The possibility of this new physics arising also in other classes of materials is currently under investigation [6]. For other scenarios of time reversal symmetry breakdown in iron-based superconductors discussed in the literature, see [7, 8].

Three band superconductors with frustrated interband Josephson couplings have a number of properties radically different from their two-band counterparts. These include (I) the appearance of a massless so-called Leggett mode at the  $\mathbb{Z}_2$  phase transition [9]; (II) the existence of new mixed phase-density collective modes in the state with broken time-reversal symmetry (BTRS) [4, 5, 10] in

contrast to the “phase-only” Leggett collective mode in two-band materials [11]; and (III) the existence of (meta-)stable excitations characterized by  $\mathbb{CP}^2$  topological invariants [12, 13].

So far the phase diagram of frustrated three-band superconductors has been investigated only at the mean-field level [3, 5]. However, the iron-based materials feature relatively high  $T_c$ , as well as being far from the type-I regime. Hence, one may expect fluctuations to be of importance.

In this paper, we study the phase diagram of a three-band superconductor in two-dimensions in the London limit, beyond mean-field approximation. The results should apply to relatively thin films of iron-based superconductors where, owing to low dimensionality, fluctuation effects are particularly important. The main findings of this work are as follows. (I) When the frustration is sufficiently strong, the phase diagram acquires an unusual fluctuation-induced metallic state which is a precursor to the BTRS superconducting phase. This metallic state exhibits a broken  $\mathbb{Z}_2$  time-reversal symmetry. A salient feature is that, although the state is metallic and non-superconducting, it nevertheless features a persistent interband Josephson current in momentum space which breaks time reversal symmetry. (II) When the frustration is weak (i.e. when phase differences are only slightly different from 0 or  $\pi$ ) we find that the system can undergo a fluctuation driven restoration of the  $\mathbb{Z}_2$  symmetry at very low temperatures.

The London model for a three-band superconductor is given by

$$F = \sum_{\alpha=1,2,3} \frac{|\psi_\alpha|^2}{2} (\nabla\theta_\alpha - e\mathbf{A})^2 + \sum_{\alpha,\alpha'>\alpha} \eta_{\alpha\alpha'} |\psi_\alpha| |\psi_{\alpha'}| \cos(\theta_\alpha - \theta_{\alpha'}) + \frac{1}{2} (\nabla \times \mathbf{A})^2. \quad (1)$$

Here,  $|\psi_\alpha| e^{i\theta_\alpha}$  denotes the superconducting condensate

components in different bands labeled by  $\alpha = 1, 2, 3$ ,

while the second term represents interband Josephson couplings. The field  $\mathbf{A}$  is the magnetic vector potential

$$F = \frac{1}{2\varrho^2} \sum_{\alpha} (|\psi_{\alpha}|^2 \nabla \theta_{\alpha} - e |\psi_{\alpha}|^2 \mathbf{A})^2 + \frac{1}{2} (\nabla \times \mathbf{A})^2 + \sum_{\alpha, \alpha' > \alpha} \frac{|\psi_{\alpha}|^2 |\psi_{\alpha'}|^2}{2\varrho^2} [\nabla(\theta_{\alpha} - \theta_{\alpha'})]^2 + \eta_{\alpha\alpha'} |\psi_{\alpha}| |\psi_{\alpha'}| \cos(\theta_{\alpha} - \theta_{\alpha'}), \quad (2)$$

where  $\varrho^2 = \sum_{\alpha} |\psi_{\alpha}|^2$ . This shows that the vector potential is coupled only to the  $U(1)$  sector of the model, and not to phase differences.

When the Josephson couplings  $\eta_{\alpha\alpha'}$  are positive, each Josephson term by itself prefers to lock phase difference to  $\pi$ , i.e.  $\theta_{\alpha} - \theta_{\alpha'} = \pi$ . Since this is not possible for three phases, the system is frustrated. The system breaks time reversal symmetry when Josephson couplings are minimized by two inequivalent phase lockings, shown in Fig. 1. The phase lockings are related by complex conjugation of the fields  $\psi_{\alpha}$ . Thus, by choosing one of these phase locking patterns the system breaks time reversal symmetry [2–4].

In this work, we address the phase transitions in a two dimensional three-band superconductor with broken time-reversal symmetry. A Berezinskii-Kosterlitz-Thouless (BKT) phase transition in  $U(1)$  systems is driven by proliferation of vortex-antivortex pairs, while an Ising phase transition is driven by proliferation of  $\mathbb{Z}_2$  domain walls. The nontriviality of the problem of phase transitions in the three-band model is due to the spectrum of topological excitations of the model. Firstly, the model features singly-quantized composite vortices where all phases wind by  $2\pi$ , i.e.  $\Delta\theta_1 \equiv \oint \nabla\theta_1 = 2\pi, \Delta\theta_2 = 2\pi, \Delta\theta_3 = 2\pi$ . We will denote them (1,1,1). As is clear from Eq. (2), such a vortex has topological charge only in the  $U(1)$  sector of the model. It has no phase winding in the phase differences and thus does not carry topological charge in  $\mathbb{Z}_2$  sector. In addition, the model features other topological defects discussed in detail in Refs. 12 and 13. These are  $\mathbb{Z}_2$  domain walls (several solutions with different energies), fractional-flux vortices with linearly divergent energy, as well as  $CP^2$  skyrmions which are combined vortex-domain wall defects carrying topological charges in both the  $U(1)$  and  $\mathbb{Z}_2$  sectors of the model. This spectrum of topological excitations distinguishes this model from other  $U(1) \times \mathbb{Z}_2$  systems, like e.g.  $XY$ -Ising model [14]. The model is also principally different from  $[U(1)]^3$  superconductors, since in such systems fractional vortices have logarithmically divergent energy and thus drive BKT phase transitions [15, 16].

In two dimensions the effective magnetic field penetration length is inversely proportional to the film thickness [17]. We thus begin by discussing the limit of very large penetration length, in which we may neglect the coupling

that couples minimally to the charged condensate matter fields. By collecting gradient terms for phase differences, it can also be rewritten as

to the vector potential. We discuss the phase diagram of the model in the case of a finite penetration length in our summary.

The partition function of the lattice version of the model (1) reads

$$Z = \prod_{\alpha, i} \left[ \int_{-\pi}^{\pi} \frac{d\theta_{\alpha, i}}{2\pi} \right] \exp(-\beta H), \quad (3)$$

where the Hamiltonian is given by

$$H = - \sum_{\langle i, j \rangle, \alpha} \cos(\theta_{\alpha, i} - \theta_{\alpha, j}) + \sum_{i, \alpha' > \alpha} g_{\alpha\alpha'} \cos(\theta_{\alpha, i} - \theta_{\alpha', i}). \quad (4)$$

$i, j \in \{1, 2, \dots, N = L \times L\}$  denote sites on a lattice of size  $L \times L$  and  $\langle i, j \rangle$  indicates nearest neighbor lattice sites (assuming periodic boundary conditions).  $\beta$  is the (properly rescaled) coupling (“inverse temperature”) and  $g_{\alpha\alpha'}$  are interband Josephson couplings. Here, we consider the case of similar prefactors for the three gradient terms.

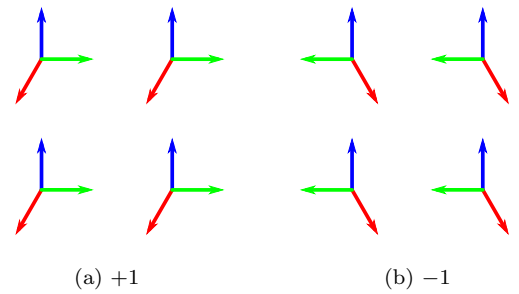


FIG. 1. (Color online) Examples of phase configurations for the two  $\mathbb{Z}_2$  symmetry classes of the ground states on sites of  $2 \times 2$  lattice. Here  $g_{12} > g_{23} > g_{13} > 0$ . The arrows ( $\rightarrow$ ,  $\leftarrow$ ,  $\rightarrow$ ) corresponds to  $(\theta_1, \theta_2, \theta_3)$ .

Algebraically decaying correlations and frustration effects typically render two dimensional  $U(1) \times \mathbb{Z}_2$ -symmetric models difficult to investigate numerically through equilibrium Monte Carlo (EMC) simulations [18]. In this work we used a non-equilibrium approach, namely that of short time critical dynamics (STCD). See

e.g. the review articles 19 and 20, and references therein. See online supplementary material for details.

First, we consider the case  $g_{12} = g_{23} = g_{31} = g$ , which is shown in Fig. 3. The phase transitions for the  $\mathbb{Z}_2$  and U(1) symmetries are close, but clearly separated for all values of  $g$ . This means that beyond the mean-field approximation there appears a new phase. As the temperature increases from the low-temperature maximally ordered phase, an unbinding of vortex-antivortex pairs of composite vortices first takes place. In the resulting state the free composite vortices  $(1, 1, 1)$  and  $(-1, -1, -1)$  do not further decompose into fractional vortices  $(1, 0, 0)$ ,  $(0, 1, 0)$ ,  $(0, 0, 1)$  because Josephson coupling provides linear confinement of the constituent fractional vortices [13]. Due to this confinement, the proliferation of  $(1, 1, 1)$  and  $(-1, -1, -1)$  vortex-antivortex pairs disorders only the U(1) sector of the model described by the first term in Eq. (2). However, these defects do not restore  $\mathbb{Z}_2$  symmetry. The resulting state is non-superconducting with broken time-reversal symmetry. This sector is described by the last terms in Eq. (2),

$$F_{\mathbb{Z}_2} = \sum_{\alpha, \alpha' > \alpha} \left\{ \frac{|\psi_\alpha|^2 |\psi_{\alpha'}|^2}{2\rho^2} [\nabla(\theta_\alpha - \theta_{\alpha'})]^2 + \eta_{\alpha\alpha'} |\psi_\alpha| |\psi_{\alpha'}| \cos(\theta_\alpha - \theta_{\alpha'}) \right\} \quad (5)$$

Secondly, at higher temperatures the  $\mathbb{Z}_2$  domain walls proliferate and restore the symmetry completely. The physical interpretation of this precursor normal state with broken time reversal symmetry is as follows. In the BTRS superconducting state there is a ground state phase difference other than 0 or  $\pi$  between components. This implies the existence of persistent interband Josephson currents. Two different  $\mathbb{Z}_2$  phase locking patterns mean that there are two inequivalent interband Josephson current “loops in  $\mathbf{k}$ -space”. Namely, one loop is of the type band 1  $\rightarrow$  band 2  $\rightarrow$  band 3  $\rightarrow$  band 1, the other is of the type band 1  $\rightarrow$  band 3  $\rightarrow$  band 2  $\rightarrow$  band 1. The non-superconducting  $\mathbb{Z}_2$ -ordered phase corresponds to the situation where superconducting phases exhibit exponentially decaying correlations due to proliferation of vortex-antivortex pairs. What sets this state apart from the situation found in conventional superconductors is that the three-band system retains a persistent interband Josephson current in  $\mathbf{k}$ -space which breaks the time reversal symmetry. The phase configurations in this state are illustrated in Fig. 2.

Next, we consider the case of a more general model where the Josephson couplings are different. By tuning some of the Josephson couplings one can make the difference between two out of three phases arbitrarily small in the BTRS ground state. This also implies that the energy of  $\mathbb{Z}_2$  domain walls can be made arbitrarily small. Thus, one can interchange critical temperatures of U(1)

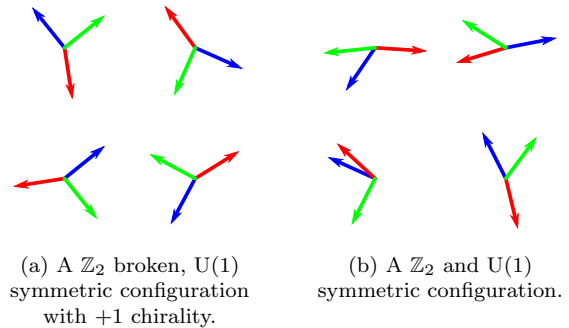


FIG. 2. (Colors online) A schematic illustration of phase configurations in the normal state which break time reversal symmetry vs the normal state which does not. Here,  $g_{12} > g_{23} > g_{13} > 0$ . The arrows ( $\rightarrow$ ,  $\rightarrow$ ,  $\rightarrow$ ) correspond to  $(\theta_1, \theta_2, \theta_3)$ .

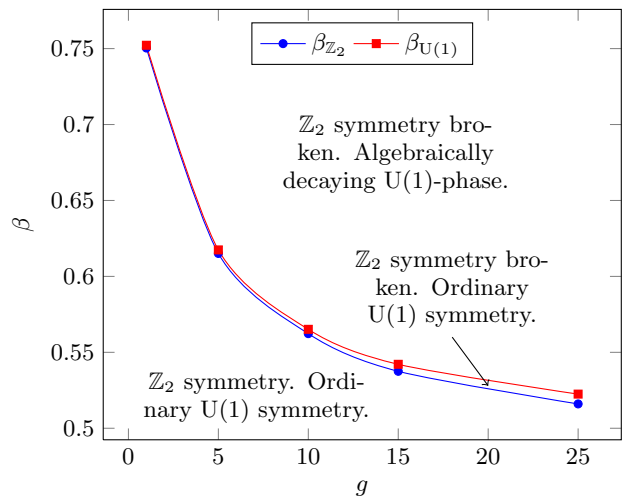


FIG. 3. (Color online) Phase diagram for the three-band model with  $g_{12} = g_{23} = g_{31} = g$ .  $g \in [1, \dots, 25]$ . The  $\beta_{U(1)}$  line lies above the  $\beta_{\mathbb{Z}_2}$  line for the investigated values of  $g$ . Error bars are smaller than symbol sizes. Lines are guides to the eye.

and  $\mathbb{Z}_2$  phase transitions. Moreover, inclusion of fluctuations can in a certain limit dramatically suppress the critical temperature of the  $\mathbb{Z}_2$  phase transition. Results from Monte-Carlo simulations shown in Fig. 4 display such behavior.

Finally, consider the effect of a finite penetration length. As can be seen from Eq. (2), the gauge field couples only to the U(1) sector of the model, making the U(1) symmetry local. It also makes the energy of  $(1, 1, 1)$  and  $(-1, -1, -1)$  finite [13]. As a result, at any finite temperature, there is a finite probability of exciting such topological defects, which from a formal viewpoint suppresses superconductivity at finite temperature in the thermodynamic limit. In a real experiment on a finite system, with large but finite penetration length, this

physics manifests itself as a conversion of a BKT transition to a broad crossover which takes place at lower characteristic temperatures than the  $U(1)$  phase transition in the global  $U(1) \times \mathbb{Z}_2$  model. Since, on the other hand  $\mathbb{Z}_2$  phase transition is not directly affected by this coupling, the  $\mathbb{Z}_2$  ordered non-superconducting state persists. Thus, in the thermodynamic limit a system with finite penetration length features  $U(1) \times \mathbb{Z}_2$  superconductivity at zero temperature, while at any nonzero temperature it resides in a  $\mathbb{Z}_2$  metallic state, up to the temperature where the  $\mathbb{Z}_2$ -symmetry is restored.

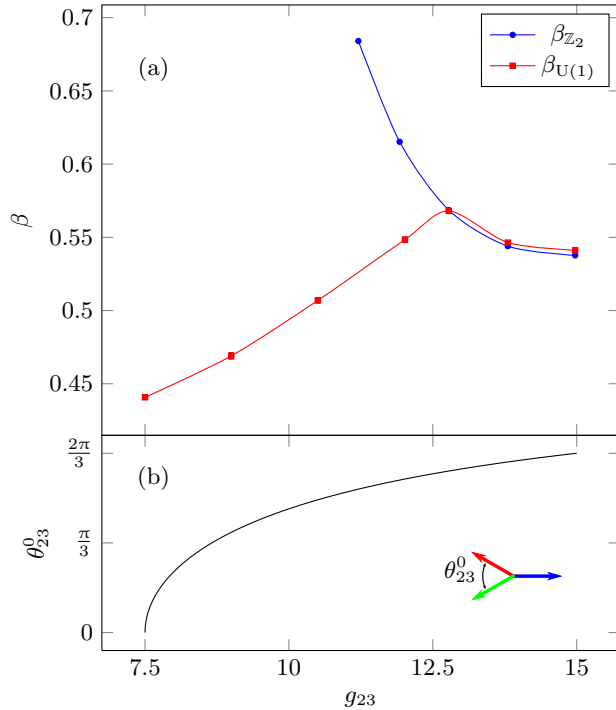


FIG. 4. (a) The phase diagram for the three-band model with unequal Josephson couplings. We set  $g_{12} = g_{13} = 15$  and varied  $g_{23}$ . Error bars are smaller than symbol sizes. Lines are guides to the eye. (b) The phase difference  $\theta_{23}^0$  between band 2 and 3 in the ground state, as defined in the phase vector inset.  $\theta_{23}^0 = 0$  for  $g_{23} < 7.5$ .  $\theta_{12}^0 = \theta_{13}^0 = (2\pi - \theta_{23}^0)/2$  for all  $g_{23}$ .

In conclusion, we have studied the phase diagram of three band superconductors with spontaneously broken time reversal symmetry due to frustrated interband Josephson-couplings, beyond mean field approximation. We have found that there is a new fluctuation-induced  $U(1)$ -symmetric state, which is a precursor to the superconducting state. However, it exhibits a broken  $\mathbb{Z}_2$  (Ising) symmetry. This state is therefore distinct from an ordinary metallic state, due to spontaneously broken time reversal symmetry associated with persistent interband Josephson currents in  $\mathbf{k}$ -space. Experimentally, it can be distinguished from superconducting and ordinary normal states by a combination of local (e.g. tunnel-

ing) and transport measurements (with later showing no superconductivity). Another way of possibly detecting this state would be by observing an Onsager anomaly in the specific heat in the normal state. Besides that the existence of broken  $\mathbb{Z}_2$  symmetry in the normal state implies the existence of mixed “phase difference-density” collective modes previously discussed in the superconducting state [4, 5, 10]. For interband coupling values where these modes exist, they may thus be observed in a similar way as the Leggett mode [21]. On the other hand, in the regime where a ground state phase differences are very close to 0 or  $\pi$ , we find that fluctuations can restore the  $\mathbb{Z}_2$  symmetry at substantially lower temperatures than those which are predicted by mean-field theory. These predictions could be used also to verify if  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  breaks time reversal symmetry at certain doping (which as recently discussed in [5] is a strong candidate for a BTRS superconductor).

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**Online supplementary material: Details about the simulations in the paper “Frustration, time-reversal symmetry breaking, and unusual metallic states in a three-band superconductor”.**

We used short time critical dynamics (STCD) to investigate the three band model. We briefly describe the procedure here, and refer to the review articles 19 and 20 and references therein for more details on this method.

The STCD method is based on the observation that when a system, initially prepared in either the ground state or in a random state, is released under a Monte Carlo (MC) dynamics of local updates, the MC time  $t$  evolution of the mean of a local observable will be very sensitive to the coupling constant of the dynamics. Typically (and this is what we have done here) one looks at some kind of magnetization  $m(t)$  when heating the ground state, as this tend to give more accurate results than cooling down a random (“infinite temperature”) initial state. When the coupling differs from the critical coupling  $\beta_c$  (which turns out to be the same as the *equilibrium* critical coupling), the magnetization goes exponentially to either zero or a constant value as  $t \rightarrow \infty$ , depending on whether the coupling is sub- or supercritical. If the coupling is critical,  $m(t) \sim t^a$ , where  $a$  is a

constant which can be related to *equilibrium* critical constants. Thus, by examining the behavior of  $m(t)$  around  $\beta_c$ , it is relatively straightforward to determine the critical point to great precision.

In practise, the time,  $t_{\text{micro}}$ , after which the  $t \rightarrow \infty$  behavior starts to emerge, is rather short: For simple non-frustrated systems it is typically in the order of tens of MC sweeps; in frustrated systems somewhat longer, a couple of orders of magnitudes more. The advantage of STCD simulations lies in the short time scale (compared to EMC). Thermalization sweeps are not necessary, and, given sufficiently large system sizes, the finite size correlation effects are exponentially damped within the simulation time, effectively making the system behave as in the thermodynamic limit. The last phenomenon can be understood from the fact that in the short time regime, correlations have not had sufficient time to expand through the entire system. Furthermore, the dynamics being far from equilibrium means that the equilibrium problem of critical slowing down is avoided, rendering the big system sizes needed for the longer simulation times of frustrated models unproblematic.

In the simulations of the three band model we have used two “magnetizations”, one for each symmetry, as local order parameters. For the  $\mathbb{Z}_2$  symmetry, we choose the mean Ising magnetization,  $m_{\mathbb{Z}_2} \equiv N^{-1} \sum_i \sigma_i$ , as order parameter. Here  $\sigma_i \equiv 1$  if the local phases are distributed as  $1 \rightarrow 2 \rightarrow 3 \rightarrow 1$  (going counterclockwise), and  $\sigma_i \equiv -1$  if they are distributed as  $1 \rightarrow 3 \rightarrow 2 \rightarrow 1$ . The  $U(1)$  symmetry order parameter is chosen to be the average of the absolute value of the mean global  $XY$  vector sum of each component,  $m_{U(1)} \equiv (3N)^{-1} \sum_\alpha |\sum_i \mathbf{s}_{\alpha,i}|$ , where  $\mathbf{s}_{\alpha,i} \equiv [\cos(\theta_{\alpha,i}), \sin(\theta_{\alpha,i})]$ . It should be noted that the magnetization curves used to obtain the results are average values of several, independent runs.

The procedure of finding the critical coupling of  $\mathbb{Z}_2$  symmetry, called  $\beta_{\mathbb{Z}_2}$ , is rather straightforward: For a given set of Josephson couplings, we start looking for the approximate critical coupling by performing some test simulations for a wide range of  $\beta$ -values, manually searching for the best power-law behavior by log-log plotting the magnetization curves. From this it is easy to obtain a relatively small interval containing  $\beta_{\mathbb{Z}_2}$ . Then we perform heavier simulations (bigger system size, larger maximal MC time,  $t_{\text{max}}$ , more runs) at  $\beta$  values spread out in this interval. Finally we use quadratic splines to interpolate between the obtained magnetization curves and search for the  $\beta$ -value which corresponds to the best power-law fit using nonlinear regression. We have used the standard deviation of the residuals as a goodness-of-fit parameter for this optimization problem.

The time correlation  $c_{\Delta t}(t) \equiv \langle m(t)m(t+\Delta t) \rangle$  is at the critical point expected to follow a power law as well,  $c_{\Delta t} \sim t^b$ . To counteract this, we let the time intervals between the simulation samplings follow a power too, keeping all the points of the regression approximately equally

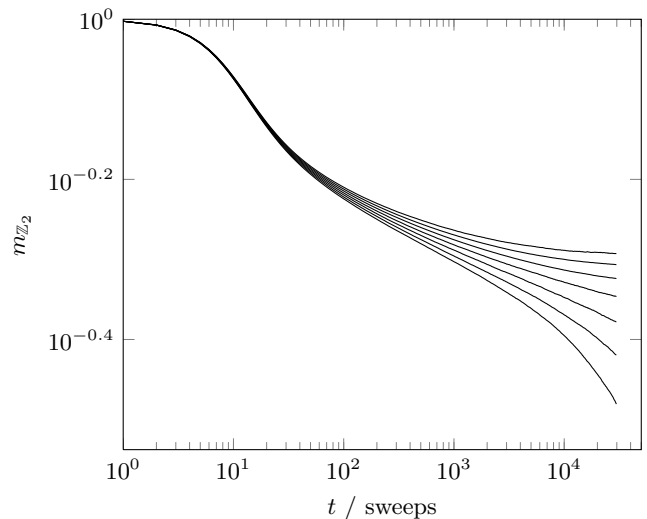


FIG. 5. log-log plot of some  $m_{\mathbb{Z}_2}(t)$ -curves for  $g = 1$  at, from below,  $\beta = 0.748, 0.749, \dots, 0.754$ . The critical coupling is determined to be  $\beta_{\mathbb{Z}_2} \approx 0.75039$ . The error is smaller than the line width.  $t_{\text{micro}} \sim \mathcal{O}(10^3)$ .

correlated.

An example of  $m_{\mathbb{Z}_2}(t)$ -curves of a STCD simulation is shown in Fig. 5.

Finding  $\beta_{U(1)}$ , the BKT transition point of the  $U(1)$  symmetry, is in many aspects similar to the  $\mathbb{Z}_2$  procedure. The main difference is that we now have to deal with the entire low temperature phase displaying power law behavior in  $m_{U(1)}(t)$ , making the search for the transition coupling more challenging. In this work we have used a technique from Ref. 22. We noted that in this case the method was less numerical stable than the  $\mathbb{Z}_2$  critical point search explained above, requiring more relying on manual initial parameter guesses. Therefore the error bars for the  $U(1)$  line in the phase diagrams of this work are probably a bit too small, but not so much that any qualitative conclusion could be changed.

An example of  $m_{U(1)}(t)$ -curves of a STCD simulation is shown in Fig. 6.

A system size of  $L = 2048$  (grid parallelized and distributed over several CPUs) was used for all results; tests against  $L = 1024$  and  $L = 4096$  simulations confirmed that this system size was sufficiently large for any finite size effects to be negligible at the Monte Carlo time scale used in the simulations.

In the determination of  $\beta_{\mathbb{Z}_2}$  and  $\beta_{U(1)}$  only data for  $t > 5000$  sweeps was used in order to avoid the non-universal microscopic time regime.  $t_{\text{max}} = 30000$  sweeps was used for the determination of all  $\beta_{\mathbb{Z}_2}$  values,  $\beta_{U(1)}$  values for the  $g_{12} = g_{23} = g_{13}$  simulations and for  $g_{23} > 12.5$  in the  $g_{12} = g_{23} \neq g_{13}$  simulations.  $t_{\text{max}} = 50000$  was used for  $g_{23} < 12.5$  in the  $g_{12} = g_{23} \neq g_{13}$  simulations.

Pseudorandom numbers were generated by the Mersenne-Twister algorithm [23]. Errors were deter-



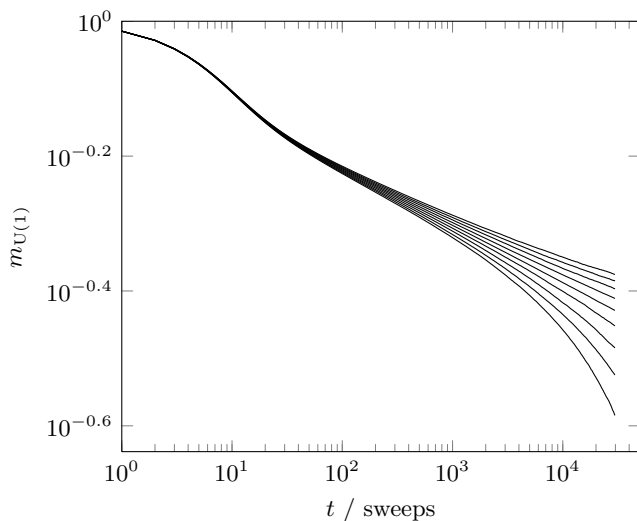


FIG. 6. log-log plot of some  $m_{U(1)}(t)$ -curves for  $g = 1$  at, from below,  $\beta = 0.748, 0.749, \dots, 0.756$ .  $\beta_{U(1)} \approx 0.752\,20$ . The error is smaller than the line width.  $t_{\text{micro}} \sim \mathcal{O}(10^3)$ .

mined using the jackknife method.

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