Nonequilibrium critical behavior from the master equation

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Kinetic models for nonequilibrium phase transitions have been extensively studied via mean-field theory, series expansions, and Monte Carlo simulation. In this paper we present a different approach based on studying the normal-mode solutions of the master equation for the system to extract information on the static and dynamic behavior of the system in the vicinity of the phase transition. This method provides insight into the physics of the transition and offers an alternative technique for analyzing kinetic phase transitions. We demonstrate the method by extracting the critical behavior of a simple model from finite-size scaling.

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I. INTRODUCTION

An intriguing feature of a continuous thermodynamic phase transition is the existence of universal behavior [1] near the transition characterized by a set of critical exponents. These exponents are universal in the sense that they are essentially independent of all microscopic details except for the symmetry properties of the Hamiltonian, the range of the interaction, and the spatial dimensionality of the system. The ideas of scale invariance and coupling of fluctuations on all length scales [1] provide an elegant picture of the long-wavelength and low-frequency behavior of a system in thermal equilibrium near a phase transition.

In recent years there has been much work devoted to understanding the behavior of a system with many degrees of freedom whose steady state is not a state of thermal equilibrium. These are typically open systems [2] driven by an external force. Examples include lasing [2], fluid turbulence [2], chemical reactions [3], epidemic models [4], and exciton dynamics [5]. These systems can exhibit phase transitions where the gross macroscopic features of the steady state change in a singular manner as the external control parameters are smoothly varied. The behavior of such a nonequilibrium system near a phase transition exhibits many characteristics similar to those seen in thermodynamic phase transitions. Thus it is natural to ask to what extent one can adapt the language and techniques developed in treating equilibrium phase transitions to the nonequilibrium case.

There have been several methods used in the past to study models of nonequilibrium phase transitions. The mean-field approach [6] replaces the complicated many-particle dynamics with a set of evolution equations for smoothed average quantities. The instabilities of the steady-state solutions, or bifurcations, have been extensively investigated [2,6]. As in the equilibrium case, we expect the mean-field approximation to fail in the vicinity of the transition if large-scale fluctuations are present. In order to go beyond the mean-field approximation, recent studies have employed series expansions [7] or numerical simulations [8–10] to extract the critical behavior.

In this paper we will investigate the static and dynamic behavior of a nonequilibrium phase transition from a different perspective, using a kinetic description based on the master equation for the probability distribution. The steady-state solutions of the master equation describe the static behavior of the system, and by decomposing the master equation into various normal modes we can study the relaxation to the steady state. We argue below that the phase transition in a nonequilibrium system is caused, in the infinite-volume limit, by the appearance of new effective steady states derived from the slowly relaxing modes of the master equation. By studying the slow modes in a large system we can thus extract information about the nature of the static and dynamic behavior near the phase transition. This method, which bears a strong resemblance to the transfer-matrix technique used in equilibrium systems, is an additional approach to finding critical behavior that complements the Monte Carlo or series-expansion method. While the method of solving a master equation by a normal-mode decomposition is well known [11] in the chemical physics literature, the present work is an attempt to combine it with finite-size scaling to study the critical behavior of a nonequilibrium phase transition.

This paper is divided into four sections. In Sec. II we will discuss the mechanism for a nonequilibrium phase transition from the point of view of the master equation. In Sec. III we will present finite-size-scaling studies of the critical behavior of a model exhibiting a nonequilibrium phase transition, and in Sec. IV we will briefly summarize our approach.

II. CRITICAL BEHAVIOR FROM NORMAL MODES

Assuming that the underlying dynamical evolution is Markovian, a complete description of the behavior of a system with many degrees of freedom is given by the master equation

$$\frac{dP_i}{dt} = \sum_j (W_{ij}P_j - W_{ji}P_i), \quad (1)$$

where P_i is the probability for the system to be in a particular state i and W_{ji} is the transition probability from state i to state j. In any particular kinetic model the transition rates are determined by a set of rate parameters that play a role similar to that played by the chemical potential, temperature, and magnetic field in an equilibrium system. The above equation was first studied by Glauber [12] for systems in thermal equilibrium, where the transition rates obey detailed balance condition $W_{ij}/W_{ji} = P_i^{\rm eq}/P_j^{\rm eq}$ with $P^{\rm eq}$ denoting the equilibrium distribution. For nonequilibrium systems, detailed balance does not hold and the static behavior must be extracted from Eq. (1).

Writing a solution to Eq. (1) of the form $P_i = C_i^\lambda \exp(-\lambda t)$, we find that the coefficients C_i^λ are the right eigenvectors of the eigenvalue equation

$$\lambda C_i^\lambda = \sum_j (W_{ji}C_i^\lambda - W_{ij}C_j^\lambda). \quad (2)$$

The dynamical evolution of the system can therefore be viewed as the evolution of various normal modes of Eq. (2). The long-time and steady-state behavior is entirely controlled by those modes with a very small or zero relaxation rate. The behavior of the system in the vicinity of the phase transition is also controlled by these same modes. The phase transition must arise from large changes in the character of the steady-state solutions to Eq. (2). Level crossing, which in quantum mechanics leads to large changes in the character of the eigenvectors, cannot occur for the steady-state solutions because the eigenvalues must remain non-negative. Thus the form of the eigenvectors generally varies smoothly at the transition, and so the transition must be accompanied by the appearance of additional steady-state solutions for some range of values of the rate parameters. Since most kinetic models have a unique steady state in a finite-size system, these extra steady-state solutions will arise only in the infinite-volume limit where their relaxation rates will vanish. For a system of finite size, we thus expect to see a change in the size dependence of the relaxation rate of these slowly decaying modes at the transition. As in an equilibrium transition, we expect that the number of independent modes that become degenerate in the infinite volume limit is determined by the symmetries of the kinetic rules and the spatial dimensionality.

As an example we consider the class of nonequilibrium models with a single absorbing state, that is, a configuration that cannot be left once it is entered. This class includes the contact process [13], epidemic models [4], and models for catalytic reactions on surfaces [14,15]. The generic behavior of the phase transition in this class of models was elucidated first by Grassberger [16] and Janssen [17], who argued that the critical behavior should be the same as directed percolation (DP) [18,19] and Reggeon field theory (RFT) [20]. This behavior has been seen in a variety of models [7–10,21,22].

For system of finite size, the only steady state of this class of models is the adsorbing state. Since the mode corresponding to the steady state has unit probability of one configuration only, there is no statistical noise in this steady state. All other modes of the master equation have nonzero amplitudes for many different configurations, and thus statistical noise. As a function of some control parameter p, there may be a critical value $p = p_c$ where for $p < p_c$ the relaxation rate γ of one excited mode into the absorbing state remains nonzero for large systems and for $p \geq p_c$ the relaxation rate of this mode vanishes in the limit of infinite system size. This mode then becomes the effective steady state for the system and the system changes from a noiseless absorbing steady state to a noisy state. Because these systems have no symmetry other than translational invariance, we expect only one mode becomes critical at p_c. This is the scenario that occurs for models that show critical behavior characteristic of directed percolation [16,21].

Our approach bears a strong resemblance to the transfer-matrix technique used for equilibrium systems [23], where the partition function is found from the largest eigenvalue of the transfer matrix and the spatial correlation length is found from the ratio of the two largest eigenvalues [23]. This technique, which is extensively used for one-dimensional systems with short-range forces, forms the basis of Onsager's solution to the two-dimensional (2D) Ising model [24], which identifies the critical temperature with the point where the two largest eigenvalues of the transfer matrix become degenerate. Kac [25] has argued that the degeneracy of eigenvalues of the transfer matrix is the generic mathematical mechanism underlying second order phase transitions with divergent spatial correlations.

The transition probability matrix in Eq. (2) acts as the nonequilibrium analog of the transfer matrix, with the time direction in the nonequilibrium problem replacing the spatial direction chosen in the transfer matrix. This means that the static correlations in the nonequilibrium case are contained not in the eigenvalues but in the eigenvectors of the slow modes, like the transverse correlations in the transfer-matrix approach. The mechanism we present here can be viewed as the extension of Kac's idea [25] to the nonequilibrium case. For example, Henkel and Privman [26] have argued that asymptotic degeneracy of the two lowest eigenvalues of the master equation is a generic feature of the directed percolation transition.

III. FINITE-SIZE SCALING STUDY

In order to demonstrate the utility of this method, we have studied a kinetic model [27] that simulates a surface catalytic reaction $A + A \rightarrow A_2$ on a chain of sites, each site of which can be either empty or occupied by one atom. Atoms may adsorb on vacant sites if both neighboring sites are also empty. If one or both of the adjacent sites are already occupied, the atom will absorb with probability p. Otherwise, with probability $(1 - p)$, it will immediately react with a neighboring atom and both atoms will instantaneously desorb to leave two vacant sites. Atoms only react immediately after being adsorbed. As a function of the single rate parameter p, the

number of vacancies in the chain shows a second order transition at a critical value $p_c \approx 0.28$. For $p < p_c$ the vacancy fraction is nonzero and a reactive steady state appears with a constant production rate of A_2. For $p \geq p_c$ the steady state is an absorbing state where every site is occupied. Both the static [8,27] and dynamic [8] critical behavior of this model have been extensively studied via Monte Carlo simulation and the critical exponents appear to be in the DP/RFT universality class, in agreement with the discussion [16,17,21] above on transitions to an absorbing state.

We calculated the transition rates W_{ij} in Eq. (2) for system sizes from $L = 3$ to 19 with various values of p and then diagonalized the resulting nonsymmetric rate matrix numerically. We used the translational invariance of the model to reduce the large size of the matrix (2^L) by a factor of L by studying modes with a definite wave vector k, since modes with different values of k are uncoupled.

We argued above that for models of the present kind, the slowest mode determines the critical behavior of the system. In Fig. 1 we show the relaxation rates of the two slowest $k = 0$ modes, and it is obvious from this figure that only one mode controls the long-time behavior of the system. We have also examined the $k \neq 0$ modes, and while for large systems some of them lie below the upper mode shown in Fig. 1, they all relax more slowly than the lowest $k = 0$ mode. This behavior supports our contention that, in addition to the absorbing state, only one ($k = 0$) mode controls the critical behavior of the system while the rest of the modes act as spectators.

As an additional check, we computed directly a quantity studied in Monte Carlo simulations on this model [8], the mean lifetime for a vacancy

$$\tau_{\rm MC} = \frac{\sum_t t\, N(t)}{\sum_t N(t)} , \tag{3}$$

where $N(t)$ is the number of vacant sites at time t. From Eq. (1) the vacancy fraction $N(t)$ behaves as

$$N(t) = \sum_i a_i\, e^{-\lambda_i t} , \tag{4}$$

where the coefficients a_i are determined by the initial conditions and λ_i is the decay rate of the ith normal mode. Then from Eq. (3) we find

$$\tau_{\rm MC} = \frac{1}{\lambda_1} \frac{a_1 + \sum_{i>1} a_i \left(\frac{\lambda_1}{\lambda_i}\right)^2}{a_1 + \sum_{i>1} a_i \left(\frac{\lambda_1}{\lambda_i}\right)} . \tag{5}$$

Contributions to $\tau_{\rm MC}$ from modes other than the slowest mode are evidently included in the summation, but if we use an initially blank lattice as used in the simulations [8], a direct calculation shows the corrections a_i/a_1 are typically of the order of a few percent for variety of lattice sizes. Thus $\tau_{\rm MC}$ should and indeed does show the same critical behavior as $\tau = 1/\lambda_1$. This is further evidence that the critical behavior of the system is solely controlled by the lowest normal mode.

We calculated the relaxation time $\tau = 1/\lambda_1$ of the slowest mode for systems of size 3 to 19 for various values of p. The critical exponents z and ν can be found by assuming that τ obeys a finite-size scaling relation

$$\tau = L^z f_1 \left[(p - p_c) L^{1/\nu}\right] , \tag{6}$$

where $f_1(x) \propto x^{z\nu}$ for $x \to \infty$.

We determined the critical point p_c and exponent z from plots of $\ln(\tau)$ vs $\ln(L)$ plots for various p values, which are shown in Fig. 2. The value of p that produces a straight line in the plot indicates the critical point p_c. We find $p_c = 0.28 \pm 0.02$, in agreement with the value of

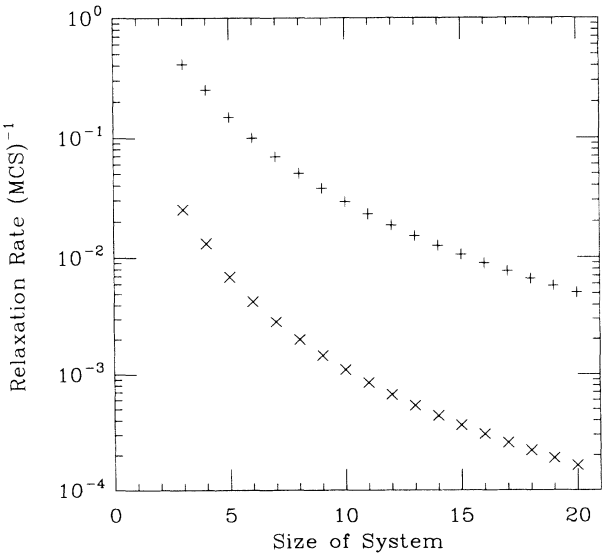

FIG. 1. The relaxation rates of the two slowest $k = 0$ modes in the vicinity of the critical point as a function of system size.

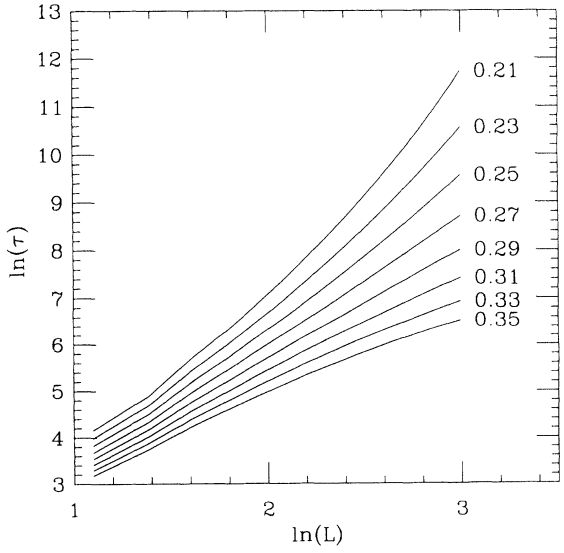

FIG. 2. Behavior of $\ln(\tau)$ vs $\ln(L)$ for various p values. The value of p which produces a straight line yields the critical point $p_c = 0.27 \pm 0.02$.

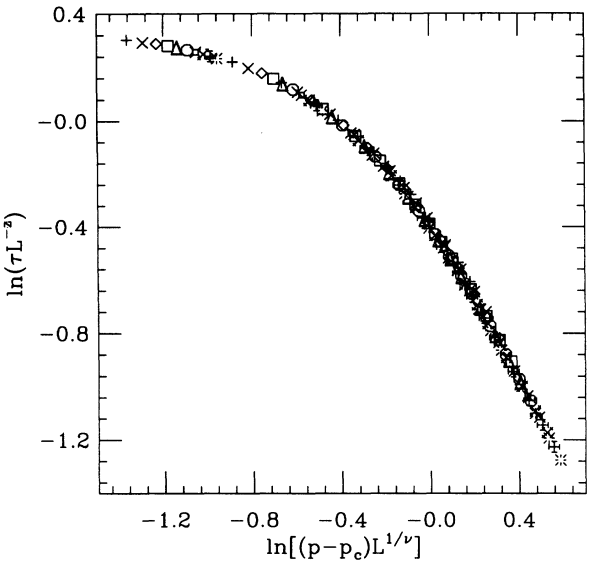

FIG. 3. A scaling plot of $\ln(\tau L^{-z})$ vs $\ln\left[(p-p_c)L^{1/\nu}\right]$ for various lattice sizes showing the collapse of the data on a single universal curve.

0.277 found by Monte Carlo simulation [8]. Furthermore we find that it is possible to scale all the data for τ for various L onto a single universal curve, as Eq. (6) would predict. In Fig. 3 we show the scaled data for $\ln(\tau L^{-z})$ vs $\ln\left[(p-p_c)L^{1/\nu}\right]$ for variety of system sizes. The scaling yields values of $z = 1.53 \pm 0.12$ and $\nu = 1.05 \pm 0.05$, which agree well with the 1+1D DP [18] values of 1.58 and 1.10, respectively. These results confirm earlier work [8] that the phase transition in this model belongs to the RFT/DP universality class.

IV. CONCLUSIONS

We have presented here a picture of the behavior of a nonequilibrium system using the modes of the master equation. Our method emphasizes the behavior of various normal modes as the system undergoes a phase transition and it gives a complementary way to study nonequilibrium transitions. The method and the underlying mechanism for the transition bear a strong resemblance to transfer-matrix techniques for equilibrium systems. Our approach also directly implements the role of symmetry in determining the critical behavior, so it should prove useful in looking for models which exhibit critical behavior different from directed percolation.

For the model studied here we have been able to isolate the relevant critical mode from the irrelevant modes and have used a finite-size-scaling approach to find the critical exponents. We have also found evidence of asymptotic degeneracy in the two largest eigenvalues, as expected for a directed percolation transition [25]. While the results we have obtained so far are not as accurate as those found by series expansion [7] and Monte Carlo [8,9,22] simulations, the results can be obtained with far less computational effort for a similar accuracy. Because we wanted to analyze the master equation exactly, the present results were limited by the size of the largest rate matrix we can diagonalize. It is possible to study larger systems by using variational techniques or by truncating the basis of states.

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