

Low-Temperature Nucleation in a Kinetic Ising Model with Soft Stochastic Dynamics

Kyungwha Park,^{1,2} Per Arne Rikvold,^{2,3} Gloria M. Buendía,^{2,3,4} and M. A. Novotny³

¹*Center for Computational Materials Science, Code 6390, Naval Research Laboratory, Washington D.C. 20375, USA*

²*School of Computational Science and Information Technology, Center for Materials Research and Technology, and Department of Physics, Florida State University, Tallahassee, Florida 32306-4120, USA*

³*Department of Physics and Astronomy and ERC Center for Computational Sciences, Mississippi State University, Mississippi State, Mississippi 39762-5167, USA*

⁴*Department of Physics, Universidad Simón Bolívar, Caracas 1080, Venezuela*

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We study low-temperature nucleation in kinetic Ising models by analytical and simulational methods, confirming the general result for the average metastable lifetime, $\langle\tau\rangle = A \exp(\beta\Gamma)$ ($\beta = 1/k_B T$) [E. Jordão Neves and R. H. Schonmann, *Commun. Math. Phys.* **137**, 209 (1991)]. Contrary to common belief, we find that both A and Γ depend significantly on the stochastic dynamic. In particular, for a “soft” dynamic, in which the effects of the interactions and the applied field factorize in the transition rates, Γ does *not* simply equal the energy barrier against nucleation, as it does for the standard Glauber dynamic, which does not have this factorization property.

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Nucleation is fundamentally important in disciplines ranging from biochemistry [1] to earth sciences [2], astrophysics [3], and cosmology [4], and it has been studied by kinetic Monte Carlo (MC) simulations in electrochemistry [5], materials science [6], magnetism [7], and atmospheric science [8], to mention just a few. However, many questions in nucleation theory are still unresolved, and recently there has been much interest in kinetic Ising systems as models for nucleation. In particular, much work has been done on their dynamical behavior at very low temperatures [9–14], where it is influenced by lattice discreteness. It is then possible to calculate exactly both the shape of the critical nucleus (the saddle-point configuration) and the most probable path during a nucleation event. In a typical numerical experiment, the system is prepared in a metastable state with all spins positive in a negative applied field. During each MC step (MCS), a randomly chosen spin is flipped with a configuration-dependent transition rate W that satisfies detailed balance, so that it will drive the system to thermodynamic equilibrium. The metastable lifetime is measured as the average number of MCS until the magnetization reaches zero. In the regime of single-droplet decay studied here, the lifetime measured in MCS is independent of the system size [9,11,15]. In the low-temperature limit the lifetime has been rigorously shown to be [9]

$$\langle\tau\rangle = Ae^{\beta\Gamma}. \quad (1)$$

Here the only dependence on the temperature T is through $\beta = 1/k_B T$, where k_B is Boltzmann’s constant (hereafter set equal to one). It is often assumed that Γ equals the energy difference between the saddle point and the metastable state [13,14], independent of the specific stochastic dynamic. In this Letter we show that this is not always so.

In particular, we describe two dynamics that both obey detailed balance but have different values of Γ and A for all values of the applied field, despite having the same saddle-point configuration.

At sufficiently low T , the saddle point was shown in Ref. [9] to be an $\ell \times (\ell - 1)$ rectangle of overturned spins with a “knob” of one overturned spin on one of its long sides. The critical length $\ell = \lfloor 2J/|H| \rfloor + 1$ for all $|H| \in (0, 4)$, where $\lfloor x \rfloor$ is the integer part of x . Here, $J > 0$ is the nearest-neighbor interaction constant of the Ising model, which will henceforth be set to unity. The critical length thus changes discontinuously at values of $|H|$ such that $2/|H|$ is an integer.

The square-lattice $S = 1/2$ Ising ferromagnet with unit interaction is defined by the Hamiltonian $\mathcal{H} = -\sum_{\langle\alpha,\beta\rangle}\sigma_\alpha\sigma_\beta - H\sum_\alpha\sigma_\alpha$, where the Ising spins $\sigma_\alpha = \pm 1$, H is the applied field, $\sum_{\langle\alpha,\beta\rangle}$ runs over all nearest-neighbor bonds on a square lattice, and \sum_α runs over all lattice sites. When this system evolves under a continuous-time Glauber dynamic with spin-flip rate [16]

$$W_G = [1 + \exp(\beta\Delta E)]^{-1}, \quad (2)$$

where ΔE is the energy change that would result from the flip, Γ in Eq. (1) is given by [9]

$$\Gamma_{\text{hard}} = 8\ell - 2|H|(\ell^2 - \ell + 1), \quad (3)$$

and from Ref. [12] $A = A_{\text{hard}} = 3/[8(\ell - 1)]$ for all $|H| < 2$. (See explanation of the subscript “hard” below.) The interpretation of Γ_{hard} is indeed the energy difference between the saddle point and the metastable state.

A characteristic feature of the Glauber dynamic is that it does not factorize into one part that depends only on the change in interaction energy, ΔE_J , and another that depends only on the change in the field energy, ΔE_H . Such

transition rates are known as hard [17]. Dynamics that do factorize this way are called “soft.” An example is the soft Glauber dynamic [18],

$$W_{SG} = [1 + \exp(\beta\Delta E_J)]^{-1} [1 + \exp(\beta\Delta E_H)]^{-1}. \quad (4)$$

In studies of field-driven Ising and solid-on-solid interfaces [18,19] it was recently shown that soft dynamics yield significantly different microscopic interface structures and mobilities than hard dynamics. Here we show that also the low-temperature nucleation properties with the soft Glauber dynamic differ significantly from those with the hard Glauber dynamic. In particular, Γ is not simply the energy difference between the saddle point and the metastable state, and the prefactor A is also different.

We obtain our results in three different ways. First, we calculate analytically by hand the first-passage time from the metastable state to an absorbing state just beyond the saddle point in an approximation that the path in configuration space corresponds to a simple one-step Markov process [20]. Second, we perform computer-aided analytical calculations using the technique of absorbing Markov chains (AMC) [10,21], allowing for multiple branching paths and “blind alleys.” Third, we perform simulations using the MC simulation with AMC (MCAMC) technique [10,22]. The first method provides the clearest physical insight, and for noninteger values of $2/|H|$ the results are fully confirmed by the other two.

The one-step Markov chain for $1 < |H| < 2$ ($\ell = 2$) corresponds to the configurations labeled $i = 0, \dots, 4$ in Fig. 1. The label i gives the number of overturned spins, such that the starting configuration has $i = 0$, and the saddle point has $i = i^* = 3$. In general the absorbing state is labeled $I \geq i^* \geq 1$. The mean time spent in state i is h_i . The rate at which the cluster grows from i to $i + 1$ overturned spins is g_i , and the rate with which it shrinks from i to $i - 1$ is s_i . These quantities satisfy the relation [20,23,24]

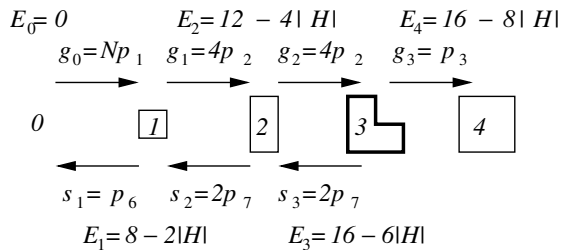


FIG. 1. The states in the one-step Markov chain of clusters of $i = 0, \dots, 4$ overturned spins, used to calculate the metastable lifetime $\langle\tau\rangle$ analytically by hand. The right-pointing arrows give the growth rates g_{i-1} , and the left-pointing arrows give the shrinkage rates s_i for $i = 1, 2$, and 3 . The energies E_i (relative to the metastable state, $i = 0$) are given at the top of the figure for even i and at the bottom for odd i .

$$h_{i-1} = (s_i h_i + N)/g_{i-1} \quad (5)$$

with boundary conditions $s_I = s_0 = 0$. The number N of sites in the system represents the total probability current through the Markov chain [20]. From Eq. (5) we obtain h_i recursively as $h_{I-1} = N/g_{I-1}$ and

$$h_i = \frac{N}{g_i} + \sum_{k=1}^{I-1-i} \frac{N}{g_{i+k}} \prod_{j=1}^k \left(\frac{s_{i+j}}{g_{i+j-1}} \right) \quad (6)$$

for $0 \leq i \leq I - 2$. Assuming that $I > i^*$ and the growth time for the supercritical droplet is negligible compared with the nucleation time [9,18,19], the lifetime $\langle\tau\rangle$ is the mean first-passage time to I , $\langle\tau\rangle = \langle\tau_I\rangle = \sum_{i=0}^{I-1} h_i$. Grouping the terms according to the “unpaired” factors N/g_{i+k} in Eq. (6) then yields

$$\langle\tau_I\rangle = \frac{N}{g_0} + \sum_{l=1}^{I-1} \frac{N}{g_l} \left(1 + \sum_{k=1}^l \prod_{j=0}^{k-1} \frac{s_{l-j}}{g_{l-j-1}} \right). \quad (7)$$

This result is general for any one-step Markov chain with absorption at I , regardless of the values of g_i and s_i [20]. However, for the Ising model the transition rates are related by detailed balance as $(s_i/n_i^s)/(g_{i-1}/n_{i-1}^s) = e^{\beta(E_i - E_{i-1})}$, where E_i is the energy of state i . The degeneracy factors n_i^s and n_{i-1}^s are the numbers of lattice sites at which a single spin-flip can shrink the cluster from i to $i - 1$ and analogously for growth from $i - 1$ to i , respectively. As a result, Eq. (7) becomes

$$\langle\tau_I\rangle = \frac{N}{g_0} + \sum_{l=1}^{I-1} \frac{N}{g_l} \left(1 + \sum_{k=1}^l e^{\beta(E_l - E_{l-k})} \prod_{j=0}^{k-1} \frac{n_{l-j}^s}{n_{l-j-1}^s} \right). \quad (8)$$

In the limit $\beta \rightarrow \infty$ Eq. (8) is dominated by the term or terms with the largest exponential factor. Their selection, which determines Γ and A , is described below, after we next find the spin-flip rates in the different dynamics.

For the square-lattice Ising system, the spins fall into ten classes [22], determined by the spin value σ (+ for the metastable direction and $-$ for the stable direction) and the number N_+ of its nearest neighbors that point in the metastable direction. The low-temperature limits of the rates p_m for flipping a spin in class m [Eqs. (2) and (4)], are shown in Table I.

Figure 1 shows a one-step Markov chain with $I = 4$. For $1 < |H| < 2$ the saddle-point configuration ($i = i^*$) is the L-shaped cluster with $\ell = 2$ ($i^* = 3$). Among the dominant terms in Eq. (8) is always the one with $k = l = i^* - 1$. For $0 < |H| < 2$, growth from $i^* - 1$ to i^* always involves adding a knob to one of the long sides of an $\ell \times (\ell - 1)$ rectangle, such that $g_{i^*-1} = 2\ell p_2$. From Table I we see that $p_2^{\text{soft}} = e^{-\beta 2|H|} p_2^{\text{hard}}$. For $2 < |H| < 4$, the saddle point is a single overturned spin ($\ell = 1$), so $i^* = 1$ and $g_{i^*-1} = g_0 = N p_1$. Again, the difference between Γ for the two dynamics is determined by the fact that $p_1^{\text{soft}} = e^{-\beta 2|H|} p_1^{\text{hard}}$. This yields our main result:

TABLE I. Rates of flipping a spin in class m , p_m , in the limit $\beta \rightarrow \infty$. Here $\sigma = + (-)$ corresponds to a spin in the metastable (stable) direction, and N_+ is the number of its nearest-neighbor spins in the metastable direction. The analytic form of p_m for the soft dynamic does not change with $|H|$.

m	σ	N_+	p_m^{soft}	p_m^{hard}	m	σ	N_+	p_m^{soft}	p_m^{hard}
1	+	4	$e^{-\beta 8}$	$e^{-\beta(8-2 H)}$ for $ H < 4$ 1/2 for $ H = 4$ 1 for $ H > 4$	6	-	4	$e^{-\beta 2 H }$	1 for $ H < 4$ 1/2 for $ H = 4$ $e^{\beta(8-2 H)}$ for $ H > 4$
2	+	3	$e^{-\beta 4}$	$e^{-\beta(4-2 H)}$ for $ H < 2$ 1/2 for $ H = 2$ 1 for $ H > 2$	7	-	3	$e^{-\beta 2 H }$	1 for $ H < 2$ 1/2 for $ H = 2$ $e^{\beta(4-2 H)}$ for $ H > 2$
3	+	2	1/2	1 for $ H > 0$	8	-	2	$e^{-\beta 2 H }/2$	$e^{-\beta 2 H }$ for all $ H $
4	+	1	1	1 for all $ H $	9	-	1	$e^{-\beta(4+2 H)}$	$e^{-\beta(4+2 H)}$ for all $ H $
5	+	0	1	1 for all $ H $	10	-	0	$e^{-\beta(8+2 H)}$	$e^{-\beta(8+2 H)}$ for all $ H $

$$\Gamma_{\text{soft}} = \Gamma_{\text{hard}} + 2|H| \quad \text{for } 0 < |H| < 4. \quad (9)$$

We emphasize that Eq. (9) is valid as $T \rightarrow 0$ for all $|H| \in (0, 4)$. For $|H| > 4$ the lifetime is the first-passage time to one overturned spin, so that $\langle \tau \rangle = \langle \tau_1 \rangle = 1/p_1$, which

$$\begin{aligned} \langle \tau_4 \rangle &= \frac{1}{p_1} + \frac{1}{4p_2} [N + e^{\beta(8-2|H|)}] + \frac{1}{4p_2} \left[N + \frac{N}{2} e^{\beta(4-2|H|)} + \frac{1}{2} e^{\beta(12-4|H|)} \right] \\ &+ \frac{1}{p_3} \left[N + \frac{N}{2} e^{\beta(4-2|H|)} + \frac{N}{4} e^{\beta(8-4|H|)} + \frac{1}{4} e^{\beta(16-6|H|)} \right] \equiv \mathcal{A} + \mathcal{B} + \mathcal{C} + \mathcal{D}. \end{aligned} \quad (10)$$

Using p_m from Table I we identify the dominant terms in $\langle \tau_4 \rangle$, and from these we obtain A and Γ for both the soft and hard Glauber dynamics for all $|H| > 1$. (Analogous calculations can be carried out for arbitrarily small $|H|$.)

Soft dynamic: For $1 < |H| < 2$ the sum is dominated by the last term in \mathcal{C} , yielding $A_{\text{soft}} = 1/8$ and $\Gamma_{\text{soft}} = 16 - 4|H|$. For $|H| = 2$ it is dominated by \mathcal{A} and the last terms in \mathcal{B} and \mathcal{C} , yielding $A_{\text{soft}} = 11/8$ and $\Gamma_{\text{soft}} = 8$. For $|H| > 2$ it is dominated by \mathcal{A} , yielding $A_{\text{soft}} = 1$ and $\Gamma_{\text{soft}} = 8$. See Fig. 2.

Hard dynamic: For $1 < |H| < 2$ the sum is dominated by the last terms in \mathcal{C} and \mathcal{D} , yielding $A_{\text{hard}} = 3/8$ and $\Gamma_{\text{hard}} = 16 - 6|H|$. For $|H| = 2$ it is dominated by \mathcal{A} and the last terms in \mathcal{B} , \mathcal{C} , and \mathcal{D} , yielding $A_{\text{hard}} = 2$ and $\Gamma_{\text{hard}} = 4$. For $2 < |H| < 4$ it is dominated by \mathcal{A} and the last term in \mathcal{B} , yielding $A_{\text{hard}} = 5/4$ and $\Gamma_{\text{hard}} = 8 - 2|H|$. (These results agree with corresponding ones in Refs. [13,14].) For $|H| \geq 4$ the system is unstable and $\langle \tau \rangle = \langle \tau_1 \rangle = \mathcal{A}$, yielding $\Gamma_{\text{hard}} = 0$ and $A_{\text{hard}} = 2$ for $|H| = 4$ and $A_{\text{hard}} = 1$ for $|H| > 4$. See Fig. 2.

We further performed computer-aided analytic calculations of $\langle \tau \rangle$ with Mathematica [25] by the AMC method [10,21], using three different classifications of the configurations: 12 transient and 9 absorbing states (denoted 12/9), as well as 7/13 and 13/13. For noninteger $2/|H|$ the results were *identical* to the 1-step approximation. However, for $|H| = 2$, A (but not Γ) was found to depend slightly on the numbers of states included in the

yields $\Gamma_{\text{soft}} = 8$ and $\Gamma_{\text{hard}} = 0$. Thus, in contrast to the hard dynamic, nucleation with the soft dynamic is always activated, even for infinitely strong fields.

To obtain the prefactors A_{soft} and A_{hard} in the two dynamics, we explicitly write out the four terms obtained from Eq. (8) for $I = 4$ with s_i , g_{i-1} , and E_i from Fig. 1:

calculation for both dynamics. With the numbers of states used, these differences were less than 0.5%. Specifically, 12/9 yielded $A_{\text{hard}} = 78\,244/45\,597 \approx 1.7160$ (1.764 by a different method in Ref. [13]) and $A_{\text{soft}} = 943/704 \approx 1.3395$. See Fig. 2.

Both sets of analytic results were checked by MC simulations for both dynamics, using the MCAMC method [10,22]. The system size was $L = 24$, and 2000 escapes were used (6000 for $|H| \geq 4$). The parameters Γ and A were determined from weighted two-parameter linear least-squares fits to plots of $T \ln \langle \tau \rangle$ vs T [inset in Fig. 2(a)]. As seen in Fig. 2, the simulation results agree with the analytical results to within two standard errors.

In conclusion, we have confirmed Eq. (1) [9] for the low-temperature metastable lifetime of a kinetic Ising model, both analytically and by MC simulations, finding both Γ and A to depend on the specific stochastic dynamic for all values of the applied field. For a soft Glauber dynamic [Eq. (4)], Γ does *not* equal the energy difference between the critical cluster and the metastable state for any value of the field and it also does *not* vanish in the strong-field limit, as it does for the conventional, hard Glauber dynamic [Eq. (2)]. Thus, nucleation under the soft dynamic remains an activated process for arbitrarily strong fields. These results are consistent with recent studies of the microstructure and mobility of field-driven Ising and solid-on-solid interfaces [18,19]. They indicate

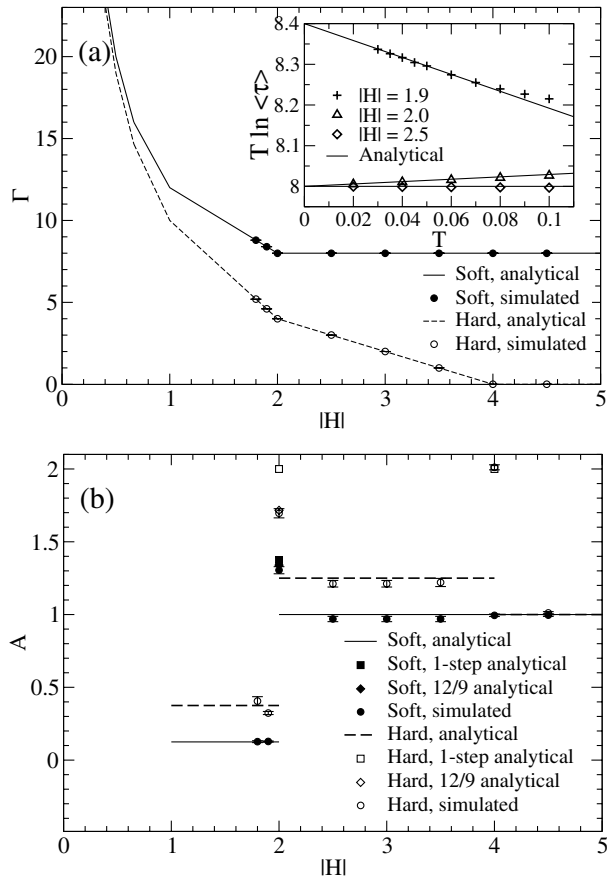


FIG. 2. Analytical and simulated results for the soft and hard Glauber dynamics for (a) Γ and (b) A . In the legends, “one-step analytical” refers to the one-step Markov-chain approximation, “12/9 analytical” to the computer-aided AMC calculations with 12 transient and 9 absorbing configurations, and “analytical” to results that are identical for all the analytical calculations. The results only differ for $|H| = 2$. The inset in (a) shows analytical (lines) and MC (data points) results for $T \ln \langle \tau \rangle$ vs T for the soft dynamic, from which Γ and A are obtained.

that great caution must be shown in formulating and interpreting stochastic models of physical systems, as even seemingly minor modifications of the transition probabilities can significantly affect the nucleation rates. It might thus be interesting to study the influence of the stochastic dynamic on dynamic phase transitions in kinetic Ising models [26]. We also note that, although our results are derived for a specific model system, qualitatively similar results should apply to kinetic MC simulations for nucleation in a wide range of scientific disciplines. On the positive side, experimental observation of the field and temperature dependences of nucleation and growth could help devise correct stochastic models of nonequilibrium phenomena.

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