Nucleation theorems applied to the Ising model

Hanna Vehkamäki* and Ian J. Ford
Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, United Kingdom
(Received 28 January 1999)

We use Monte Carlo simulations to study a single cluster of ''up'' spins in a sea of ''down'' spins in the three-dimensional Ising model. We evaluate the growth and decay rates for clusters of different sizes, identify the critical size for which these rates are equal, and obtain the internal energy of the critical size cluster. The results of the simulations at different temperatures and magnetic fields are used together with the first and second nucleation theorems to predict how the cluster nucleation rate changes when the external magnetic field and the temperature are changed. Our results are in agreement with literature values, but our method requires significantly less computational effort than the simulations reported earlier and avoids the difficult evaluation of free energies. [S1063-651X(99)08206-9]

PACS number(s): 05.50.+q, 02.70.Lq, 64.60.Qb

I. INTRODUCTION

The nucleation of clusters of spins in Ising systems has been studied widely in the literature (see, e.g., [1,2]). The conventional direct method to obtain information about nucleation is to follow the evolution of a population of clusters of different sizes in a large system. The critical size for which decay is just as likely as growth is obtained from the size distribution of clusters or the size dependence of the cluster free energy. Counting the number of clusters reaching the critical size or following the evolution of the magnetization then gives the nucleation rate.

In contrast, we study a single cluster and determine its growth and decay rates to obtain the critical size. Our simulations give the size and the excess internal energy of the critical cluster as functions of temperature and external magnetic field. We use this information in conjunction with the first and second nucleation theorems [3–6] to obtain the *derivatives* of the nucleation rate with respect to temperature and external field. If we know the nucleation rate for one set of conditions, we can therefore use the nucleation theorems to predict the nucleation rates for other temperatures and fields. Our predictions are consistent with the nucleation rates reported in earlier studies as well as with the predictions of classical nucleation theory.

The required number of lattice points in our three-dimensional (3D) Ising model calculations is of the order of 10^3, whereas the studies involving the entire population of clusters require lattices with more than 100^3 points. The amount of CPU time needed to extract the size and energy of the critical cluster using our method is about 1% of the time needed for direct nucleation simulations.

II. THEORY

The Ising lattice is an array of magnetic particles (spins) that interact with each other and with an external field. The energy of the system is given by

$$E = -J\sum_{\langle lm\rangle} s_l s_m - h\sum_l s_l , \tag{1}$$

where $\langle lm\rangle$ denotes summation over nearest neighbors (each pair counted only once), J is the spin-spin coupling constant, and h is the parameter describing the interaction with an external field. ($h=\mu H$, where μ is the magnetic dipole moment of the spin particles and H is the external magnetic field.) The values of the z component of the spins s_l are restricted to ± 1. For a three-dimensional cubic lattice the coupling constant may be expressed in terms of the numerically determined critical temperature T_c according to $J/kT_c = 0.221\,656$ [7]; T_c marks the transition temperature between the ferromagnetic and paramagnetic states at $H=0$.

The kinetics of nucleation can be analyzed if the rates of growth and decay of a cluster of i up spins can be evaluated [8]. The steady-state rate of nucleation of clusters I is the net number of clusters that grow through the critical size per Monte Carlo step and lattice site. The mean rates of growth and decay of an i cluster in the Monte Carlo stochastic dynamics are written β_i and γ_i, respectively. Under conditions where the population of isolated up spins in the system divided by the number of lattice sites N is n_1, I is given by

$$I = \frac{\beta_1 n_1}{1+\sum_{i=2}^{\infty}\prod_{j=2}^{i}(\gamma_j/\beta_j)} . \tag{2}$$

Upon applying the principle of detailed balance to relate the γ_i to the β_i, we obtain $I=[N\Sigma_{i=1}^{\infty}P(i)]^{-1}$, where $P(i) = \beta_i^{-1}\exp[F_x(i)-2hi]/kT$, k is Boltzmann's constant, and $F_x(i)$ is the excess Helmholtz free energy of the i cluster [6]. From these relations it is easy to prove that

$$\left(\frac{\partial \ln I}{\partial(2h/kT)}\right)_T = \langle i\rangle + \left\langle \left(\frac{\partial \ln \beta_i}{\partial(2h/kT)}\right)_T \right\rangle , \tag{3}$$

where the angular brackets denote an average weighted by the $P(i)$, for example, $\langle g(i)\rangle = \Sigma_{i=1}^{\infty} g(i)P(i)/\Sigma_{i=1}^{\infty}P(i)$, for some function $g(i)$. If $P(i)$ is sharply peaked, the averages

*On leave from Department of Physics, P.O. Box 9, 00014 University of Helsinki, Finland.

can be expressed as $\langle g(i)\rangle \approx g(i^*)$, where the critical size i^* is the size of cluster for which $\gamma_i = \beta_i$, which maximizes $P(i)$.

This allows us to derive the first nucleation theorem [3,4] for the Ising model:

$$\left(\frac{\partial \ln I}{\partial(2h/kT)}\right)_T = i^* + \left(\frac{\partial \ln \beta_i}{\partial(2h/kT)}\right)_{T,i}^{i^*} \approx i^* + 1. \tag{4}$$

The final form has been obtained using an approximate analysis of β_i, which is described in the Appendix. Furthermore, the second nucleation theorem [5,6] takes the form

$$\left(\frac{\partial \ln I}{\partial T}\right)_{h/kT} = \frac{E_x^*}{kT^2} + \left(\frac{\partial \ln \beta_i}{\partial T}\right)_{h/kT,i}^{i^*} \approx \frac{E_x^*}{kT^2} + \frac{2J}{kT^2}, \tag{5}$$

again using an approximate form for the growth rate β_i to obtain the final term. The superscript i^* means that the value $i = i^*$ is inserted after evaluating the derivative. The excess internal energy of a critical cluster E_x^* is defined as the difference in the coupling part of the internal energy [the first term in Eq. (1)] between a system containing one cluster in a sea of down spins and a homogeneous system (all spins up). All these results are independent of the cluster definition.

Our simulations, described in Sec. III, produce data for the size and excess internal energy of the critical cluster as functions of temperature T and external field parameter h. By integrating the nucleation theorems we can then determine the behavior of the nucleation rate $I(h/T,T)$ when the temperature and the external field are changed. The integration constant can be determined if the nucleation rate is known at a reference temperature T_0 and magnetic field h_0. Integrating Eqs. (4) and (5) gives

$$\ln I(h/T_0,T_0) - \ln I(h_0/T_0,T_0) = \int_{h_0/kT_0}^{h/kT_0} 2(i^*+1)\, d\left(\frac{h}{kT_0}\right) \tag{6}$$

and

$$\ln I(h_0/T_0,T) - \ln I(h_0/T_0,T_0) = \int_{T_0/T_c}^{T/T_c} \left(\frac{E_x^*}{kT}\frac{T_c}{T} + \frac{2J}{kT_c}\frac{T_c^2}{T^2}\right) d\left(\frac{T}{T_c}\right). \tag{7}$$

These results have been written in a manner that makes explicit the dimensionless form of the integrands and integration variables. The properties i^* and E_x^* of the critical cluster are functions of h and T.

The classical formulas for the Ising model are presented here for completeness since we compare the results of our simulations with the predictions of the classical theory. According to the classical theory [8,9], the nucleation rate has the form $I = Z\beta_{i*} n_1 \exp[-\Delta F(i^*)/kT]$, where, for Ising systems, $\Delta F(i)$ is the (Helmholtz) free energy associated with the formation of an i cluster, Z is the Zeldovich factor, and n_1 is the concentration of isolated up spins. The free energy of formation is given by $\Delta F(i) = \Gamma kT i^{2/3} - 2hi$, where Γ is a dimensionless parameter related to the surface tension, taken from Heermann *et al.* [10] (note that $h_{\text{Heermann}} = 2h_{\text{this work}}$). Thus the critical cluster determined by the condition $\partial \Delta F(i)/\partial i = 0$ has the size $i^* = [\Gamma kT/3h]^3$ and the free energy of formation $\Delta F(i^*)/kT = 4\Gamma^3/27(2h/kT)^2$. The Zeldovich factor takes the form

$$Z \equiv \left[\frac{-1}{2\pi kT}\left(\frac{\partial^2 \Delta F(i)}{\partial i^2}\right)^{i^*}\right]^{1/2} = \frac{1}{3}\sqrt{\frac{\Gamma}{\pi}}\, i^{*(-2/3)}. \tag{8}$$

The growth rate is $\beta_{i^*} \propto i^{*(2/3)}$ and n_1 is proportional to the exponential of the energy of a single isolated up spin, $n_1 \propto \exp[2(h-6J)/kT]$. The nucleation rate can only be evaluated up to an unknown proportionality factor. Finally, the excess internal energy at the critical size in the classical framework is given by

$$E_x = \Delta F_x + T\Delta S_x = \Delta F_x - T(\partial \Delta F_x/\partial T) = -kT^2\left(\frac{\Gamma kT}{3h}\right)^2 \frac{\partial \Gamma}{\partial T}, \tag{9}$$

where $F_x = \Gamma kT i^{2/3}$.

III. SIMULATIONS

We use a simple cubic lattice and set up a cluster of i spins with $s_l = +1$ surrounded by a sea of spins with $s_l = -1$. A spin is defined to be part of the cluster if it and at least one of its nearest neighboring spins have spin value $s_l = +1$. We generate a sequence of configurations representative of the canonical ensemble by following the Metropolis [11] scheme, such that the probability for a spin to flip is $\min(1, \exp[-\Delta E/kT])$, where ΔE is the change in the energy of the system due to the flip.

Our strategy is to find the critical cluster kinetically. Clusters above the critical size tend to grow in the stochastic dynamics of the Monte Carlo simulation. This is a reflection of the fact that the system free energy can be reduced by doing so. Similarly, clusters smaller than the critical size tend to shrink, for the same reason. The critical size has equal rates of growth and decay or, equivalently, it denotes the cluster with the highest free energy.

In principle, simply by observing the evolution histories of many individual clusters under the Monte Carlo dynamics, the relative rates of growth and decay as a function of size may be extracted. However, the tendency for clusters to move away from the critical size means that information in this important region will be relatively sparse. Instead, we calculate growth and decay rates as ensemble averages of certain well-defined quantities at a fixed cluster size. In this way, uniform statistics may be gathered over the interesting range of cluster sizes.

The simulation begins with a single up spin in the middle of the lattice. This seed is grown by selecting a nearest-neighbor lattice point at random and flipping it according to the Metropolis probability. This ''select-and-try-to-flip'' procedure is repeated until the cluster has been grown to a desired size. After the growth and decay rates have been calculated for this particular size, we can grow or shrink the cluster to any other size.

The calculational procedure for a cluster of a particular size can then be divided into two parts. One task is to obtain the probabilities that a particular configuration of the cluster

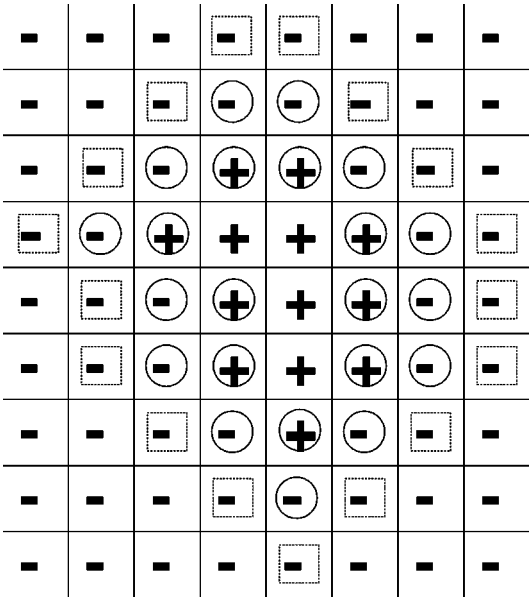

FIG. 1. Schematic picture of a cluster in the Ising lattice; see the text for details.

should grow or decay in the next Monte Carlo step and the other task is to change the configuration appropriately to obtain ensemble averages of these probabilities.

To evaluate the *decay* probability for a particular realization (shape) of the cluster, we go through all the spins with $s_l=1$ that are on the surface of the cluster (the circled plusses in Fig. 1) and add up the Metropolis probabilities for a spin flip. The *growth* probability for the Monte Carlo step is obtained by adding up the flip probabilities of the boundary spins that are nearest neighbors to the cluster (the circled minuses in Fig. 1). The expressions for the total probabilities for growth and decay are derived in the Appendix.

Note that we do not actually implement a growth or decay step in this procedure, except in the following case. We allow changes in the configuration (shape) of the cluster by first applying the select-and-try-to-flip procedure to the nearest-neighbor sites of the cluster (the circled plusses in Fig. 1) until the cluster has grown by one spin. Then the select-and-try-to-flip procedure is applied to the spins that are on the surface of the cluster (the circled minuses in Fig. 1) until the cluster has shrunk back to its original size. Every time a flip occurs, the list of surface spins and the list of nearest-neighbor spins are updated accordingly. Kawasaki dynamics [12] could of course be used to simulate this shape exploration at a fixed cluster size, but for simplicity we retain the Metropolis scheme.

This procedure clearly restricts the cluster to change size only by one spin at a time, located at the cluster surface. It turns out that this is slightly too restrictive for this system. However, a simple modification allows additional growth events to be taken into account.

In simulations where the potential spin flips are not limited to the region adjacent to the cluster, occasional events occur where clusters merge: A change in size greater than unity is achieved in a single Monte Carlo step. The most frequent event of this type is where a growing cluster merges with a single isolated up spin. If such a spin existed at a site two lattice spacings removed from a cluster spin, then the successful flip of the intervening spin would increase the size of the cluster by two spins in one Monte Carlo step.

We can estimate the likely importance of this effect. For the 3D cubic lattice, the probability for an isolated down spin to flip per attempt is $\exp[2(h-6J)/kT]$, which is around 0.02 for the conditions studied. The number of second nearest neighbors (the squared minuses in Fig. 1) is estimated to be of the order of $4(i^{2/3}+2i^{1/3})$, which is the result for a cubic cluster if $i^{1/3}$ is an integer number. This implies that for $i>8$ there is likely to be at least one spin with $s_l=1$ among them if the system is allowed to evolve without constraining the flips to the boundary region. We call these *satellite spins*.

It turns out that we have to take the effect of satellite spins into account to reproduce the results of earlier studies. To do so, when adding up the probabilities for boundary spins to flip, we also calculate the probability for the boundary spin to have n_s satellite spins ($n_s=1,\ldots,5$), multiply this by the probability for a spin with n_s satellites to flip, and add the resulting probability to the appropriate growth probability. All the flips leading to size $(i+1)$ (for example, the merging of one satellite with a cluster of size $i-1$) are added to the growth probability of size i. Similarly, we account for the fact that a decay may occur due to the flip of a spin *inside* the cluster, but this has a minor effect on the results. We therefore make a near-exact evaluation of the growth and decay probabilities per Monte Carlo step for a particular configuration and sampling is only involved when changing the shape of the cluster.

The critical size is found by studying different cluster sizes and identifying the size for which the growth and decay rates are equal. When the critical size is known, it is straightforward and fast to average the coupling part of the internal energy over different shapes to get the excess internal energy E_x^*.

The lattice size for all our calculations was chosen to be $10\times10\times10$, although test calculations with different lattice sizes were performed to make sure that the results are independent of this choice. Moreover, the simulation was terminated if the cluster reached the lattice boundaries, so we do not need to specify boundary conditions. An additional check was performed by using a lattice with periodic boundary conditions and allowing the cluster to change its size freely. During the control simulation the excess energy and the size of the cluster were recorded, allowing us to calculate the average energy of the cluster as a function of size. Since the average energies obtained from the actual simulation and lengthier free growth and decay runs agree, we are confident that we are sampling the shape space correctly.

The simulations for one temperature and magnetic field were completed in about 5 min using an ALPHA EV5 333-MHz workstation with one processor. In comparison, Acharyya and Stauffer [13] report that they used a lattice of size 256^3 and CPU time of around 1.5 h on a CRAY-T3E with 32 processors to perform their direct nucleation simulations. Thus, compared to direct nucleation simulations, our method is significantly faster. The difference in time is naturally due to the fact that we can perform the simulation in much smaller lattices and to the fact that we restrict the trial flips to the boundary region around the cluster. We can also confine our simulation to the sizes around the critical size to identify the exact watershed, which is a further economy.

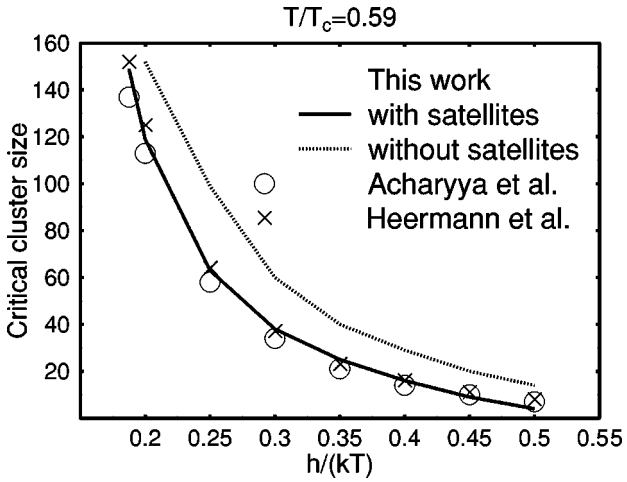

FIG. 2. Critical cluster size as a function of external field parameter h, compared with other studies. The predictions of classical theory coincide with the results of Heermann *et al.* [10].

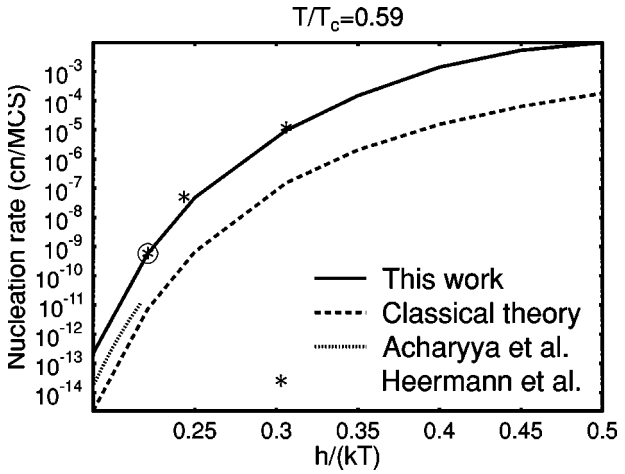

FIG. 3. Nucleation rate as a function of the magnetic field parameter h. The unit of the nucleation rate is the number of critical nuclei formed per Monte Carlo step and lattice site. The circled point was used to determine the integration constant required in our calculations. The vertical positionings of the dashed lines are arbitrary and they have been placed close to our results for clarity.

The present method of accounting for the effect of satellite spins is not expected to work well when the clusters are less compact, which is the case when the temperature approaches the critical temperature T_c. To find the critical size in these cases, we would have to abandon the idea of focusing on a single cluster and take into account the merging of two clusters explicitly. We would no longer be able to concentrate entirely on spin flips in the boundary region around the cluster, with a small perturbation due to the effect of satellites.

IV. RESULTS

We focused on conditions for which nucleation rates obtained from direct simulation have been reported, namely, a temperature $T=0.59T_c$ [10,13], and compared our results with these earlier studies as well as the predictions of classical nucleation theory. Figure 2 shows the critical cluster size as a function of magnetic field parameter at this temperature. The agreement between our results and the earlier results of Heermann *et al.* [10] and Acharyya and Stauffer [13] is very good. The sizes given by Heermann *et al.* [10] agree with the predictions of classical theory. We also show the critical sizes obtained by our method when neglecting the effect of satellite spins upon the growth and decay rates. In this case the growth rates are clearly underestimated and the critical sizes are larger.

Figure 3 shows the behavior of the nucleation rate as the magnetic field is changed, while the temperature is kept constant at $T_0=0.59T_c$. We use Eq. (6) to predict the nucleation rate, having chosen reference values $h_0/kT_0=0.221\,15$ and $I_0=I(h_0/T_0,T_0)=5.81\times10^{-10}$. This reference nucleation rate is the number of critical nuclei formed per Monte Carlo step and lattice site calculated using direct simulation by Heermann *et al.* [10] (the circled point in Fig. 3). Our predictions agree well with the further results of Heermann *et al.* [10].

Acharyya and Stauffer [13] report the nucleation times in this system, which are taken to be inversely proportional to the nucleation rate. Since the proportionality factor is unknown, only the slope of the dashed line representing their results in Fig. 3 is significant. The classical theory predictions suffer from the same lack of information about the proportionality constant. For convenience the vertical displacement of these curves is arbitrarily chosen so that they lie close to the line representing our results and it is clear that the slopes are in good agreement.

Unfortunately, direct simulations of nucleation appear only to have been carried out in the literature for the single temperature $0.59T_c$. Nevertheless, we are able to compare our results with classical theory for other temperatures. We calculated the excess internal energies for the critical cluster at temperatures between $0.54T_c$ and $0.70T_c$ with $h_0/kT_0=0.243\,35$, and performed the integration in Eq. (7). The excess internal energy of the critical cluster and the critical cluster size as a function of the temperature are shown in Fig. 4, together with the classical theory predictions. The deviations from classical theory are seen to be small. The temperature dependence of the nucleation rate obtained using Eq. (7) and the classical predictions are shown in Fig. 5. The reference values were $T_0=0.59T_c$ and $I_0=4.44\times10^{-8}$, again

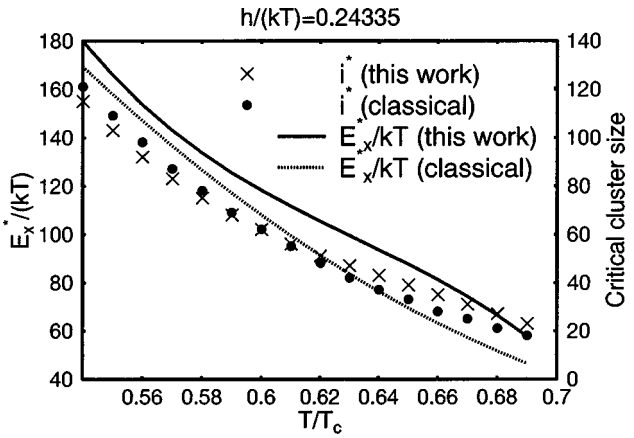

FIG. 4. Critical cluster size (i^*) and excess internal energy (E_x^*) of the critical cluster as a function of temperature.

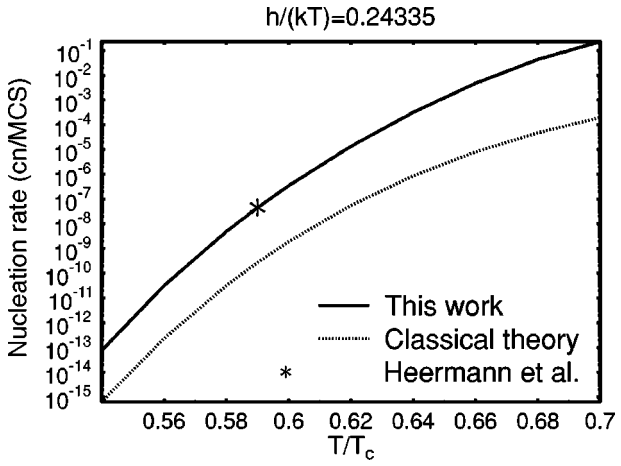

FIG. 5. Nucleation rate as a function of temperature. The unit of the nucleation rate is the number of critical nuclei formed per Monte Carlo step and lattice site. The point marked with an asterisk, taken from the work of Heermann *et al.* [10], was used to determine the integration constant in our rates. The vertical positioning of the dashed line is arbitrary.

taken from Heermann *et al.* [10]. Again, only the slope of the classical curve is significant.

Since it is our aim to demonstrate how the second nucleation theorem can be used to extrapolate nucleation rate data to new regions of parameter space, we present the rates, rather than their derivatives. The slope of the classical curve agrees well with our results at the low temperatures studied, but starts to deviate when the temperature is higher. When applied to the formation of liquid droplets from supersaturated vapors, classical theory tends to predict nucleation rates with an incorrect temperature dependence and so it is interesting to note that it behaves in the same way in the case of the Ising model.

Finally, our simulations provide numerical values of the growth rates β_i and so we are able to test the validity of the approximation made in obtaining the right-hand sides of Eqs. (4) and (5). We found that the derivative $[\partial \ln \beta_i/\partial(2h/kT)]^{i^*}_{T,i}$ is within 30% of the assumed value of unity for all the conditions studied and that the derivative $(\partial \ln \beta_i/\partial T)^{i^*}_{h/kT,i}$ is about three times the assumed value $2J/kT^2$. These deviations from the values used in Eqs. (4) and (5) do not affect the calculated nucleation rates significantly.

V. CONCLUSIONS

We used single-cluster simulations to determine kinetically the size and excess internal energy of the critical cluster in a 3D cubic Ising model at various temperatures and external magnetic fields. The critical size is that which is equally likely to grow and decay, and our results agree with those reported earlier. The advantage of our method is that results are produced with remarkably less computational effort. We concentrate on single clusters rather than populations of clusters, which allows us to use small lattice sizes. We also avoid the laborious task of evaluating free energies.

We can predict the dependence of the nucleation rate on the external magnetic field using the first nucleation theorem and our simulation data for the critical size. We find that a one-site growth mechanism is not accurate enough to describe the nucleation rate observed in direct nucleation studies. The effect of the merging together of one or more satellite spins with the cluster has to be taken into account.

We demonstrate how the second nucleation theorem and the easily obtainable data for the ensemble-averaged excess internal energies of critical clusters can be used to predict the temperature dependence of the nucleation rate. Direct simulation studies at a wider range of temperatures could be made in order to check these predictions. In the near future the simulation technique outlined here will be used to study the nucleation of molecular clusters.

ACKNOWLEDGMENTS

This work was funded by the United Kingdom Engineering and Physical Science Research Council (EPSRC) under Grant No. GR/L78499 and the Academy of Finland (Project No. 41886). H.V. thanks K. Arstila for useful discussions.

APPENDIX

In this appendix, we go through an evaluation of the growth and decay rates β_i and γ_i. We define these to be the probability per Monte Carlo step that the cluster will change size by plus or minus one. A Monte Carlo step is the random selection of a site in the Ising lattice followed by an attempt to flip the spin at that site.

The probability that a particular site j should be selected is simply $1/N$, where N is the number of sites in the lattice. The probability that cluster growth should then result is the product of three factors: first, a factor ω_j, which is zero if the spin is already up and unity otherwise; then a factor C_j, which is zero if the spin if flipped, would create a configuration disallowed under the cluster definition, but unity otherwise; finally, a factor p_j, given by the Metropolis function. The total probability for growth given a certain configuration K is then the sum of these probabilities over all sites in the lattice: $\beta_i^K = N^{-1}\Sigma_j \omega_j C_j p_j$. In practice, ω_j and C_j are unity only for down spins adjacent to the cluster: the sites shown as circled minus signs in Fig. 1. The mean growth probability β_i is then just the ensemble average of β_i^K over all configurations K of a single cluster of size i.

Similarly, the probability for growth given a particular configuration K is given by $\gamma_i^K = N^{-1}\Sigma_j(1-\omega_j)C_j p_j$ and in practice the sum is restricted to the outermost sites of the cluster: the circled plus signs in Fig. 1. The flip of a spin within the body of the cluster can be taken into account, but it has only a minor effect at the temperatures we are studying. This expression is then averaged over the ensemble. In fact, we should consider the probability per sweep of the lattice to be the correct growth rate since β_i and γ_i would otherwise be system size dependent, but this detail is not important for our purposes.

The expression for β_i^K can be used to estimate the derivatives required to simplify Eqs. (4) and (5). It turns out that the Glauber function [14] for the flip probability is more useful for this purpose than the Metropolis function. We write the simpler expression $\beta_i^K = N^{-1}\Sigma_k p_k^G$, where the index k runs over allowed sites for growth, where

$$p_k^G = \frac{1}{2}\left[1 - \tanh\left(\frac{\Delta E}{2kT}\right)\right],$$

where ΔE is the change in energy of the configuration associated with the growth event, and then $\beta_i = \langle \beta_i^K \rangle_i$ where the angular brackets denote ensemble averaging over configurations K of a cluster of size i. For growth, we have $\partial \Delta E / \partial h = -2$ [see Eq. (1)]. Therefore,

$$\left(\frac{\partial \beta_i}{\partial (2h/kT)}\right)_{T,i} = \left\langle N^{-1} \sum_k p_k^G [1 + \exp(-\Delta E/2kT)]^{-1} \right\rangle_i$$

and so

$$\left(\frac{\partial \beta_i}{\partial (2h/kT)}\right)_{T,i} \leqslant \left\langle N^{-1} \sum_k p_k^G \right\rangle_i = \beta_i,$$

so that $(\partial \ln \beta_i / \partial (2h/kT))_{T,i} \leqslant 1$.

The derivative of β_i with respect to T is less easy to characterize, since the statistical weight of the configurations depends on T. If we ignore this fact, the previous derivation may be repeated, yielding the result

$$\left(\frac{\partial \beta_i}{\partial T}\right)_{h/kT,i} = \left\langle \sum_k p_k^G \frac{2 \ell_k J/kT^2}{N[1 + \exp(-\Delta E/2kT)]} \right\rangle_i,$$

where ℓ_k is the change in the number of up-down nearest-neighbor pairs produced by the spin flip at site k when in configuration K. Even this expression cannot be analyzed further: We simply assume that ℓ_k can be replaced by a mean value of order one and replace the term in square brackets in the denominator by unity to obtain

$$\left(\frac{\partial \beta_i}{\partial T}\right)_{h/kT,i} \sim 2J/kT^2 \beta_i,$$

so that $(\partial \ln \beta_i / \partial T)_{h/kT,i} \sim 2J/kT^2$, which is the approximation used in Eq. (5).

[1] K. Binder and H. Müller-Krumbhaar, Phys. Rev. B **9**, 2328 (1973).
[2] P. A. Rikvold, H. Tomita, S. Miyashita, and W. Sides, Phys. Rev. E **49**, 5080 (1994).
[3] Y. Viisanen, R. Strey, and H. Reiss, J. Chem. Phys. **99**, 13 (1993).
[4] D. W. Oxtoby and D. Kashchiev, J. Chem. Phys. **100**, 7665 (1994).
[5] I. J. Ford, J. Chem. Phys. **105**, 8324 (1996).
[6] I. J. Ford, Phys. Rev. E **56**, 5615 (1997).
[7] D. Stauffer, Int. J. Mod. Phys. C **3**, 287 (1992).
[8] R. Becker and W. Döring, Ann. Phys. (Leipzig) **24**, 719 (1935).
[9] J. Zeldovich, Sov. Phys. JETP **12**, 525 (1942).
[10] D. W. Heermann, A. Coniglio, W. Klein, and D. Stauffer, J. Stat. Phys. **36**, 447 (1984).
[11] N. Metropolis *et al.*, J. Chem. Phys. **21**, 1087 (1953).
[12] K. Kawasaki, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1972), Vol. 2, p. 443.
[13] M. Acharyya and D. Stauffer, Eur. Phys. J. B **5**, 571 (1998).
[14] R. J. Glauber, J. Math. Phys. **4**, 294 (1963).