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Evaporation/Condensation of Ising Droplets*

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Recently Biskup *et al.* [*Europhys. Lett.* **60** (2002) 21] studied the behaviour of d-dimensional finite-volume liquid-vapour systems at a fixed excess δN of particles above the ambient gas density. They identify a dimensionless parameter $\Delta(\delta N)$ and a universal constant $\Delta_c(d)$ and show that for $\Delta < \Delta_c$ a droplet of the dense phase occurs while for $\Delta > \Delta_c$ the excess is absorbed in the background. The fraction λ_Δ of excess particles forming the droplet is given explicitly. Furthermore, they state, that the same is true for solid-gas systems.

To verify these results, we have simulated the spin-$1/2$ Ising model on a square lattice at constant magnetisation equivalent to a fixed particle excess in the lattice-gas picture. We measured the largest minority droplet, corresponding to the solid phase, at various system sizes ($L = 40, \ldots, 640$). Using analytic values for the spontaneous magnetisation m_0, the susceptibility χ and interfacial free energy τ_W for the infinite system, we were able to determine λ_Δ in very good agreement with the theoretical prediction.

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1. Introduction

One of the longstanding problems in statistical mechanics concerns the formation and dissolution of equilibrium droplets at a first-order phase transition. Interesting quantities in this context are the size and the free energy of a "critical droplet" that needs to be formed before the decay of the metastable state via homogeneous nucleation can start. In the analysis done so far it was implicitly assumed that the size of the droplet is of the order of the system, which is not the case when the droplet forms first [1, 2]. In this work, the region of system parameters that lead to the formation/dissolution of a droplet is examined by means of Monte Carlo simulations. We follow closely the theoretical ideas of Biskup *et al.* [3]; still, there are also different descriptions of the same physical phenomena [2, 4, 5, 6].

In the remaining sections, after a brief introduction to the classical droplet theory, first the new results of Biskup *et al.* [3] are summarized. Then our Monte Carlo measurements supporting the theoretical results are presented, and finally some preliminary conclusions are drawn.

2. Theory

In the following the term "droplet" will be used in the sense of particles (or spins) that are grouped together in a purely geometric way. One of the basic papers in this context is due to M. E. Fisher [7] where he discusses "... a gas of particles interacting with repulsive cores and short-range attractive forces ...". There, he mentions that "... the typical configuration at low densities and temperatures will consist of essentially isolated clusters of one, two, three or more particles" while "A sufficiently large cluster is just a small droplet of the liquid ..." and "Condensation in this picture corresponds to the growth of a macroscopic droplet of the liquid." It should be emphasised that the *geometric* definition of a cluster or droplet must not be confused with *stochastic* "Fortuin-Kasteleyn clusters" or anything the like where it is not possible to identify a cluster by just "looking" at a spin configuration.

All arguments in this section are based on the work of Fisher [7] and especially Biskup *et al.* [3]. Due to the fact that the Monte Carlo simulations were done using the Ising model, the theory is presented in terms of a lattice gas which mainly is a change of notation but does not alter the theory. This is also supported by Biskup *et al.*'s presentation where the general results are additionally given in the special context of the two-dimensional Ising model with Hamiltonian

$$\mathscr{H} = -J \sum_{\langle i,j \rangle} \sigma_i \sigma_j - h \sum_i \sigma_i \,, \tag{2.1}$$

where $\sigma_i = \pm 1$ and $\langle i, j \rangle$ denotes a nearest-neighbour pair. Having no external field ($h = 0$) the second term vanishes. When an up-spin ($\sigma_i = 1$) is treated as a particle and a down-spin ($\sigma_i = -1$) is treated as a vacancy the system can be interpreted as a lattice gas of atoms.

Considering such a system, classical droplet theory assumes that there are two contributions to the free energy. First, there can be local fluctuations and the probability to find a difference in the magnetisation (excess in the magnetisation compared to $M_0 = m_0 V$ with $V = L^2$) of $\delta M = M - M_0$ can be expressed in terms of a Gaussian distribution as

$$\exp\left[-\frac{(\delta M)^2}{2V\chi}\right] = \exp\left[-\frac{(2m_0 v_L)^2}{2V\chi}\right]. \tag{2.2}$$

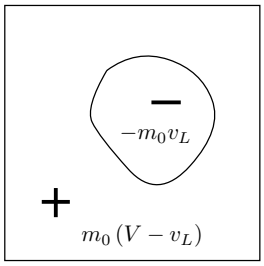

Figure 1: Ising system of size V with a minority droplet of volume v_L of negative spins surrounded by positive spins with an volume $(V - v_L)$. Here we have assumed for simplicity that the total excess in magnetisation is concentrated in the droplet, i.e. $v_d = v_L$.

Here, $m_0 = m_0(\beta)$ is the spontaneous magnetisation and $\chi = \chi(\beta)$ the susceptibility, both quantities at an inverse temperature β and in the thermodynamic limit. Assuming that the magnetisation in the background and inside a droplet is m_0, the total magnetisation can be written as $M = -m_0 v_L + m_0(V - v_L)$, where v_L is the volume of the droplet and V the volume of the system (see Fig. 1). Rearranging this expression for the difference in the magnetisation yields

$$\delta M = M - V m_0 = -2 v_L m_0 \tag{2.3}$$

which was used in Eq. (2.2).

The second contribution to the free energy stems from an interface of a droplet of volume v_L. The cost to form it is given in two dimensions by [8]

$$\exp\left[-\tau_{\mathrm{W}} \sqrt{v_L}\right] , \tag{2.4}$$

where $\tau_{\mathrm{W}} = \tau_{\mathrm{W}}(\beta)$ is the interfacial free energy per unit volume of an ideal shaped droplet which is also known as the free energy of a droplet of Wulff shape.

Comparing the exponents of Eq. (2.2) to (2.4) gives:

$$\Delta = \frac{(2 m_0 v_L)^2/(2V\chi)}{\tau_{\mathrm{W}} \sqrt{v_L}} = 2 \frac{m_0^2}{\chi \tau_{\mathrm{W}}} \frac{v_L^{3/2}}{V} . \tag{2.5}$$

With $\Delta \stackrel{!}{=} 1$ and Eq. (2.3), the difference in the magnetisation is

$$\delta M = \theta V^{2/3} \qquad \text{with} \qquad \theta = -\left(\frac{2\chi\tau_{\mathrm{W}}}{\sqrt{2 m_0}}\right)^{2/3} . \tag{2.6}$$

This means, when $\delta M \gg \theta V^{2/3}$ the droplet mechanism dominates, while the fluctuation mechanism dominates for $\delta M \ll \theta V^{2/3}$. Biskup *et al.* [3] studied the crossover region $\delta M \approx \theta V^{2/3}$. By isoperimetric reasoning, they showed that in this range no droplets of intermediate size exist. There is at most a single large droplet of size $v_{\mathrm{d}} < v_L$ with costs giving in Eq. (2.4) that absorbs δM_{d} of the excess of the magnetisation δM while the rest goes into the fluctuations of the background. This justifies the following Ansatz for the probability that the droplet contains the fraction $\lambda = \delta M_{\mathrm{d}}/\delta M$ of the excess in magnetisation:

$$\exp\left[-\tau_{\mathrm{W}}\sqrt{v_{\mathrm{d}}} - \frac{(\delta M - \delta M_{\mathrm{d}})^2}{2V\chi}\right] = \exp\left[-\tau_{\mathrm{W}}\sqrt{\frac{-\delta M}{2 m_0}}\Phi_\Delta(\lambda)\right] , \quad \Phi_\Delta(\lambda) = \left[\sqrt{\lambda} + \Delta(1-\lambda)^2\right] , \tag{2.7}$$

(a) Evaporated system where a large number of very small clusters exist (1 to 3 spins)

(b) Condensed system with a single large cluster that has absorbed nearly all small clusters

Figure 2: Snapshots of a two-dimensional Ising lattice; white squares correspond to up-spins (atoms), while black squares correspond to down-spins (vacancies).

where Δ is defined in Eq. (2.5). Since $\tau_W\sqrt{-\delta M/2m_0}$ and Δ are constants, the fraction of excess that is most probable is obtained by minimising $\Phi_\Delta(\lambda)$. In two dimensions, the minimisation leads to $\Delta_c = (1/2)(3/2)^{3/2} \approx 0.92$. For values $\Delta < \Delta_c$ the global minimum of $\Phi_\Delta(\lambda)$ is reached for $\lambda = 0$, while for $\Delta > \Delta_c$ it is located at a nontrivial value $\lambda_\Delta > 0$. At the transition point $\Delta = \Delta_c$, the value is $\lambda_c = 2/3$. The solid line in Fig. 4 below shows the graph of λ_Δ. Its interpretation is as follows: for $\Delta < \Delta_c$ all of the excess is absorbed in the background fluctuations, then, at the transition point $\Delta = \Delta_c$ a value of $2/3$ of the excess forms a droplet while the rest of the excess remains as background fluctuations. For values $\Delta > \Delta_c$ the droplet grows and absorbs most of the background fluctuations. For an illustration with actual simulation data, see Fig. 2.

3. Simulations

The main goal of our work was to check the theoretical results presented in the last section. In order to do so the fraction of the excess of magnetisation in the largest droplet was measured. To this end a Monte Carlo simulation was set up with a fixed excess of magnetisation. To keep the magnetisation during the simulation constant, a Metropolis update with Kawasaky dynamics was chosen. After every sweep a cluster decomposition was performed (using the Hoshen-Kopelman algorithm) and the volume of the largest cluster was measured. It is important to note that in the present context the volume of the cluster includes overturned spins within the cluster (which was implemented with a so-called "flood-fill" routine).

The simulations were performed at 38 different magnetisations chosen to have enough data points in the vicinity of the transition. The temperature was set to $T = 1.5$ and altogether five different square lattice sizes were taken into account ($L = 40, 80, \ldots, 640$). Every simulation took 20000 sweep for thermalisation and 200000 sweeps for measurements. To obtain the error bars, 10 independent simulations were run for each data point.

In order to get the correct scaling for the abscissa, $\Delta(M, M_0, \chi, \tau_W)$ has to be calculated. For the spontaneous magnetisation there exists Onsager's famous analytic solution and for the free energy of the Wulff droplet, Rottman and Wortis [9] were able to derive an analytic expression, while for the susceptibility only series expansions are known; see, e.g., Enting, Guttmann and Jensen [10] (but up to order 323!).

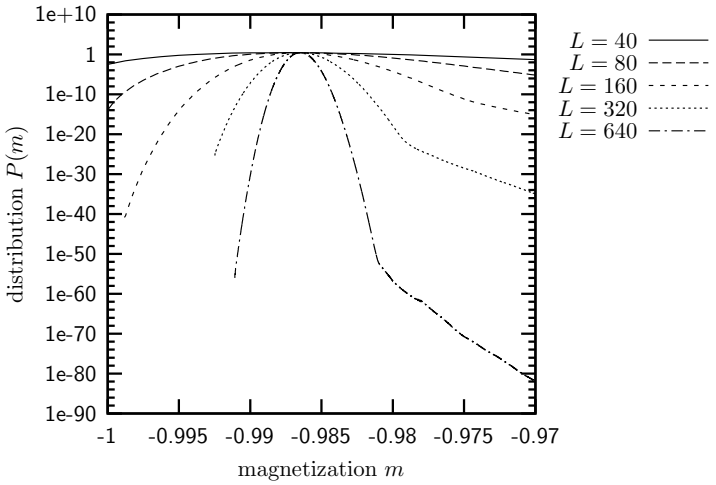

Figure 3: Distribution of the magnetisation for the two-dimensional Ising model for different system sizes L at the temperature $T = 1.5$. The cusp indicates the transition region. On the left side of the cusp (evaporated system) a Gaussian peak is clearly visible, while on the right side of the cusp (condensed system) the stretched exponential behaviour can be seen.

4. Results

Figure 3 shows the result of our "Multimagnetic" (Multicanonical for the magnetisation) simulations. The distribution of the magnetisation shows at larger lattices sizes a cusp which divides the evaporated and condensed region. Within the condensed region it has a Gaussian form according to Eq. (2.2) while in the condensed region a stretched exponential behaviour is visible, cf. Eq. (2.4). All data points in Fig. 4 are in the vicinity of this cusp. To get a feeling how the different configurations actual look like, Figs. 2(a) and 2(b) show an evaporated and condensed system, respectively. Both systems have the same magnetisation which was chosen to be the one at the transition point. They represent extremes since during one simulation run these are the configurations where the largest droplet is either minimal or maximal.

Figure 4 shows the results for the fractions λ_Δ for various lattice sizes. The solid line represents the analytical value (result of the minimisation of Eq. (2.7)). Clearly, for larger lattice sizes the result of the simulations approach the theoretical values nicely. The jump from $\lambda_\Delta \approx 0$ to $\lambda_\Delta \approx 2/3$ at $\Delta_c \approx 0.92$ confirms the theoretical prediction that at the evaporation/condensation transition only $2/3$ of the excess of the magnetisation goes into the droplet.

The increase of λ_Δ for $\Delta \to 0$ can be explained by the fact, that the minimal cluster size is 1 (and not an arbitrarily small fraction) but the excess that can be fixed is smaller than 1.

5. Conclusion

Our Monte Carlo data clearly confirm the theoretical results of Biskup *et al.* [3]. The observed finite-size scaling behaviour fits perfectly with their predictions. All simulations were performed in thermal equilibrium and the abundance of droplets of intermediate size could be confirmed. At the moment, additional simulations for different models are performed that should prove the universal aspects of the theory.

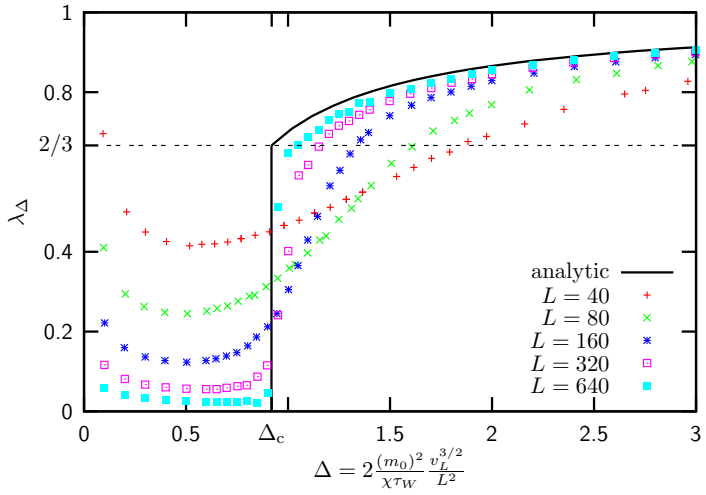

Figure 4: Two-dimensional Ising model with nearest-neighbour interaction on a square lattice ($L = 40, 80, \ldots, 640$) at the temperature $T = 1.5 \approx 0.7\,T_c$. The error bars are not plotted since their size is much smaller than that of the data symbols.

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