Nonequilibrium critical relaxation at a first-order phase transition point

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Abstract – We study numerically the nonequilibrium dynamical behavior of an Ising model with mixed two-spin and four-spin interactions after a sudden quench from the high-temperature phase to the first-order phase transition point. The autocorrelation function is shown to approach its limiting value, given by the magnetization in the ordered phase at the transition point, \( m_c \), through a stretched exponential decay. On the other hand relaxation of the magnetization starting with an uncorrelated initial state with magnetization, \( m_i \), approaches either \( m_c \), for \( m_i > 0.5 \), or zero, for \( m_i < 0.5 \). For small \( m_i \) and for \( m_i \) slightly larger than 0.5 the relaxation of the magnetization shows an asymptotic power law time dependence, thus from a nonequilibrium point of view the transition is continuous.

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Introduction. – Most of our knowledge about the properties of phase transitions has accumulated for continuous or second-order transitions for which the concepts of scaling and universality, as well as the application of the method of renormalization group, have provided a deep understanding [1]. Considerably less is known about singularities at first-order phase transitions, although in nature this type of transition is very common [2]. At a first-order phase transition point one observes phase coexistence where several thermodynamical quantities, such as the internal energy and the order parameter (magnetization), have a discontinuity, whereas the correlation length is generally finite. In spite of this finite correlation length some response functions, such as the magnetic susceptibility, have a scaling behavior in a finite system [3] that involves the discontinuity fixed-point exponent \( y_d = d \) where \( d \) denotes the dimension of the system [4]. From a dynamical point of view the relaxation time is finite at a first-order transition point, yielding equilibrium autocorrelations and relaxation functions that decay exponentially.

Many studies have been devoted to the investigation of the formation of the ordered phase when the temperature is lowered below the phase transition point, \( T_c \). Thus, the nonequilibrium dynamics of the nucleation process [5] and the morphology of the solidification [6] are key problems in material science. From a more theoretical perspective one is interested in the coarsening process [7] that takes place when the system is suddenly quenched from the high-temperature phase at or below the phase transition point. If the quench is performed below \( T_c \), then the competition between the stable ordered phases leads to the same type of qualitative picture independently of the order of the transition at \( T_c \) [8]. If, however, we quench the system at the very transition point, important differences are expected in the two cases due to the different behavior of the correlation length.

In the past this type of phenomena, i.e. nonequilibrium critical relaxation, has been thoroughly studied in the case of a second-order transition point [9,10], whose investigations were also motivated by the appearance of ageing. On the contrary only little attention has been paid to the case when the transition is of first order. It has been demonstrated that nonequilibrium relaxation starting from an ordered or a mixed-phase initial state can be used to locate accurately the phase transition point and to decide about the order of the transition [11,12]. For models with quenched disorder the tricritical value of
the dilution has also been studied by this method [13]. However, we are not aware of any study in which the singularities in the dynamical quantities are systematically investigated. In this paper we perform an investigation of the nonequilibrium dynamics at a first-order phase transition that addresses questions well established for a second-order transition point. In particular we consider the asymptotic behavior of the autocorrelation function as well as the relaxation of the order parameter after a sudden quench from the high-temperature phase to the first-order transition point.

The structure of the paper is the following. After a brief recapitulation of the known results of nonequilibrium critical relaxation we present a detailed numerical study performed on a square lattice Ising model with mixed two-spin and four-spin product interactions [14,15]. This system has a first-order phase transition and the location of the transition point is known by duality. The numerical results obtained in this model are consistent with the existence of a divergent nonequilibrium relaxation time. We interpret its origin and argue that the observed nonequilibrium dynamical behavior is generic at first-order phase transitions.

**Nonequilibrium critical dynamics: a reminder.** – At a second-order transition point both the correlation length, $\xi$, and the relaxation time, $\tau$, are divergent, and they are related through: $\tau \sim \xi^\theta$. The dynamical exponent, $\theta$, which generally depends on the local dynamics, on conservation laws, and on symmetries, is enough to classify the dynamical universality class at equilibrium [16]. In a nonequilibrium situation, when the system is quenched from the high-temperature phase as an initial state to the critical point, new nonequilibrium exponents have to be defined [17,18]. The reason for this is the broken time translation invariance due to a discontinuity at the time horizon (“time-surface”). As a result the autocorrelation function, $C(s,t)$, is generally non-stationary: it depends on both the waiting time, $s$, and the observation time, $t$. In the limit $t \gg s$ we have asymptotically: $C(s,t) \sim t^{-\lambda/\nu}$, which involves the nonequilibrium autocorrelation exponent $\lambda$ [18]. In another related process the initial state is prepared with a small, non-vanishing magnetization, $m_i$, and one measures its relaxation, which for short times behaves as $m(t) \approx m_i t^\theta$. Here the initial slip exponent $\theta$ satisfies the scaling relation: $\lambda = d - \theta \nu$ [9,17]. We note that in mean-field theory $\theta = 0$, whereas in real systems the exponent $\theta$ is generally larger than zero and can be expressed as $\theta = (x_1 - x) / \nu$, where $x = \beta / \nu$ is the anomalous dimension of the bulk magnetization ($\beta$ and $\nu$ are the standard magnetization and correlation length exponents, respectively) and $x_1$ is called the anomalous dimension of the initial magnetization.

**Ising model with multispin interaction.** – At a first-order transition point the correlation length stays finite, $\xi < \infty$, and the same is true for the equilibrium relaxation time, $\tau < \infty$. The magnetization displays a jump from zero above $T_c$ to some value $m_c > 0$ below $T_c$. In this case the static critical exponents are formally given by $x = \beta = 0$ and $\nu = 1 / d$, where the latter is the discontinuity fixed-point value [4]. The specific model we study numerically in the following belongs to a class of square lattice Ising models with two-spin interactions in the vertical direction and $n$-spin product interactions in the horizontal direction that is defined by the Hamiltonian

$$H = - \sum_{i,j} \left( J_2 \sigma_{i,j} \sigma_{i,j+1} + J_n \prod_{k=0}^{n-1} \sigma_{i+k,j} \right).$$

The model is self-dual [14,15,19] and the self-dual point is located at: $\sinh(2J_2/kT_c) \sinh(2J_n/kT_c) = 1$. For $n = 2$ we of course recover the standard Ising model. Whereas for $n = 3$ the transition is found to be continuous and to belong to the universality class of the four state Potts model [20–25], for $n \geq 4$ the transition is of first order [26,27]. In the following we consider the model with four-spin interaction, in which case the ordered phase has eightfold degeneracy$^1$ and the phase transition of the system is comparable with that in the eight-state Potts model. If one takes $J_2 = J_3$, numerical results [27] indicate a latent heat of $\Delta/kT_c = 0.146(3)$ and a jump of the magnetization from zero to $m_c = 0.769(6)$. The correlation length has not been measured in this system, but from the snapshots of the simulations one expects somewhat different correlation lengths in the horizontal and in the vertical directions that are both of the order of ten lattice constants.

**Single-spin autocorrelation function.** – The single-spin autocorrelation function of the model is defined as $C(s,t) = \langle \sigma_{i,j}(s) \sigma_{i,j}(t) \rangle$, which is then averaged over all spins, $1 \leq i, j \leq L$, and periodic boundary conditions are applied in both directions. Starting with a disordered initial state, which corresponds to $T = \infty$, we apply non-conserving spin-flip dynamics at the temperature $T = T_c$. In the following we consider no waiting time, i.e. $s = 0$, and use the notation: $C(0,t) \equiv C(t)$. In order to monitor finite-size effects we simulated systems containing $L^2$ spins with $L$ ranging from 60 to 640. For the largest size we averaged over 2000 different samples with different realizations of the noise, more samples were simulated for smaller system sizes. As usual, time is measured in Monte Carlo steps where on average every spin is flipped once per step. The calculated autocorrelation functions for different finite systems are plotted in fig. 1. Rather large systems are needed in the calculation in order to handle the finite-size effects. From fig. 1a we can identify three different time regimes. i) For short times, which are shorter than the equilibrium relaxation time, $t < \tau$, there is a fast decay of $C(t)$, which is described by a stretched

$^1$The ordered states have a strip-like structure build from $n = 4$ units in the direction of the four-spin interaction. These are $+++ +, + + - - , + - - + , - + - +$ and four others by interchanging $+$ and $-$. In the following we refer to the homogeneous state with $++ + +$ as the ordered state.
exponential function. ii) This initial decay is followed by a relatively slow approach to the equilibrium limiting value \( \lim_{t \to \infty} \lim_{L \to \infty} C(t) = m_c \). Indeed the plateau of \( C(t) \) in fig. 1a gives an accurate estimate \( m_c = 0.770(3) \) in accordance with previous numerical results [27]. For the largest finite system we have checked the behavior of the connected autocorrelation function and we have obtained a stretched exponential form: \( C(t) - m_c \sim \exp(-bt^a) \), with an exponent \( a = 0.198(3) \), see fig. 1b. We note that in the short-time regime the decay is described by a somewhat larger effective exponent in the stretched exponential, \( a \approx 0.32 \). iii) The third regime of \( C(t) \) is seen in finite systems [28], in which it starts to decrease from the saturation value after passing a cross-over time: \( t_L \sim L^2 \). The dynamical exponent measured in this part of the figure is found to be \( z = 2.05(10) \), which is consistent with \( z = 2 \).

At this point we can conclude that for large systems the nonequilibrium autocorrelation function at a first-order transition point approaches the bulk magnetization, \( m_c \). Thus although the average magnetization of the system is vanishing (see the next section) the dynamical correlations display a finite asymptotic value. On the other hand the connected autocorrelation function has an asymptotic stretched exponential decay.

**Relaxation of the magnetization.** – Next we investigate the relaxation of the magnetization at \( T_c \) where we start with an uncorrelated initial state with a magnetization \( m_i \leq 1 \). In order to obtain reliable data for this noisy quantity, we averaged over typically 250000 different samples with different initial states and different realizations of the noise. The data discussed in the following have been obtained for systems containing \( 240^2 \) spins. We checked that the presented data are free of finite-size effects by simulating some larger systems for selected values of the initial magnetization \( m_i \).

The results are shown in fig. 2 for different values of the initial magnetization. Here one can observe two fixed points concerning the limiting value of \( \lim_{t \to \infty} m(t) \equiv m_\infty \), see figs. 2a and b. For large enough initial order, \( m_i > m^* \), the magnetization approaches its finite equilibrium value in the ordered phase, \( m_\infty = m_c \), whereas for weak initial order, \( m_i < m^* \), the limiting magnetization is vanishing: \( m_\infty = 0 \). According to our numerical estimates the border between the two regimes is given by \( m^* = 0.500(1) \), and starting the relaxation from this initial value the magnetization approaches a finite limiting value: \( m_\infty = 0.3844(3) \), which is compatible with \( m_c / 2 \). At this special value of \( m_i \) there is first a drop of the magnetization in the early times when nuclei of the coexisting phases are created (resulting in an average magnetization of \( \approx m_c / 2 \)), which afterwards takes part in a special coarsening process during which the coexisting ordered and disordered phases are separated and thus the interface between the two phases are reduced. The evaluation of the structure of the system in time is shown in fig. 3. Here we present also results for \( m_i \ll m^* \), when the mass of the ordered domains and thus the magnetization in the system is continuously decreasing.

In the regime \( m_i > m^* \), the \( t \)-dependence of \( m(t) \) is non monotonic when \( m_i \) is close to \( m^* \): in the early time steps there is a sudden drop of the magnetization due to the formation of disordered clusters, which is then followed
by an increase of $m(t)$ that results from the formation of ordered domains. In this regime the relaxation for \( t > \tau \) is described in a power law form: \( m(t) - m_c/2 \sim (m_i - m^*)(t - \tau)^{\theta} \). Here the measured effective exponent has a strong \( m_i \) dependence and close to \( m_i = m^* \) it seems to approach \( \theta = 0.50(3) \). Starting with the maximally ordered initial state with \( m_i = 1 \) the relaxation of the magnetization is monotonous and follows very closely the decay of the autocorrelation function, \( C(t) \), which is measured by preparing the system in the fully disordered initial state, see fig. 1. As for the autocorrelation function the approach of the limiting value, \( m_\infty = m_c \), is asymptotically given in a stretched exponential form: \( m(t) - m_c \sim \exp(-b t^{a'}) \), and the measured exponent is \( a' = 0.196(5) \), which coincides with \( a \) within the error of the calculation. We can thus conclude that the exponents for the two functions are probably identical and they are given by: \( a = a' = 0.197(4) \). Note that at a second-order transition point \( (m_i = 0) \) the decay with \( m_i = 1 \) is given by \( m(t) \sim t^{-(\xi - 2)/2} \) and our result does not fit with a naive application of the discontinuity fixed-point value, \( x = 0 \).

In the regime with \( m_i < m^* \) and for \( m_i \) close to \( m^* \) the decay seems to be symmetric to the relaxation process observed for \( m_i > m^* \). On the other hand for a small \( m_i/m^* \leq 0.2 \) the data points scales with \( m_i(t)/m_i \), and here we can observe also two different relaxation regions. As shown in fig. 2c, for short times, \( t < \tau \), the decay is very slow, which is well fitted by a logarithmic dependence: \( m(t)/m_i \sim c - d \ln t \), with \( c = 0.7 \) and \( d = 0.1 \). In the long-time limit, \( t > \tau \), the decay becomes faster and is described by a power law form: \( m(t) \sim t^{\theta'} \), with \( \theta' \approx -1 \), as shown in fig. 2d.

**Discussion.** – Nonequilibrium relaxation at a first-order transition point has considerable differences with the case when the quench is performed to a second-order transition point. This is due to the facts that i) the correlation length is finite, which leads to a finite initial time regime, \( t < \tau \), and more importantly ii) in the equilibrium state there is phase coexistence between an ordered phase, having \( m_c > 0 \), and a disordered phase with vanishing magnetization. In the relaxation process the system typically evolves towards one of these phases depending on the magnetization of the uncorrelated initial state, \( m_i \). Just at \( m_i = 0.5 \), which separates the two regimes, the system evolves into phase coexistence. At this special value of \( m_i \) the ordered and disordered phases are separated during a special coarsening process, thus the average magnetization approaches \( m_\infty = m_c/2 \). If the initial magnetization is slightly above the border-line value, \( m_i = 0.5 \), after the initial drop of the magnetization the ordered phase starts to grow around the interface of the extra droplets of the ordered phase. Here we use formally the scaling relation valid at a second-order transition point, \( \theta = (x_i - x)/z \), in which we identify the anomalous dimension of the initial magnetization with the dimension of the interface, \( x_i = d - 1 \), whereas we set \( x = 0 \) at a first-order transition. Consequently we have an initial slip exponent: \( \theta = (d - 1)/z \), which is compatible with the numerical results in \( d = 2 \). In the other case, when the initial magnetization is small, then in the initial state there are no long-range correlations, thus the system has a mean-field character. Since the transition point of the mean-field model is \( T_c(mf) > T_c \), an effective coarsening process takes place during which the magnetic moments of the domains, \( \mu_c \), grow practically with their volume, \( \mu \sim m_i l(t)^d \sim t^{d/z} \). As a consequence the total magnetization of the system decays very slowly with time, slower than any power of time. This coarsening goes on until the linear size of the domains \( l(t) \sim t^{1/z} \) approaches the correlation length \( \xi \). For longer times the magnetic moment of the domains stay constant, \( \mu^* \sim m_i \), so that the magnetization start to decay as \( m(t) \sim t^{-d/z} \), which is compatible with the measured value in \( d = 2 \).

The observed power law dependences of the magnetization are somewhat unexpected, since it means that there is no relevant time scale in the problem. Thus in this sense we have critical nonequilibrium relaxation, even though the equilibrium transition is of first order. To understand this phenomenon we draw an analogy between nonequilibrium
relaxation, i.e. the time dependence of the magnetization, \(m(t)\), after a quench from \(T = \infty\) at \(t = 0\), and the behavior of the equilibrium magnetization, \(m(y)\), in a semi-infinite system measured at a distance, \(y\), from the free surface located at \(y = 0\). In the two systems time, \(t\), and distance, \(y\), play analogous roles and they can be related as \(t \sim y^z\), and similarly: \(\tau \sim \xi^z\). In both systems translational invariance is broken at \(t = 0\) and \(y = 0\), respectively. At a second-order transition point the equivalent initial-slip behaviors are given by: \(m(t) \sim t^{(x_i-x)/z}\), and \(m(y) \sim y^{(x_s-x)}\), respectively, thus the anomalous dimension of the initial magnetization, \(x_i\), and the anomalous dimension of the surface magnetization, \(x_s\), are analogous quantities [29–33]. There is also an analogy when starting with a fully ordered initial state, \(m(t = 0) = 1\), and having an ordered surface, \(m(y = 0) = 1\), when the asymptotic decays are given by \(m(t) \sim t^{-x/z}\) and \(m(y) \sim y^{-x}\), respectively. Now let us see how these analogies work if the transition in the system is of first order. The properties of the surface transition in systems having a first-order transition in the bulk have been studied in the literature [34–39]. Interestingly, the surface transition is of second-order for weak surface fields, \(h_s < h_s^*\), which turns to first-order for \(h_s > h_s^*\), and the two regimes are separated by a tricritical point. This phenomena for \(h_s < h_s^*\) is known as surface induced disorder and is analogous to the wetting transition [40]. Indeed in the surface region the correlation length is divergent, and one should even define two diverging correlation lengths, \(\xi_{\parallel}\) and \(\xi_{\perp}\), which are related as \(\xi_{\parallel} \sim \xi_{\perp}^z\), with an anisotropy exponent, \(\zeta > 1\). Now turning back to the non-equilibrium relaxation process we can interpret our results for small initial magnetization and for \(|m_i - 1/2| \ll 1/2\) as due to a quench-induced disordering effect, so that the non-equilibrium dynamics is critical, although the equilibrium phase transition is of first order. A closer analogy with surface induced disorder is seen if the relaxation starts with \(m_i = 0.5\), where the system, after the initial short time decay, remains at the magnetization \(m_\infty < m_c\) for any finite times. We note that this phenomenon is different to other kinetic analogs of surface-induced disorder [41] and that this type of analogy does not apply to the case of an ordered initial state and an ordered surface. The static magnetization profile at the ordered surface shows an exponential decay to \(m_c\), whereas in the non-equilibrium relaxation problem the decay is found to be stretched exponential.

The results obtained in this paper about the specific model in eq. (1) are presumably generally valid for other systems having a first-order phase transition. In particular the functional form of the relaxation function close to \(m_i = 0\) and \(m_i = 0.5\) are assumed to be described by universal exponents, which could depend only on the dimension of the system. In this respect the type of dynamics (conserving and non-conserving) does matter, e.g. for conserving dynamics the dynamical exponent should become \(z = 3\) instead of 2. On the other hand the stretched exponential form of the autocorrelation function is probably system dependent. It would be interesting to study these questions also for other models.

Fig. 3: Snapshots of the system starting with an uncorrelated initial state having an initial magnetization \(m_i = 0.5\) (upper panels) and \(m_i = 0.1\) (lower panels) at times \(t = 5\), \(t = 100\) and \(t = 500\), from left to the right. The four-spin interaction is given in the vertical direction.
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