# Asymptotic forms and scaling properties of the relaxation time near threshold points in spinodal-type dynamical phase transitions

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We study critical properties of the relaxation time at a threshold point in switching processes between bistable states under change in external fields. In particular, we investigate the relaxation processes near the spinodal point of the infinitely long-range interaction model (the Husimi-Temperley model) by analyzing the scaling properties of the corresponding Fokker-Planck equation. We also confirm the obtained scaling relations by direct numerical solution of the original master equation, and by kinetic Monte Carlo simulation of the stochastic decay process. In particular, we study the asymptotic forms of the divergence of the relaxation time near the spinodal point and re-examine its scaling properties. We further extend the analysis to transient critical phenomena such as a threshold behavior with diverging switching time under a general external driving perturbation. This models photoexcitation processes in spin-crossover materials. In the ongoing development of nanosize fabrication, such size-dependence of switching processes should be an important issue, and the properties obtained here will be applicable to a wide range of physical processes.

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## I. INTRODUCTION

Relaxation phenomena in strongly interacting systems have been studied extensively, including various types of threshold phenomena. One of the most typical examples is the decomposition at the coercive field (the end of the hysteresis loop of a ferromagnetic system). This phenomenon appears in the field dependence of the order parameter in the ordered phase. In mean-field studies, this phenomenon is well described by the change in the free energy as a function of the magnetization (the order parameter of a ferromagnetic system). That is, in the ordered state, the free energy has two minima representing the symmetry-broken states. When we apply an external field, one of them is selected to be the equilibrium state. For a weak field, however, the other state remains as a metastable state. When the field becomes strong, the metastable state finally becomes unstable. This point is called the spinodal point. However, in models with short-range interactions, fluctuations cause the system to escape from the metastable state through nucleation phenomena, and thus the change in the relaxation time is smeared. Thus, although there are several studies on the divergence of the relaxation near the spinodal points [1], the spinodal phenomenon in short-range systems must be defined only as a crossover [2].

Recently, however, it has been pointed out that the mean-field universality class is realized in spin-crossover type systems [3]. In these materials, the volume of the unit cell changes, depending on the local twofold states [say, the high-spin (HS) and low-spin (LS) states]. The volume change causes a lattice distortion, and the elastic interactions among the local distortions cause an effective long-range interaction among the spin states. The critical properties of the spin-crossover system turned out to belong to the mean-field universality class. It was also found that the finite-size analysis for various quantities is very similar to the long-range, weakly interacting model (the Husimi-Temperley model).

In spin-crossover systems, one may expect that the dynamics also corresponds to that of the mean-field model. In particular, various threshold phenomena have been pointed out in the dynamics of spin-crossover type materials. For example, a spinodal phenomenon without nucleation type clustering was reported in numerical calculations [3]. The change is considered not to be a crossover process but a change with a true critical singularity that can be described by mean-field dynamics [4]. Moreover, threshold phenomena have been pointed out in the dependence of the photoexcitation process on the strength of the photoirradiation, which can be modeled as a kind of spinodal phenomenon [5]. The metastable state does not relax to the stable state in the mean-field approximation, which corresponds to the infinite system size in the Husimi-Temperley model.

In recent extensive studies on nanosize systems, finite-size effects turn out to play important roles. Therefore, it would be useful to study finite-size effects on the spinodal phenomenon as a critical dynamical process. The critical phenomena of the phase transition were studied in a dynamical mean-field model. The relaxation time diverges when the parameter approaches the threshold value. Binder studied the divergence of the relaxation time in spinodal phenomena in the mean-field model by a Monte Carlo method [1]. Although the metastable state does not relax to the stable state in the mean-field approximation, Paul *et al.* studied the

 

relaxation from the metastable side in finite-size systems and obtained a finite-size scaling form of the relaxation time [6]. The size dependence of the relaxation was also discussed recently [7].

In the present paper, we study the asymptotic behavior of the relaxation time of the infinitely long-range interaction model (the Husimi-Temperley model) near the spinodal point including the metastable region, the spinodal point, and the unstable region. We re-examine the scaling properties near the spinodal point, which have been proposed by Paul *et al.* [6].

As in the previous studies, we adopt the Glauber dynamics, and derive a master equation for the probability distribution of the total magnetization. We first derive a master equation as a function of the total magnetization, which is possible in the Husimi-Temperley model because of the long-range nature of the interactions. Then, we derive a Fokker-Planck equation by using an expansion in terms of the inverse system size, which is an example of the van Kampen Omega expansion [8,9]. We analyze the Fokker-Planck equation and derive a scaling relation and also asymptotic forms of the relaxation times. These properties are confirmed by direct numerical investigations of the original master equation, as well as corresponding Monte Carlo simulations of the stochastic decay process.

We also extend the analysis to general threshold phenomena, such as switching from LS to HS states in spin-crossover materials by photoirradiation. We obtain the effects of the photoirradiation on the master equation, and show that the critical properties of these processes are the same as those of the spinodal phenomena.

The rest of this paper is organized as follows. In Sec. II, we review the spinodal phenomena and define the master equation for the long-range model. Then, we study the asymptotic size dependence of the relaxation time near the spinodal point in Sec. III Numerical confirmation of the asymptotic forms is given in Sec. IV Next, we extend our study to threshold phenomena under external pumping in Sec. V Finally, a summary and discussion are given in Sec. VI.

## II. INFINITE LONG-RANGE MODEL AND THE SPINODAL POINT

We investigate the relaxation phenomena near the spinodal point in the infinitely long-range model. This is a spin model in which each spin interacts equally with every other spin. The Hamiltonian is

$$\mathcal{H} = -\frac{J}{2N}M^2 - HM, \quad M = \sum_{i=1}^{N} \sigma_i, \tag{1}$$

where $H$ is the magnetic field and $\sigma_i = \pm 1$. It is well-known that the mean-field theory is exact for this model in the limit of $N \to \infty$. This model shows a second-order phase transition at $T = T_C = J$, $H = 0$. Below the critical temperature, a metastable state exists for weak magnetic fields. When the magnetic field becomes strong, the metastable state becomes unstable at a certain point, known as the spinodal point. In order to determine the spinodal point, we consider the extended free energy, i.e., the free energy for fixed total magnetization. In the infinitely long-range model, this is given by

$$f(m) = -\frac{J}{2}m^2 - Hm - \frac{1}{\beta N}\ln_N C_{(N+M)/2}$$
$$\sim -\frac{J}{2}m^2 - Hm + \frac{1}{\beta}\left(\frac{1+m}{2}\ln\frac{1+m}{2} + \frac{1-m}{2}\ln\frac{1-m}{2}\right), \tag{2}$$

in the limit $N \to \infty$ where we use Stirling's formula and $m$ is the magnetization per spin ($m = M/N$). The spinodal point is given by the following conditions:

$$\frac{\partial f}{\partial m} = 0 \quad \text{and} \quad \frac{\partial^2 f}{\partial m^2} = 0,$$

which give

$$H_{SP} = \mp J\sqrt{1 - \frac{1}{\beta J}} \pm \frac{1}{2\beta}\ln\frac{1 + \sqrt{1 - \frac{1}{\beta J}}}{1 - \sqrt{1 - \frac{1}{\beta J}}}, \tag{3}$$

at which the magnetization is given by

$$m_{SP} = \pm\sqrt{1 - \frac{1}{\beta J}}. \tag{4}$$

In this paper, we consider the case where we increase the field from a negative value. Therefore, we consider the behavior of a locally stable state at negative magnetization around $m_{SP} < 0$.

We study the dynamics via the standard master equation

$$\frac{\partial P(S,t)}{\partial t} = -\sum_{S'} W_{S \to S'}P(S) + \sum_{S'} W_{S' \to S}P(S'), \tag{5}$$

where $S$ and $S'$ denote states of the system and $W_{S \to S'}$ is a transition rate from $S$ to $S'$. The probability of the state $S$ at time $t$ is denoted by $P(S,t)$.

Among the many possible dynamical models (choices of the transition rate), we adopt the Glauber dynamics in this work. In the Glauber dynamics, the transition takes place as a flip of a local spin, and the transition rate $w_{ij}$ from a local spin state $i$ to a local spin state $j$ is given by

$$w_{ij} = \frac{1}{\tau_0}\frac{e^{-\beta E_j}}{e^{-\beta E_i} + e^{-\beta E_j}}, \tag{6}$$

where $E_i$ denotes the energy of the system in spin state $i$, and $\tau_0$ is some characteristic time scale. In this paper, we scale the time by $\tau_0$ and set $\tau_0 = 1$ for simplicity.

With this transition rate, we construct a master equation for the mean-field model. As the Hamiltonian depends only on the magnetization $M$, the master equation is written in closed form for $M$. Thus, the master Eq. (5) can be expressed as a function of $M$. Let $P(M)$ be the probability that the system has the total magnetization $M$. The master equation for $P(M)$ is given by

$$\frac{\partial P(M,t)}{\partial t} = \frac{1}{\tau_0}\left\{ -\frac{N+M}{2}\frac{\exp\{-\beta[J(M-1)/N+H]\}}{2\cosh\{\beta[J(M-1)/N+H]\}}P(M) - \frac{N-M}{2}\frac{\exp\{\beta[J(M+1)/N+H]\}}{2\cosh\{\beta[J(M+1)/N+H]\}}P(M) \right.$$
$$\left. + \frac{N-M+2}{2}\frac{\exp\{\beta[J(M-1)/N+H]\}}{2\cosh\{\beta[J(M-1)/N+H]\}}P(M-2) + \frac{N+M+2}{2}\frac{\exp\{-\beta[J(M+1)/N+H]\}}{2\cosh\{\beta[J(M+1)/N+H]\}}P(M+2)\right\}, \quad (7)$$

where $N$ is the number of spins and $M$ takes discrete values $-N,-N+2,\ldots,N$ (see Appendix A). For finite $N$, we can solve the simultaneous equations for $P(-N,t),P(-N+2,t),\ldots,P(N,t)$, as well as perform a Monte Carlo simulation of the model.

## III. ASYMPTOTIC SIZE-DEPENDENCE OF THE RELAXATION TIME NEAR THE SPINODAL POINT

Next, we study the scaling properties of the relaxation time near the spinodal point. For large $N$, we set $m=M/N$ and regard $m$ as a continuous variable. Expanding the RHS of Eq. (7) in a series in $\varepsilon=1/N$, we obtain the following Fokker-Planck equation:

$$\frac{\partial P(m)}{\partial t} = \frac{\partial}{\partial m}g_1(m)P(m) + \varepsilon\frac{\partial^2}{\partial m^2}g_2(m)P(m) + O(\varepsilon^2), \quad (8)$$

where

$$g_1(m) = m - \tanh[\beta(Jm+H)] + \varepsilon\frac{\beta Jm}{\cosh^2[\beta(Jm+H)]},$$

$$g_2(m) = 1 - m\tanh[\beta(Jm+H)]. \quad (9)$$

The last term of $g_1(m)$ gives the correction to the spinodal point due to the finite-size effect. Hereafter, we ignore this term as it is very small. Near the spinodal point, we expand $g_1(m)$ and $g_2(m)$ around the spinodal point. We set $x=m-m_{\mathrm{SP}}$, and $y=\beta(H-H_{\mathrm{SP}})\equiv h-h_{\mathrm{SP}}$, and then we have

$$g_1(m) \simeq -\frac{y}{\beta J} - \eta(\beta J x^2 + 2xy),$$

$$g_2(m) \simeq \frac{1}{\beta J}, \quad (10)$$

where $\eta=|m_{\mathrm{SP}}|=\sqrt{1-1/\beta J}$.

Let us consider the time evolution of $x$, starting from $x=0$. The distribution of $x$ evolves according to Eq. (8) with $g_1\sim -y/\beta J$ and $g_2\sim 1/\beta J$. When $x$ approaches $x\sim y^{1/2}$, the correction term $-\eta\beta J x^2$ in $g_1$ becomes relevant, but other correction terms in $g_1$, such as $-2\eta xy$, are still irrelevant. Therefore, in order to determine the relaxation time, we can use the approximation that $g_1\simeq -y/\beta J-\eta\beta J x^2$ in the early stage of the phase change. Then, the Fokker-Planck equation takes the form

$$\frac{\partial}{\partial t}P(x,t) = \left[-\frac{\partial}{\partial x}\left(\frac{y}{\beta J}+\eta\beta J x^2\right) + \frac{\varepsilon}{\beta J}\frac{\partial^2}{\partial x^2}\right]P(x,t). \quad (11)$$

Now, we introduce the scaled parameters,

$$\xi = x|y|^{-1/2}, \quad (12)$$

and

$$\Lambda = N^{2/3}y. \quad (13)$$

Then, for nonzero $y$, the Fokker-Planck equation is given by

$$\frac{\partial}{\partial t}P(\xi,t) = \varepsilon^{1/3}\left\{-|\Lambda|^{1/2}\frac{\partial}{\partial\xi}\left(\pm\frac{1}{\beta J}+\eta\beta J\xi^2\right) + \frac{1}{\beta J|\Lambda|}\frac{\partial^2}{\partial\xi^2}\right\}P(\xi,t), \quad (14)$$

where $\Lambda>0$ for the upper sign, and $\Lambda<0$ for the lower sign. Because Eq. (14) depends only on $\Lambda$ except for the factor $\varepsilon^{1/3}$, the relaxation time is expected to be given in the form

$$\tau \sim N^{1/3}f(\Lambda) = N^{1/3}f(N^{2/3}(h-h_{\mathrm{SP}})). \quad (15)$$

This is the finite-size scaling for the relaxation time near the spinodal point. This form has been pointed out by Paul *et al.* [6]. Here, we re-examine the form of the scaling function by studying the asymptotic forms of the relaxation time.

In the above argument leading to the scaling relation, it is necessary to pay attention to the range of the parameters, in which the application of the above estimate can be verified. We may regard the relaxation time as a time when the magnetization $x$ becomes $O(1)$. Then, $\xi$ becomes $O(|y|^{-1/2})$. This implies that we may need to include an additional $y$ dependence in the relaxation time. Thus, we cannot immediately conclude that the finite-size scaling has the simple form of Eq. (15). Indeed, such consideration is essential for the relaxation after rapid quenching at zero field and has been studied as a scaling theory of the relaxation at unstable point [10]. In fact, the relaxation time is proportional to $\ln N$ in the relaxation at the unstable point, which is unexpected from the form of the Fokker-Planck equation. This problem is investigated in Appendix B, where we show that the additional contribution does *not* need to be taken into account in the relaxation at the spinodal point, and the finite-size scaling is correctly given by Eq. (15). In the following, we investigate asymptotic forms of the relaxation time for the cases $y<0$, $y=0$, and $y>0$.

In order to consider more general cases later (Sec. V), we rewrite the Fokker-Planck equation with general coefficients $\alpha$, $\gamma$, and $\delta$,

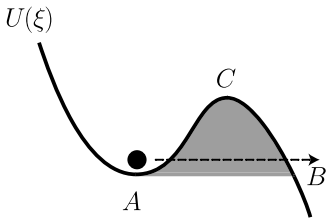


FIG. 1. The scaled potential $U(\xi)$ in Eq. (19). A: the metastable state. B: a point in the basin of attraction of the stable state. C: the unstable state.

$$\frac{\partial}{\partial t}P(\xi,t) = \varepsilon^{1/3}\left\{-|\Lambda|^{1/2}\frac{\partial}{\partial \xi}(\pm\alpha+\gamma\xi^2)+\frac{\delta}{|\Lambda|}\frac{\partial^2}{\partial \xi^2}\right\}P(\xi,t). \tag{16}$$

The coefficients $\alpha$, $\gamma$, and $\delta$ are defined as follows:

$$g_1 \simeq -\alpha y - \gamma x^2,$$

$$g_2 \simeq \delta. \tag{17}$$

In the case of the relaxation from the mean-field spinodal point, $\alpha=\delta=1/(\beta J)$, $\gamma=\eta\beta J$.

### A. Metastable region: The $y<0$ case

This case corresponds to the relaxation from the metastable state to the equilibrium state. We can estimate the relaxation time by Kramers' method of escape over a potential barrier [11]. We rewrite Eq. (16) in the following form ($\Lambda<0$):

$$\frac{\partial P}{\partial t} = \varepsilon^{1/3}\frac{\partial}{\partial \xi}\left(\frac{\delta}{|\Lambda|}e^{-|\Lambda|^{3/2}U(\xi)/\delta}\frac{\partial}{\partial \xi}e^{|\Lambda|^{3/2}U(\xi)/\delta}P\right), \tag{18}$$

where

$$U(\xi) = \alpha\xi - \gamma\xi^3/3 \tag{19}$$

is the scaled potential. We consider the escape of probability from the valley to the outside ($A\rightarrow B$) as depicted in Fig. 1 [11]. There, the probability current $\sigma$ is given by

$$\sigma = -\varepsilon^{1/3}\frac{\delta}{|\Lambda|}e^{-|\Lambda|^{3/2}U(\xi)/\delta}\frac{\partial}{\partial \xi}e^{|\Lambda|^{3/2}U(\xi)/\delta}P. \tag{20}$$

We consider the stationary current ($\sigma$=const.) and integrate it between the two points $A$ and $B$.

$$\sigma = -\varepsilon^{1/3}\frac{\delta}{|\Lambda|}\frac{[e^{|\Lambda|^{3/2}U(\xi)/\delta}P]_A^B}{\int_A^B e^{\beta J|\Lambda|^{3/2}U(\xi)/\delta}d\xi}. \tag{21}$$

If the number of spins $N$ is very large, we can use the steepest-descent method, and we have

$$\int_A^B e^{|\Lambda|^{3/2}U(\xi)/\delta}d\xi \approx \int_{-\infty}^{\infty} e^{(1/\delta)|\Lambda|^{3/2}U(C)+(1/2\delta)\beta J|\Lambda|^{3/2}U''(C)(\xi-C)^2}d\xi$$

$$= \sqrt{\frac{-2\pi\delta}{|\Lambda|^{3/2}U''(C)}}e^{|\Lambda|^{3/2}U(C)/\delta}. \tag{22}$$

This approximation is valid for a sufficiently large $|\Lambda|$. We consider the early stage of the relaxation and assume the relaxation has not occurred yet, namely, $P(B)\approx 0$. Then, we obtain the estimate

$$\sigma = P(A)\varepsilon^{1/3}\sqrt{\frac{-\delta U''(C)}{2\pi|\Lambda|^{1/2}}}e^{-|\Lambda|^{3/2}[U(C)-U(A)]/\delta}. \tag{23}$$

The probability distribution near the point $A$ is approximately given by

$$P(\xi) \simeq n_A\frac{\exp\left[-\frac{|\Lambda|^{3/2}}{\delta}\left\{U(A)+\frac{1}{2}U''(A)(\xi-A)^2\right\}\right]}{Z}, \tag{24}$$

where $Z$ is a partition function,

$$Z = \int_{-\infty}^{\infty}d\xi\exp\left[-\frac{|\Lambda|^{3/2}}{\delta}\left\{U(A)+\frac{1}{2}U''(A)(\xi-A)^2\right\}\right]$$

$$= e^{-|\Lambda|^{3/2}U(A)/\delta}\sqrt{\frac{2\pi\delta}{|\Lambda|^{3/2}U''(A)}}. \tag{25}$$

Namely, the probability distribution near the point $A$ is given by the equilibrium distribution of the approximate harmonic potential. The variable $n_A$ represents the total probability near the point $A$. This quantity evolves as

$$\frac{d}{dt}n_A = -\sigma = -\frac{1}{\tau}n_A, \tag{26}$$

where $\tau$ is the relaxation time that we want to know. As the probability at the point $A$ is given by

$$P(A) \simeq n_A\frac{\exp\left[-\frac{|\Lambda|^{3/2}}{\delta}U(A)\right]}{Z} = n_A\sqrt{\frac{|\Lambda|^{3/2}U''(A)}{2\pi\delta}}, \tag{27}$$

[see Eq. (24)], combining Eqs. (23), (26), and (27), we obtain

$$\tau \sim N^{1/3}2\pi|U''(A)U''(C)|^{-1/2}|\Lambda|^{-1/2}$$

$$\times\exp\left\{\frac{|\Lambda|^{3/2}}{\delta}[U(C)-U(A)]\right\}. \tag{28}$$

This is the result of the well-known Kramers' formula for the escape rate, and it agrees with the finite-size scaling Eq. (15).

The potential $U(\xi)$ is $U(\xi)=\alpha\xi-\gamma\xi^3/3$, and the two points $A$ and $C$ are given by the condition $dU/d\xi=0$. Therefore,

$$A = -C = -\frac{\alpha}{\gamma},$$

$$U(C) - U(A) = \frac{4\alpha}{3}\sqrt{\frac{\alpha}{\gamma}},$$

$$U''(C) = -2\sqrt{\alpha\gamma}. \tag{29}$$

Hence, the relaxation time for sufficiently large $|\Lambda|$ is

$$\tau \sim N^{1/3}\frac{\pi}{2\sqrt{\alpha\gamma|\Lambda|}}\exp\left\{\frac{4\alpha}{3\delta}\sqrt{\frac{\alpha}{\gamma}}|\Lambda|^{3/2}\right\}. \tag{30}$$

In the case of Eq. (10), $\alpha=\delta=1/(\beta J)$, $\gamma=\eta\beta J$, and we have

$$\tau \sim N^{1/3}\frac{\pi}{2\sqrt{\eta|\Lambda|}}\exp\left\{\frac{4}{3\beta J\eta^{1/2}}|\Lambda|^{3/2}\right\}. \tag{31}$$

Here, it should be noted that from Eq. (2) the following relation holds:

$$\frac{4}{3\beta J\eta^{1/2}}\Lambda^{3/2} = \beta\Delta F \equiv \beta[F(C) - F(A)].$$

Therefore, the relaxation time obtained above is roughly $\tau \sim e^{\beta\Delta F}$, which is the well-known Arrhenius formula. Another derivation of Eq. (30) uses the WKB approximation as discussed by Tomita *et al.* [12]. We can derive the same result (see Appendix C).

In the infinite long-range model, microscopic fluctuations do not grow to become macroscopic because the long-range interaction suppresses clustering and nucleation. It should be noted that, in the limit of $N\to\infty$, the system remains at the metastable or marginally stable point, and the relaxation time from that point becomes infinite. In contrast, in systems with short-range interactions, the nucleation process causes the system to relax to the equilibrium state in a finite relaxation time. Thus, the divergence of the relaxation time does not take place, and so far the divergence of the relaxation time has not been considered seriously. However, as has been pointed out [3], effective long-range interactions appear in systems in which elastic deformation mediates interactions among the spins. In such systems, the long-range interaction model is effectively realized, and we expect that the finite-size scaling discussed here would be relevant.

### B. At the spinodal point: The $y=0$ case

Next, we consider the relaxation just at the spinodal point, $y=0$. Substituting $y=0$ in the Fokker-Planck Eq. (11), we obtain

$$\frac{\partial}{\partial t}P(x,t) = \gamma\frac{\partial}{\partial x}[x^2P(x,t)] + \varepsilon\delta\frac{\partial^2}{\partial x^2}P(x,t). \tag{32}$$

Putting $x=\varepsilon^{1/3}z$,

$$\frac{\partial}{\partial t}P(z,t) = \varepsilon^{1/3}\left\{-\gamma\frac{\partial}{\partial z}z^2 + \delta\frac{\partial^2}{\partial z^2}\right\}P(z,t). \tag{33}$$

By using the scaled variable $s=t\varepsilon^{1/3}$, we can eliminate the $\varepsilon$-dependence. Thus, as pointed out by Kubo *et al.* [9], the relaxation time behaves as

$$\tau \propto \varepsilon^{-1/3} = N^{1/3}. \tag{34}$$

The relaxation time diverges in the limit of $N\to\infty$ just at the spinodal point. In the limit of $N\to\infty$, the system remains at the unstable point, and the relaxation time becomes infinite. This divergence is again due to the long-range interaction.

### C. Unstable region: The $y>0$ case

Finally, we consider the case $y>0$. In this case, even if $N=\infty$ ($\varepsilon=0$), the relaxation takes place. Namely, the relaxation time saturates at a finite value at large $N$. Therefore, we consider only the limit $N\to\infty$. The Fokker-Planck Eq. (14) then becomes

$$\frac{\partial P}{\partial t} = -y^{1/2}\frac{\partial}{\partial\xi}(\alpha + \gamma\xi^2)P. \tag{35}$$

Therefore, the relaxation time is expected to scale as

$$\tau \sim y^{-1/2} \sim (h - h_{\mathrm{SP}})^{-1/2}. \tag{36}$$

In the limit of $N\to\infty$, there is no diffusion term, so we can derive the time evolution of the scaled magnetization $\xi(t)$ directly. Namely, putting $P(\xi,t)=\delta[\xi-\xi(t)]$, we obtain

$$\dot{\xi}(t) = y^{1/2}(\alpha + \gamma\xi(t)^2). \tag{37}$$

The solution of Eq. (37) is given by

$$\xi(t) = \sqrt{\frac{\alpha}{\gamma}}\tan(\sqrt{\alpha\gamma y}t) \tag{38}$$

for $\sqrt{\alpha\gamma y}t < \pi/2$. At a time $\sqrt{\alpha\gamma y}t=\pi/2$, the above expression indicates that $\xi(t)$ would diverge. However, the higher order terms in the original Fokker-Planck equation (8) prevent this divergence of the magnetization. Thus, the relaxation time is estimated as

$$\tau \sim \frac{\pi}{2}(\alpha\gamma y)^{-1/2}. \tag{39}$$

In the scaling form,

$$\tau \sim N^{1/3}\frac{\pi}{2}(\alpha\gamma\Lambda)^{-1/2}. \tag{40}$$

This result is consistent with that obtained by Binder [1], who showed that for $h>h_{\mathrm{SP}}$, the relaxation time behaves as $\tau\sim(h-h_{\mathrm{SP}})^{-1/2}$.

## IV. NUMERICAL RESULTS

We have seen that the relaxation time $\tau$ obeys the finite-size scaling form (15)

$$\tau(y,N) = N^{1/3}f(N^{2/3}y),$$

and we have derived the asymptotic forms of the relaxation time, i.e., in the case of Eq. (10),

$$f(\Lambda) \sim \begin{cases} \pi(\eta|\Lambda|)^{-1/2}\exp\left\{\dfrac{4}{3\beta J\eta^{1/2}}|\Lambda|^{3/2}\right\} & \text{for} \quad -\Lambda \gg 1 \\ \dfrac{\pi}{2}(\eta\Lambda)^{-1/2} & \text{for} \quad \Lambda \gg 1. \end{cases} \tag{41}$$

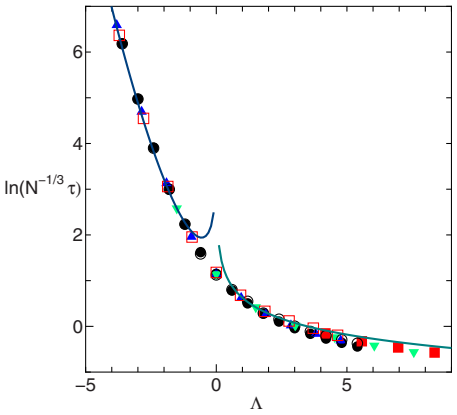


FIG. 2. (Color online) Comparison of the asymptotic form of the relaxation time with numerical results. Data are plotted in the coordinates $[\Lambda, \ln(N^{-1/3}\tau)]$. The solid lines denote the asymptotic form (41). The symbols are interpreted as follows: closed circles, upward triangles, downward triangles, and closed squares denote the data obtained by the master equation for $N=1000$, 2000, 4000, and 10 000, respectively. Open circles and squares denote the data obtained by the Monte Carlo method for $N=1000$ and 10 000, respectively.

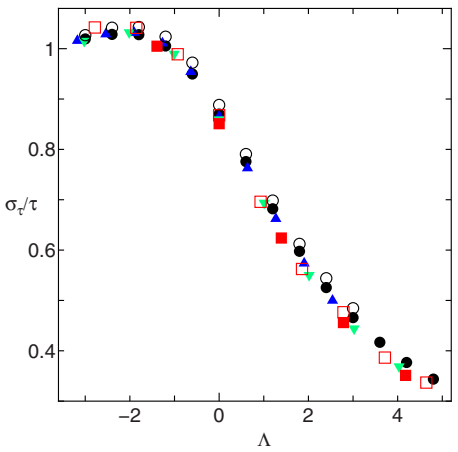


FIG. 3. (Color online) Ratio of the standard deviation of the lifetime, $\sigma_\tau$, to the lifetime, $\tau$, as obtained from the master equation and MC simulations. The data are shown as a scaling plot vs the scaling variable $\Lambda$. The symbols have the same interpretations as in Fig. 2.

In this section, we check these results by numerical studies. We solved the original master Eq. (7) numerically, and also performed kinetic MC simulations (see Appendix D). The parameters are set as $\beta=1$ and $J=2$. The relaxation time is defined as the time at which the magnetization of a sample reaches a certain value $m_0$. Here, we adopt $m_0=0$. In the Monte Carlo simulations, the relaxation time is measured directly in each simulation. On the other hand, in the master equation we have to define it from the change in the probability distribution $P(M,t)$. Namely, we obtain the average of the relaxation time with the formula,

$$\tau = -\int_0^\infty dt \sum_{M<0} \dot{P}(M,t)t, \tag{42}$$

and its standard deviation as

$$\sigma_\tau = -\int_0^\infty dt \sum_{M<0} \dot{P}(M,t)t^2 - \tau^2. \tag{43}$$

We plot data for various parameters in a scaling plot in Fig. 2, namely, in the coordinates $[\Lambda = N^{2/3}y, \ln(N^{-1/3}\tau)]$. We confirmed that both methods give the same results, as they should. In the Monte Carlo simulations, each data point is an average over 1000 samples, and the error bars are smaller than the symbol size in the figure. All the data collapse well onto a scaling function, which indicates that the finite-size scaling works well. For large $\Lambda$, data points for different $N$ deviate slightly from the scaling function. This fact is explained as follows: the condition for the finite-size scaling to hold is that the system size $N$ is sufficiently large and the magnetic field is sufficiently close to the spinodal point. This implies $N \gg 1$ and $|y| \ll 1$. However, an even stronger condition is required for the finite-size scaling. As we assumed $x = m - m_{\rm SP} \ll 1$ to derive the Fokker Planck equation and $x$ was rescaled as $x = \xi|y|^{1/2}$, not only $|y| \ll 1$ but also $|y|^{1/2} \ll 1$ was necessary. Therefore,

$$|\Lambda|^{1/2} \ll N^{1/3}$$

is required for the finite-size scaling.

Now, we compare the numerical results of the master equation with the asymptotic form for the relaxation time $\tau$, Eq. (41). In Fig. 2, we compare numerical results and the asymptotic forms, Eq. (41). Here, we find that the asymptotic formulas describe the scaling form well in the large-$\Lambda$ region, where the data points approach the asymptotic forms when the size increases. Here, we find that the scaling property (15) holds, and the asymptotic forms also hold asymptotically.

As the scaling variable $\Lambda$ goes from large positive to large negative values, the lifetime distribution goes from a narrow peak in the unstable region to an exponential distribution deep in the metastable region. This is illustrated in Fig. 3 by the ratio of the standard deviation of the lifetime, $\sigma_\tau$, to the lifetime, $\tau$, as obtained from the Master equation and also by MC simulations. These results also show good scaling collapse, although the convergence to the asymptotic limit is somewhat slower for $\sigma_\tau$ than for $\tau$, as one would expect for a quantity involving a second moment of the lifetime distribution.

## V. THRESHOLD PHENOMENA UNDER EXTERNAL PUMPING

In this section we study switching in spin-crossover (SC) materials under photoirradiation, which is representative of a wide range of switching processes under external driving forces. The pumping effect is described by the following process in the master equation. The pumping operation causes a down spin (LS) to flip to the up state (HS) with a transition rate $a$ per unit time. Thus, the total transition rate (6) from down to up is now given by

$$w(- \rightarrow +) = \frac{1}{\tau_0} \frac{e^{-\beta E_+}}{e^{-\beta E_+} + e^{-\beta E_-}} + a, \tag{44}$$

where $E_\pm$ denote the energies with the spin + and −, respectively. On the other hand, the transition rate from up to down remains unchanged and is given by

$$w(+ \rightarrow -) = \frac{1}{\tau_0} \frac{e^{-\beta E_-}}{e^{-\beta E_+} + e^{-\beta E_-}}. \tag{45}$$

These transition rates do not satisfy detailed balance, but they produce a stationary state. Thus, as far as we consider the system at a given temperature, we have a well-defined relaxation process to the stationary state. Here, we study the parameter dependence of the relaxation time. To express the pumping, we add the term

$$-a\frac{N-M}{2}P(M) + a\frac{N-(M-2)}{2}P(M-2) \tag{46}$$

in the master equation. In the large $N$ limit, this modification gives the additional terms in the Fokker-Planck equation

$$-a\frac{\partial}{\partial m}\{(1-m)P(m,t)\} + a\varepsilon\frac{\partial^2}{\partial m^2}\{(1-m)P(m,t)\}. \tag{47}$$

Thus, $g_1(m)$ and $g_2(m)$ in Eq. (9) are now replaced by

$$g_1(m) = m - \tanh[\beta(Jm+H)] - a(1-m),$$

$$g_2(m) = 1 - m\tanh[\beta(Jm+H)] + a(1-m). \tag{48}$$

The point corresponding to the spinodal point is given by

$$g_1(m) = 0 \quad \text{and} \quad \frac{\partial}{\partial m} g_1(m) = 0, \tag{49}$$

which are given by

$$\tanh z = \frac{1+a_c}{\beta J} z - \frac{H}{J}(1+a_c) - a_c, \tag{50}$$

and

$$\cosh^2 z = \frac{\beta J}{1+a_c}, \tag{51}$$

where $z=\beta(Jm_{\mathrm{SP}}+H)$. For the SC material, $H$ consists of the following two ingredients: the crystal field which gives the energy difference between HS and LS states, and a time-dependent field which represents the different degeneracies of the HS and LS states [13].

Expanding the variable near the spinodal point: $x=m-m_{\mathrm{SP}}$ and $y=a-a_c$, we have

$$g_1 \simeq -\alpha y - \gamma x^2, \tag{52}$$

where

$$\alpha = \frac{1}{1+a_c}\left(1 - \sqrt{1 - \frac{1+a_c}{\beta J}}\right), \tag{53}$$

and

$$\gamma = \beta J(1+a_c)\sqrt{1 - \frac{1+a_c}{\beta J}}, \tag{54}$$

and

$$g_2 \simeq \frac{1}{\beta J} + \frac{2a_c}{1+a_c}\left(1 + \sqrt{1 - \frac{1+a_c}{\beta J}}\right) \equiv \delta. \tag{55}$$

As $g_1$ and $g_2$ have the same forms as Eq. (17), the same analysis as in the previous section remains valid, and the finite-size scaling is again given in the form

$$\tau = N^{1/3} f(N^{2/3}(a-a_c)). \tag{56}$$

The asymptotic form is also given by Eqs. (30) and (40).

## VI. SUMMARY AND DISCUSSION

Mean-field type critical behavior takes place in systems with effective long-range interactions, as has been pointed out for spin-crossover type materials [3]. We expect that the dynamical critical properties such as the spinodal phenomena are realized in those systems. In models with short-range interactions, there exists a mode of relaxation from the metastable state through nucleation of localized clusters. Thus, the relaxation time around the spinodal point changes smoothly, and the critical properties at the spinodal point in the mean-field theory are smeared out. However, in long-range interaction models, the relaxation time diverges as described by the mean-field theory. It is, therefore, necessary to study the finite-size scaling properties of the critical behavior. Thus, we here studied the size-dependence of the relaxation time near the spinodal point in the Husimi-Temperley model. Starting from the master equation for the probability density of the total magnetization under the Glauber dynamics, the Fokker-Planck equation for large $N$ was obtained as in previous work [6]. Using this Fokker-Planck equation, we investigated the relaxation processes near the spinodal point. As a result, we obtained asymptotic forms and a finite-size scaling function for the relaxation time, which cover both sides of the spinodal point, i.e., the metastable side and the unstable side.

We further extended the analysis to systems that are pumped by an external field. The critical properties obtained in the present work should apply widely to threshold phenomena in long-range interacting models, such as the threshold phenomena found in the excitation process by photoirradiation from the low-temperature phase to a photoexcited high-temperature phase in spin-crossover materials [5].

In the ongoing development of nanosize fabrication, switching processes in nanosize systems are an important issue, and the size-dependence of relaxation times of the switching processes must be precisely understood. We hope the scaling properties presented here will help to analyze such processes in experimental systems.

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## APPENDIX A: THE DERIVATION OF THE MASTER EQUATION

In this appendix, we derive the master Eq. (7) for Hamiltonian (1) and the transition rate (6). The probability of the state $\{\sigma_1,\sigma_2,\ldots,\sigma_N\}$ at a time $t$, which is denoted by $P(\sigma_1,\ldots,\sigma_N;t)$, evolves according to

$$\frac{\partial}{\partial t}P(\sigma_1,\ldots,\sigma_N;t) = -\sum_{i=1}^{N}\omega_M(\sigma_i\to-\sigma_i)P(\sigma_1,\ldots,\sigma_N;t) + \sum_{i=1}^{N}\omega_{M-2\sigma_i}(-\sigma_i\to\sigma_i)P(\ldots,-\sigma_i,\ldots;t), \tag{A1}$$

where

$$\omega_M(\sigma_i\to-\sigma_i) = \frac{1}{\tau_0}\frac{\exp\{-\beta\sigma_i[J(M-\sigma_i)/N+H]\}}{2\cosh\{\beta[J(M-\sigma_i)/N+H]\}}, \tag{A2}$$

and $M$ is the total magnetization, i.e., $M=\Sigma_i\sigma_i$. We consider the time evolution of the probability of $M$

$$P(M,t) \equiv \sum_{\sigma_1,\sigma_2,\ldots,\sigma_N=\pm1}\delta\left(\sum_{i=1}^{N}\sigma_i,M\right)P(\sigma_1,\ldots,\sigma_N;t),$$

where $\delta(a,b)$ denotes the Kronecker delta. After some calculation from Eq. (A1), the equation of motion for $P(M,t)$ is obtained in the form

$$\begin{aligned}\frac{\partial}{\partial t}P(M,t) = &-\frac{N+M}{2}\omega_M(+1\to-1)P(M,t)\\ &-\frac{N-M}{2}\omega_M(-1\to+1)P(M,t)\\ &+\frac{N-(M-2)}{2}\omega_{M-2}(-1\to+1)P(M-2,t)\\ &+\frac{N+(M+2)}{2}\omega_{M+2}(+1\to-1)P(M+2,t).\end{aligned} \tag{A3}$$

The meaning of this equation is clear. The first and second terms correspond to the transition from the magnetization $M$ to $M-2$ and $M+2$, respectively. The third and fourth terms represent the transitions from $M-2$ to $M$ and from $M+2$ to $M$, respectively. This equation gives Eq. (7).

## APPENDIX B: CUTOFF INDEPENDENCE OF THE FOKKER-PLANCK EQUATION (16)

In Sec. II, we remarked on the possibility of an additional $y$-dependence in the relaxation time. Namely, if we regard the relaxation time as the time when the magnetization $x$ becomes $O(1)$, this corresponds to the time when $\xi$ becomes $O(|y|^{-1/2})$, and this implies that we cannot conclude the finite-size scaling of the relaxation time, Eq. (15), from the form of the Fokker-Planck equation (16). In other words, although the Fokker-Planck equation (16) seems to depend only on $\Lambda$, we must restrict the range of the variable $\xi < O(y^{-1/2})$ and this *cutoff* of $\xi$ can induce an additional $y$-dependence in the relaxation time. We note that the relaxation time indeed depends on the cutoff in other situations. One example is the relaxation from the mean-field unstable fixed point. In this case, the Fokker-Planck equation is given by

$$\frac{\partial}{\partial t}P(x,t) = -\frac{\partial}{\partial x}xP(x,t) + \varepsilon\frac{\partial^2}{\partial x^2}P(x,t), \tag{B1}$$

(we set some coefficients equal to unity). We can transform this equation to the scaling form similarly. If we set $x=\varepsilon^{1/2}\xi$,

$$\frac{\partial}{\partial t}P(\xi,t) = -\frac{\partial}{\partial \xi}\xi P(\xi,t) + \frac{\partial^2}{\partial \xi^2}P(\xi,t). \tag{B2}$$

This equation is apparently independent of $\varepsilon$. Is the relaxation time independent of the system size $N=1/\varepsilon$? The answer is no. It is known that the relaxation time in this case is $\tau\sim\ln N$ [10]. We show that this $N$-dependence stems from the finite cutoff. We can solve Eq. (B2) for the initial condition $P(\xi,0)=\delta(\xi)$,

$$P(\xi,t) = \frac{1}{\sqrt{2\pi}\sigma(t)}\exp\left[-\frac{\xi^2}{2\sigma(t)^2}\right] \tag{B3}$$

where $\sigma(t)$ is given by

$$\sigma(t) = \sqrt{e^{2t}-1} \sim e^t \tag{B4}$$

It takes infinite time for $\xi$ to reach infinity. As $x=\varepsilon^{1/2}\xi$, $\overline{x^2}(t)\sim\varepsilon\sigma(t)^2=\varepsilon e^{2t}$. We consider the relaxation time as the time when $\overline{x^2}(t)$ reaches 1, i.e., $\overline{x^2}(\tau)\sim 1$, the relaxation time is proportional to $\ln N$,

$$\tau\sim\ln N. \tag{B5}$$

In this way, we found out that the cut-off dependence could actually affect the relaxation time, but this cutoff played no role in the case of the relaxation near the spinodal point.

Hence, here we show that the relaxation time does not depend on the cutoff of $\xi$ if this cutoff is very large. If we denote the average of $\xi^n$ over $P(\xi)$ by $\overline{\xi^n}$, the time evolution of $\overline{\xi}$ is given by

$$\dot{\overline{\xi}}(t) = \varepsilon^{1/3}|\Lambda|^{1/2}[\pm\alpha+\gamma\overline{\xi^2}(t)] \geq \varepsilon^{1/3}|\Lambda|^{1/2}[\pm\alpha+\gamma\overline{\xi}(t)^2]. \tag{B6}$$

If $\xi(t_0)$ is larger than $\sqrt{\alpha/\gamma}\equiv\nu$, it can be shown from Eq. (B6) that

$$\bar{\xi}(t) \geq \frac{1+A(t)}{1-A(t)}\nu, \tag{B7}$$

where $A(t)$ is given by

$$A(t) = \frac{\bar{\xi}(t_0)-\nu}{\bar{\xi}(t_0)+\nu}\exp[2\varepsilon^{1/3}\Lambda^{1/2}\nu\gamma(t-t_0)]. \tag{B8}$$

The average of the scaled magnetization $\bar{\xi}(t)$ reaches infinity when $A(t)=1$, namely,

$$t-t_0 = \frac{1}{2\varepsilon^{1/3}|\Lambda|^{1/2}\nu\gamma}\ln\left(\frac{\bar{\xi}(t_0)+\nu}{\bar{\xi}(t_0)-\nu}\right). \tag{B9}$$

Because $t_0$ is finite, it takes only a finite time for $\bar{\xi}(t)$ to reach infinity. Therefore, there is no cut-off dependence on the relaxation time in the Fokker-Planck equation (16).

## APPENDIX C: DERIVATION OF EQ. (30) BY THE WKB APPROXIMATION

In the body of this paper, we estimated the relaxation time for $\Lambda<0$ and $|\Lambda|\gg 1$ according to Kramers' argument. Here, we give another derivation by using the WKB approximation. The following derivation is essentially the same as that of Tomita, *et al.* [12]. The Fokker-Planck Eq. (18) can be transformed into the "Schrödinger equation"

$$\frac{\partial Q}{\partial t} = \varepsilon^{1/3}\frac{\delta}{|\Lambda|}\frac{\partial^2 Q}{\partial \xi^2} - \varepsilon^{1/3}V(\xi)Q(\xi) \equiv \varepsilon^{1/3}\mathcal{H}Q(\xi) \tag{C1}$$

by substituting

$$P = \exp\left(-\frac{1}{2\delta}|\Lambda|^{3/2}U(\xi)\right)Q(\xi).$$

The scaled potential $U(\xi)$ is given by Eq. (19), and the Schrödinger potential $V(\xi)$ is

$$V(\xi) = \frac{1}{4\delta}|\Lambda|^2U'(\xi)^2 - \frac{1}{2}|\Lambda|^{1/2}U''(\xi). \tag{C2}$$

If the eigenvalues of $\mathcal{H}$ are $\lambda_i$ $(i=0,1,2,\ldots)$ and the eigenfunctions are $\phi_i$, we can expand $Q(\xi,t)$ as

$$Q(\xi,t) = \sum_i c_i\phi_i(\xi)e^{-\varepsilon^{1/3}\lambda_i t}. \tag{C3}$$

The lowest eigenvalue is $\lambda_0=0$, and the corresponding eigenfunction is

$$\phi_0(\xi) = \frac{1}{Z^{1/2}}e^{-|\Lambda|^{3/2}U(\xi)/(2\delta)}, \tag{C4}$$

which corresponds to the equilibrium state. $Z$ is the normalization factor for $\int\phi_0^2 d\xi=1$. The second lowest eigenfunction $\phi_1$ will represent the metastable mode, and the corresponding eigenvalue will be connected with the inverse of the lifetime of the metastable state, $\varepsilon^{1/3}\lambda_1 \sim 1/\tau$.

From the Schrödinger equation (C1),

$$0 = -\frac{1}{2m}\phi_0'' + V\phi_0, \tag{C5}$$

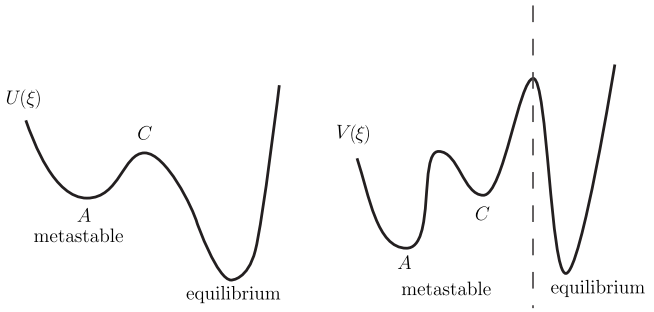


FIG. 4. Rough sketches of the scaled potential $U(\xi)$ and the corresponding Schrödinger potential $V(\xi)$.

$$\lambda_1\phi_1 = -\frac{1}{2m}\phi_1'' + V\phi_1. \tag{C6}$$

The mass $m$ is $m=|\Lambda|/(2\delta)$. If we multiply Eq. (C5) by $\phi_1$ and Eq. (C6) by $\phi_0$ and subtract the two equations, we obtain

$$\lambda_1\phi_0\phi_1 = \frac{1}{2m}(\phi_0'\phi_1 - \phi_1'\phi_0)'. \tag{C7}$$

Integrating this equation from $-\infty$ to the point $C$ (see Fig. 4), we obtain

$$\lambda_1 = -\frac{\phi_1'(C)\phi_0(C)}{2m\int_{-\infty}^{C}\phi_0\phi_1 d\xi} \tag{C8}$$

because $\phi_0'(C)=0$. Here, we consider the *metastable wave function* $\phi_{\rm ms}$, which corresponds to the localized canonical distribution at the valley of the potential. Hence, we assume for $\xi\lesssim C$

$$\phi_0 \approx u\phi_{\rm ms} \quad \text{for} \quad \xi\lesssim C, \tag{C9}$$

where $u$ is a constant given by

$$u^2 = \int_{-\infty}^{C}\phi_0(\xi)^2 d\xi \sim Z^{-1}\sqrt{\frac{2\pi\delta}{|\Lambda|^{3/2}U''(A)}}e^{-|\Lambda|^{3/2}U(A)/\delta}. \tag{C10}$$

Besides we assume that the first excited eigenfunction is also proportional to $\phi_{\rm ms}$ in the range $\xi\lesssim C$,

$$\phi_1 \approx v\phi_{\rm ms} \quad \text{for} \quad \xi\lesssim C, \tag{C11}$$

because $\phi_1$ is considered to represent the metastable mode. Under these assumptions, we get

$$\int_{-\infty}^{C}\phi_0\phi_1 d\xi = uv\int_{-\infty}^{C}\phi_{\rm ms}^2 d\xi \approx uv. \tag{C12}$$

Using the WKB approximation, it is obtained that

$$\phi_1'(C) \approx \sqrt{2mV(C)}\phi_1(C) = \frac{v}{u}\sqrt{2mV(C)}\phi_0(C). \tag{C13}$$

Substituting Eqs. (C12) and (C13) into Eq. (C8),

$$\lambda_1 = \frac{1}{u^2}\sqrt{\frac{V(C)}{2m}}\phi_0(C)^2. \tag{C14}$$

After some calculation, we obtain

$$\lambda_1 = \frac{1}{2}\sqrt{\frac{|\Lambda|}{\pi}|U''(A)U''(C)|}e^{-\beta\Delta F} \tag{C15}$$

where the free energy barrier $\beta\Delta F$ is $\beta\Delta F = |\Lambda|^{3/2}[U(C) - U(A)]/\delta$. Therefore, the lifetime of the metastable state $\tau$, which is equivalent with the relaxation time, is

$$\tau \sim 2N^{1/3}\sqrt{\pi}|U''(A)U''(C)|^{-1/2}|\Lambda|^{-1/2}e^{\beta\Delta F}. \tag{C16}$$

Comparing with the relaxation time obtained by Kramers' argument, Eq. (28), they agree with each other except for the minor difference in the constant prefactor.

## APPENDIX D: MONTE CARLO SIMULATION

We also obtained data by performing kinetic Monte Carlo simulations to confirm the data obtained by solving the master equation. In the Monte Carlo simulations, each data point is an average over 1000 samples, and the error bars are smaller than the symbol size in Fig. 2.

The algorithm of the Monte Carlo simulations is as follows. We choose a spin at a site $i$ randomly, and update the spin with the probability corresponding to the Glauber model given by Eq. (6)

$$\omega_M(\sigma_i \to -\sigma_i) = \frac{\exp\{-\beta\sigma_i[J(M-\sigma_i)/N + H]\}}{2\cosh\{\beta[J(M-\sigma_i)/N + H]\}}\Delta t,$$

where $\sigma_i$ is the spin state of the $i$-th spin ($\sigma_i = \pm 1$). In principle, a small time increment $\Delta t \ll 1$ is necessary to reproduce the result of the master equation given as a differential equation. However, we found almost the same result with the different time division, $\Delta t = 0.01$ and 1 for the quantities plotted in Fig. 2. Hence, we obtained the data with $\Delta t = 1$. During one Monte Carlo step we perform single spin flips $N$ times. Therefore, the time $t$ is related to the Monte Carlo step $s$ by $t = s$ because $\Delta t = 1$. The initial condition of each Monte Carlo simulation is set to the spinodal magnetization.


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