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Droplet dynamics in the two-dimensional kinetic Ising model

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Abstract. We discuss the microscopic dynamics of isolated clusters in the two-dimensional Ising model with Glauber spin-flip dynamics. Rate equations for the evolution of a droplet at low temperature are expressed in terms of the geometry of this droplet.

From these equations, the existence of a critical droplet size \mathcal{C}_c can be inferred. An exact lower bound for \mathcal{C}_c is obtained. Predictions on \mathcal{C}_c and the evolution of droplets are confirmed by computer experiments.

I. Introduction

It is well known that metastability is a kinetic phenomenon [1]. However, an exact microscopic dynamical theory of metastability does not exist yet. Dynamical aspects have been introduced in the study of metastability mainly in the two following ways (for a review of molecular theories of metastability, see [1]).

In the first approach (proposed by Lebowitz and Penrose [2]), one assumes that the static properties of a metastable state can be described by an appropriate distribution on phase space chosen on physical grounds. One then verifies a posteriori that this distribution has a long life time under a suitable dynamical process. This program is pursued in [2, 3, 4] and in [5] with a different choice for the metastable distribution. In the second approach, one tries to obtain an intrinsic characterization of metastability in a purely dynamical framework. Here, metastability has been defined as a 'flatness' property of the relaxation curve of relevant observables [6, 7, 8] (cf. also [19]), or linked to specific spectral properties of the evolution (approximate degeneracy of the ground state) [9, 10].

In any case, the physical mechanism underlying the formation and the decay of a metastable state is the nucleation process; clearly, a process which has to be understood at the molecular level. Usually nucleation is described by the classical theory of Becker and Döring [11, 12] and refinements of it. In this theory, one writes phenomenological rate equations for the dynamics of clusters; the rates are then related to the microscopy by invoking the detailed balancing condition and

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calculating the thermodynamical potential of a single droplet. Here, the existence of a critical droplet size does not result from a direct analysis of the molecular dynamics, but from thermodynamical considerations involving the bulk versus surface contributions to this potential.

The main point of this paper is to provide a detailed analysis of the low temperature evolution of a single critical or over critical cluster in a regime where such clusters already exist. In particular we focus attention on the microscopic dynamical mechanisms leading to the existence of the critical size. We will not be concerned with the general multicluster dynamics, nor do we attempt to give nucleation rates and full relaxation curves as in earlier work on the subject [7, 8].

We shall use the Glauber spin flip dynamics [13] in the two dimensional next neighbour Ising model as our basic process, and keep the magnetic interpretation. (In the lattice gas interpretation, the spin flip dynamics corresponds to the situation where there is an additional infinite reservoir emitting and absorbing particles at constant rates; for a study of the conservative Kawasaki or exchange dynamics see [14] and references quoted there; for a recent investigation paralleling ours, see [15]).

Consider an initial distribution of (-1) droplets in a sea of $(+1)$ spins corresponding to some prescribed value m of the average magnetization and let it evolve towards equilibrium in a small negative magnetic field $h = -|h|$.

At subcritical temperatures, the magnetization $m(t)$ relaxes according to the following qualitative patterns

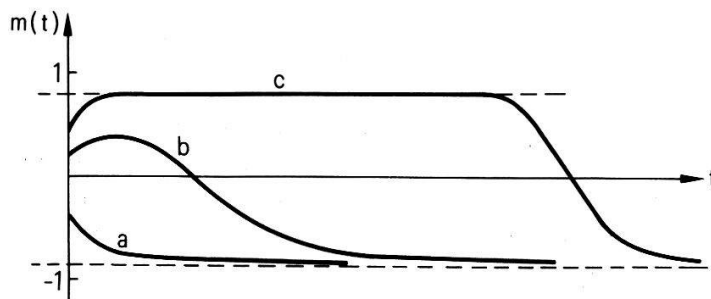


Figure 1
Qualitative magnetization relaxation curves

We note that there are two distinct possibilities: Either $m(t)$ tends immediately to equilibrium (curve *a*), or it is first attracted towards a metastable level (curves *b* and *c*). Curves of the type (*b*, *c*) cannot occur in one dimension since it can be shown that $dm(t)/dt \leq 0$ for $m(t) \geq 0$ [13].

The fact that a magnetization below its metastable value may first increase towards the metastable level, by the disappearance of subcritical droplets, is a phenomenon characteristic of metastability. The ultimate transition of $m(t)$ to its (negative) equilibrium value is dependent on the formation of supercritical droplets. This is illustrated by the following computer film corresponding to a very low temperature (Fig. 2).

By ferromagnetic interactions, an initial random configuration of (-1) spins in a background of $(+1)$ spins (*a*), coagulates rapidly into a pattern of clusters of various sizes and shapes (*b*). Subcritical droplets disappear gradually (*c*, *d*), whereas a supercritical droplet starts growing, retaining a typical near square shape,

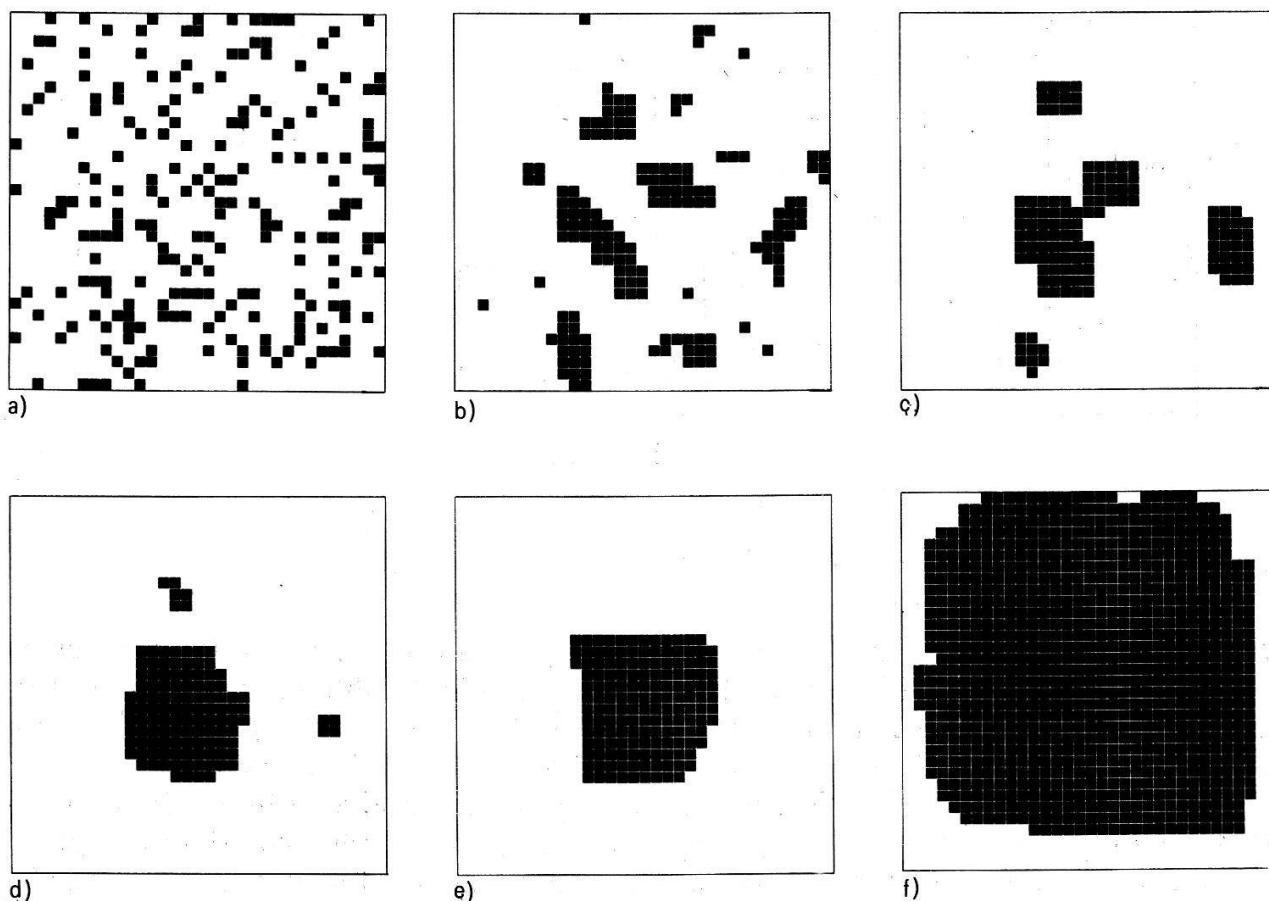


Figure 2

Computer film of nucleation. Torus 33×33 ; initial percentage of (-1) spins $\sim 20\%$; $\alpha = \text{th}(\beta h) = -0.3$; $\gamma = \text{th}(2\beta J) = 0.99$. Black square indicate (-1) spins.

(a) $t=0$ $m=0,614$, (b) $t=5$ $m=0,719$, (c) $t=20$ $m=0,778$
 (d) $t=50$ $m=0,787$, (e) $t=100$ $m=0,735$, (f) $t=500$ $m=-0,590$

(See the remark on computer simulations at the end of Section IV).

(e), until it fills the lattice and completes the transition towards the stable (-1) -phase (f).

We also note that the initial configuration evolves rapidly toward the characteristic configuration of a two phases equilibrium system, exhibiting typically a single almost square shaped domain of (-1) spins in a background of $(+1)$ spins [16].

In the example of Fig. 2, the appearance of a supercritical droplet right from the start, forces the state to relax towards the stationary state after a relatively short excursion towards the metastable level (curve b). If all droplets had happened to be subcritical, the state would have reached the metastable region and remained there for a very long time (curve a), due to the improbability of the formation of a critical droplet by thermal fluctuations.

Various qualitative features concerning growth and criticality of droplets can be obtained from simple arguments. In the Glauber dynamics, the probability of a flip at site x is proportional to $W(s) = \frac{1}{2}(1 - \text{th}[2\beta J(2-s)])$ where s is the number of spins neighbouring x with opposite sign (see section II). At low temperature $\beta = (kT)^{-1} \gg 1$ we may consider the approximation $W(0) = \frac{1}{2}(1 - \text{th} 4\beta J) \sim 0$ in

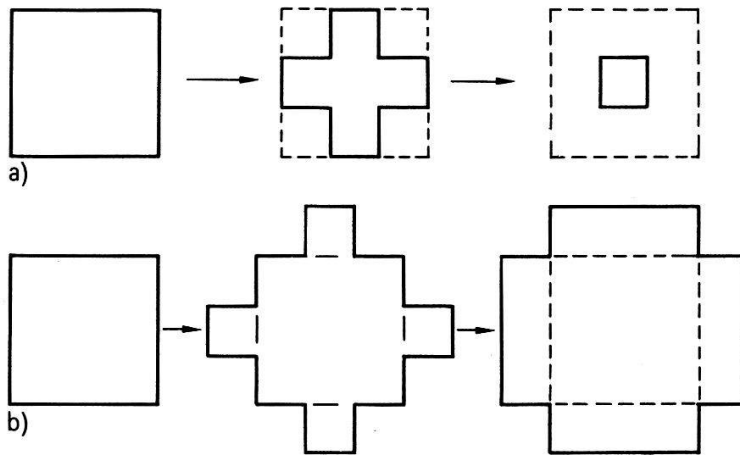


Figure 3
(a) Erosion of a droplet. (b) Growth of a droplet.

which no isolated new droplets can form. Thus, all flips occur at the boundaries of already existing ones, and original droplets remain identifiable in the course of the time (see Fig. 2). Since $W(1) = \frac{1}{2}(1 - \tanh 2\beta s) \ll W(s)$ for $s \geq 2$, $\beta \gg 1$, the $s = 1$ events at the boundary are much less probable than the $s \geq 2$ events.

The phenomenon of criticality can now be understood as follows: If we were to take only the rapid $s \geq 2$ flips into account, the droplet could never grow beyond the initial boundary of its circumscribed square, but it could eventually shrink away from it by the process of erosion schematically represented in Fig. 3a. The possibility for a droplet to exhibit long time growth therefore depends on the less frequent $s = 1$ processes by which protuberances exceeding the original square may be formed (Fig. 3b).

The argument then rests on the observation that, for increasing droplet size, the probability of creating new protuberances (thus new rows) increases whereas the probability that full rows disappear by $s \geq 2$ transitions decreases. Hence, there is a critical droplet size at which growth and shrinkage are balanced.

The paper is organized as follows. In section II we introduce the spin flip process and the notation. In section III we characterize an appropriate class of droplets by geometrical quantities and establish a formula for the infinitesimal change of the bulk of such droplets. Section IV contains a discussion of the critical size and qualitative properties of the evolution of a droplet. In section V we derive droplet dynamics from the general dynamics of spin correlations by interpreting the latter in geometrical terms.

II. The spin flip process

We consider the two dimensional periodic next neighbour ferromagnetic Ising model on the square lattice $\Lambda = \{x = (n_1, n_2); 0 \leq n_i \leq N-1\}$. We denote by $e_1 = (1, 0)$ and $e_2 = (0, 1)$ the two fundamental unit vectors and by ω a spin configuration in Λ :

$$\begin{aligned} \omega(x) &= \pm 1 & x \in \Lambda \\ \omega(x + Ne_1) &= \omega(x + Ne_2) = \omega(x) \end{aligned} \tag{1}$$

The hamiltonian of the spin system in an external field h is

$$\begin{aligned}
 H(\omega) &= -J \sum_{\substack{x, y \\ \text{next neighbour}}} \omega(x)\omega(y) - h \sum_x \omega(x), \quad J > 0 \\
 &= -\frac{J}{2} \sum_x \omega(x)\mu(x) - h \sum_x \omega(x)
 \end{aligned}
 \tag{2}$$

where $\mu(x) = \omega(x + e_1) + \omega(x + e_2) + \omega(x - e_1) + \omega(x - e_2)$ is the magnetization due to the four neighbours of x .

The evolution of a state ρ (i.e. a probability distribution $\rho(\omega)$ on spin configurations) is governed by the master equation for the spin flip process

$$\frac{d\rho_t(\omega)}{dt} = \sum_x (W(\omega | \omega^x)\rho_t(\omega^x) - W(\omega^x | \omega)\rho_t(\omega))
 \tag{3}$$

$W(\omega^x | \omega)$ is the transition rate per unit time of a spin flip in the configuration ω at lattice site x , where

$$\omega^x(y) = \begin{cases} \omega(y) & y \neq x \\ -\omega(y) & y = x \end{cases}
 \tag{4}$$

We assume that W obeys the detailed balancing condition

$$W(\omega^x | \omega) \exp[-\beta H(\omega)] = W(\omega | \omega^x) \exp[-\beta H(\omega^x)]
 \tag{5}$$

with $H(\omega^x) = H(\omega) + J\omega(x)\mu(x) + 2h\omega(x)$. The process (3) then admits as a stationary state the Gibbs distribution

$$\left[\sum_x \exp(-\beta H(\omega)) \right]^{-1} \exp[-\beta H(\omega)]$$

at temperature β^{-1} .

A transition rate compatible with (2) and (5) is

$$W(\omega^x | \omega) = \tau^{-1} [1 - \omega(x) \text{th}(\beta J \mu(\omega))] [1 - \omega(x) \alpha]
 \tag{6}$$

where $\alpha = \text{th} \beta h$ and τ is a time scale factor so adjusted that $0 \leq W(\omega^x | \omega) \leq 1$. (In the computer experiments $\tau = \frac{1}{2}(1 + |\alpha|)$.) Note that (5) only defines $W(\omega^x | \omega)$ up to an arbitrary symmetric function of ω^x and ω . The choice (6) coincides with that of [13] in the one dimensional case.

The first parenthesis in (6) is proportional to the probability of a flip due to its ferromagnetic binding, and the second parenthesis expresses the modification due to the external field h .

In terms of the variable $s(x) = 2 - [\omega(x)\mu(x)/2]$, i.e. the number of neighbouring spins to x with sign opposite to $\omega(x)$, the transition rate can be written $W(\omega^x | \omega) \equiv W(\omega, s)$ with

$$W(\omega, s) = \tau^{-1} [1 - \text{th}(2\beta J(2 - s))] [1 - \omega \alpha]
 \tag{7}$$

We classify the processes according to the possible values of $s = 0, 1, \dots, 4$. Since $J > 0$, the probability of s -processes increases with s .

At low temperature, the 0-process (i.e. the creation of a +1 or -1 island in a

sea of opposite sign) is of order

$$1 - \text{th } 4\beta J = 0(\exp(-8\beta J))$$

whereas the 1-process is of order

$$1 - \text{th } 2\beta J \equiv 1 - \gamma = 2 \exp(-4\beta J) + 0(\exp(-8\beta J))$$

As mentioned in the introduction, we shall consider the low temperature domain in which 0-processes are neglected and single droplet dynamics therefore makes sense.

III. Rate equations for droplets

Let us specify a class of droplets for which rate equations at low temperature can be derived in simple geometrical terms.

We first note that, up to rotations, the nearest neighbourhood of any site x is uniquely defined by $s(x)$, except in the case $s(x) = 2$ in which there are two possibilities characterized by

$$\omega(x + e_1)\omega(x - e_1) = \omega(x + e_2)\omega(x - e_2) = a(x); \quad a(x) = \pm 1$$

Consider a configuration ω containing a single droplet of (-1) spins with simple boundary (a simple boundary is a closed connected and non self-intersecting contour Γ in Λ). The droplet is *admissible* if, for all x ,

- (i) $s(x) \neq 4$
- (ii) if $s(x) = 2$, then $a(x) = -1$.

(i) excludes monomers and (ii) excludes droplets with "filaments". A droplet with filaments is shown in Fig. 4b.

For an admissible droplet we define the geometric quantities:

- n : volume = number of (-1) spins
- \mathcal{C} : perimeter = length of Γ
- v_{\pm} : number of positive (negative) corners = number of right angles in Γ enclosing a spin (± 1)
- p_{\pm} : number of (\pm) -protuberances = number of sites x with $s(x) = 3$, $\omega(x) = \pm 1$.

We denote by $v = v_+ + v_-$ the total number of corners and note that

$$v_- - v_+ = 4, \quad 4 \leq v \leq \mathcal{C} \quad (8)$$

As an example, an admissible droplet with $n = 13$, $\mathcal{C} = 20$, $v_- = 9$, $v_+ = 5$, $p_+ = 1$, $p_- = 3$ may look as in Fig. 4a. We now derive an expression for the initial volume change n' of an admissible droplet at low temperature.

Obviously n increases if a positive spin flips and n decreases if a negative spin flips. Hence

$$n' = \sum_{\substack{x \\ \omega(x)=1}} W(\omega, s) - \sum_{\substack{x \\ \omega(x)=-1}} W(\omega, s) \quad (9)$$

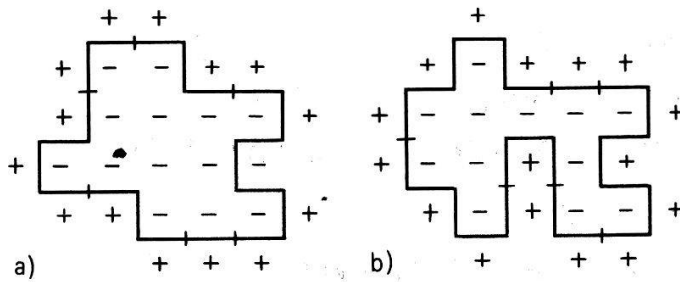


Figure 4
 (a) An admissible droplet. (b) A droplet with filaments.

Since monomers have been excluded, $s(x)$ will never equal 4. Since we neglect bulk processes, the sites at which $s(x) = 0$ do not contribute to the sum (9) either. Under these restrictions only six combinations of ω and s occur and the summand in (9) only takes six values. Hence, we can write:

$$\begin{aligned}
 n' = & W(1, 1)\#\{x \mid \omega(x) = 1, s(x) = 1\} + W(1, 2)\#\{x \mid \omega(x) = 1, s(x) = 2\} \\
 & + W(1, 3)\#\{x \mid \omega(x) = 1, s(x) = 3\} - W(-1, 1)\#\{x \mid \omega(x) = -1, s(x) = 1\} \\
 & - W(-1, 2)\#\{x \mid \omega(x) = -1, s(x) = 2\} \\
 & - W(-1, 3)\#\{x \mid \omega(x) = -1, s(x) = 3\}
 \end{aligned} \tag{10}$$

Next we express the various sums appearing in (10) in terms of the geometrical quantities n, \mathcal{C} , etc.

The definition of a protuberance implies that

$$p_+ = \#\{x \mid \omega(x) = 1, s(x) = 3\}, \quad p_- = \#\{x \mid \omega(x) = -1, s(x) = 3\} \tag{11}$$

We remark that a site with $\omega(x) = 1, s(x) = 3$ contributes two positive corners, whereas an admissible site with $\omega(x) = 1$ and $s(x) = 2$ contributes a single positive corner. Consequently,

$$\begin{aligned}
 v_+ = & 2\#\{x \mid \omega(x) = 1, s(x) = 3\} + \#\{x \mid \omega(x) = 1, s(x) = 2\} \\
 = & \#\{x \mid \omega(x) = 1, s(x) = 2\} + 2p_+ \\
 v_- = & \#\{x \mid \omega(x) = -1, s(x) = 2\} + 2p_-
 \end{aligned} \tag{12}$$

Finally, we derive an expression for \mathcal{C} .

Since a site characterized by $\omega(x) = 1$ and $s(x)$ gives a contribution $s(x)$ to the total length of the boundary,

$$\begin{aligned}
 \mathcal{C} = & \#\{x \mid \omega(x) = 1, s(x) = 1\} + 2\#\{x \mid \omega(x) = 1, s(x) = 2\} \\
 & + 3\#\{x \mid \omega(x) = 1, s(x) = 3\} \\
 = & \#\{x \mid \omega(x) = 1, s(x) = 1\} + 2v_+ - p_+
 \end{aligned} \tag{13}$$

and similarly,

$$\mathcal{C} = \#\{x \mid \omega(x) = -1, s(x) = 1\} + 2v_- - p_-$$

From (11), (12) and (13) now follows that:

$$n' = (\mathcal{C} - 2v_+ + p_+)W(1, 1) + (v_+ - 2p_+)W(1, 2) + p_+W(1, 3) \\ - p_-W(-1, 3) - (v_- - 2p_-)W(-1, 2) - (\mathcal{C} - 2v_- - p_-)W(-1, 1)$$

or with the explicit expression (7) for W

$$\tau n' = (2\gamma - 1)(1 - \alpha)v_+ - (2\gamma - 1)(1 + \alpha)v_- - 2\alpha(1 - \gamma)\mathcal{C}$$

with

$$\alpha = \text{th } \beta h, \quad \gamma = \text{th } 2\beta J$$

(Note that the p 's drop out altogether).

Using (8), the rate equation for the bulk of a single admissible negative droplet becomes

$$\tau n' = -(2\gamma - 1)(4 + \alpha v) - 2\alpha(1 - \gamma)\mathcal{C} \quad (14)$$

We also mention the following rate equation for the perimeter \mathcal{C} of an admissible droplet

$$\tau \mathcal{C}' = -4[p_+(1 - \gamma\alpha) + p_-(1 + \gamma\alpha) - (1 - \gamma)(\mathcal{C} - v + p_- - p_+ + 4\alpha)] \quad (15)$$

Although (15) could be obtained from direct geometrical consideration, we refer the reader to section V where equations (14) and (15) will be rederived in the general context of the dynamics of spin correlations.

IV. Time evolution and criticality

We shall now interpret the rate equations (14) and (15) for admissible droplets. In the low temperature domain considered here, all events occur at the boundary of the droplet. They are of two kinds: whereas the $s = 2$ processes leave the perimeter \mathcal{C} invariant and induce a change in the bulk n only, the $s = 1$ and $s = 3$ processes modify \mathcal{C} by creation and destruction of a protuberance. Moreover, a p_- -protuberance can occur either by erosion of the droplet due to $(-1) \rightarrow (+1)$ flips (2-processes), or by a spontaneous $(+1) \rightarrow (-1)$ flip at the boundary (1-process).

As discussed in the introduction, erosion and disappearance of a p_- -protuberance by $s = 2$ and $s = 3$ processes cause the shrinkage of the droplet, whereas creation of protuberances by $s = 1$ processes causes its growth. In order to decouple the phenomena of growth and shrinkage, it is convenient to consider first the following limiting situation

A. Zero temperature limit

We take the limit $\beta \rightarrow \infty$ ($\gamma \rightarrow 1$) and $h \rightarrow 0$ with $\alpha = \text{th } \beta h$ fixed, $|\alpha| \leq 1$. The probability of the s -processes is now strictly zero for $s < 2$ and $\tau^{-1}(1 \pm \alpha)$ for $s = 2$.

For negative magnetic field $h = -|h|$, $\alpha = -|\alpha|$, equations (14) and (15) imply, in this limit:

$$\tau n' = -(4 - |\alpha|v) \quad (16)$$

$$\tau \mathcal{C}' = -4(p_+(1 + |\alpha|) + p_-(1 - |\alpha|)) \quad (17)$$

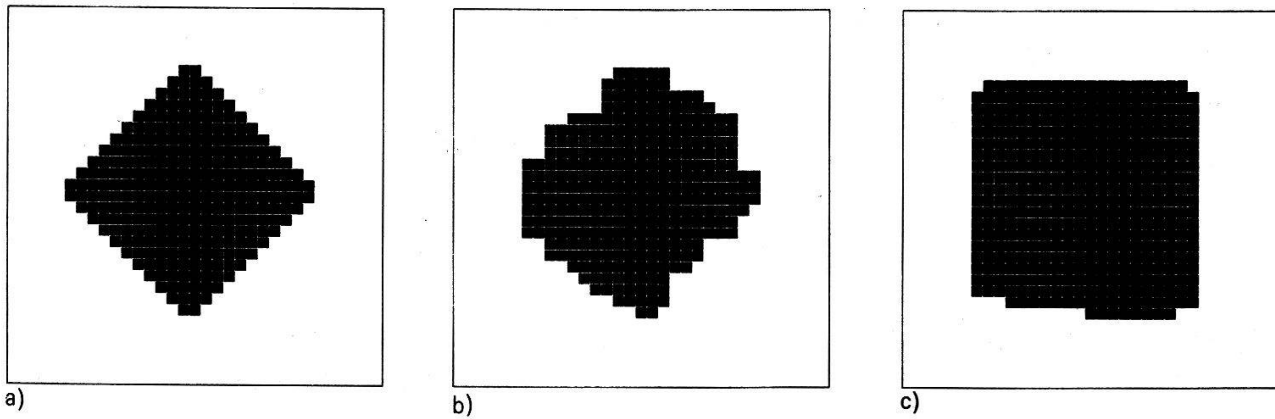


Figure 5
Approach of an initial diamond to the typical shape of a 'round-off' square

If the initial droplet has no protuberances ($p_+ = p_- = 0$), then $\mathcal{C}' = 0$. At $t = 0$, the shape of the droplet will therefore change, but not its perimeter.

For an initial square ($v = 4$)

$$n' = -4(1 - |\alpha|) < 0$$

is always negative, while for an initial diamond ($v = \mathcal{C} - 4$),

$$n' = -(4 - |\alpha|)(\mathcal{C} - 4)$$

is positive provided that $\mathcal{C} > 4/|\alpha| + 4$. This is readily understood by observing that the square can only lose its corners (thus decreasing n), whereas the bulk of the diamond can increase by 2-processes. It is then to be expected that, in the early stage of its evolution, any droplet will reach the typical shape of a square with round-off corners (Fig. 5). It turns out that the erosion of corners at zero temperature is closely related to a one dimensional random walk process. This will be discussed in the appendix.

B. Criticality

If the temperature is not strictly zero, one has to take into account the second term in the right hand side of (14). This term, which is positive and proportional to \mathcal{C} incorporates the $s = 1$ processes responsible for the growth of the droplet. A droplet will increase its bulk if $n' > 0$. A necessary condition for the growth of an admissible droplet is therefore

$$|\alpha|(1 - \gamma)\mathcal{C} - (\gamma - \frac{1}{2})(4 - |\alpha|v) > 0, \quad v \leq \mathcal{C} \tag{18}$$

Hence

$$\mathcal{C} \geq \mathcal{C}^* = \frac{8}{|\alpha|} (\gamma - \frac{1}{2}) = \frac{4}{\text{th } \beta |h|} (1 - 4 \exp(-4\beta J)) \tag{19}$$

where \mathcal{C}^* is an exact lower bound for the critical perimeter at low temperature.

The actual critical perimeter \mathcal{C}_c can be estimated as follows. The 2-processes by which a droplet approaches its typical shape are much faster than the 1-processes responsible for its ultimate growth. Instantaneous increase of the bulk

($n' > 0$) is therefore only significant for the long term growth of the droplet, if the latter has already reached its typical shape. In the typical shape, we can admit that $v = \lambda \mathcal{C}$ with $0 < \lambda < 1$ (see next sub-section, point b).

Inserting this in (14) and setting $n' = 0$, the estimate of the critical length \mathcal{C}_c becomes

$$\mathcal{C}_c = \frac{4}{|\alpha|} \frac{(\gamma - \frac{1}{2})}{1 - \lambda/2 - \gamma(1 - \lambda)} \geq \mathcal{C}^* \quad (20)$$

\mathcal{C}_c has the qualitative property $\mathcal{C}_c = 0(1/|h|)$ for $|h| \rightarrow 0$, as expected from the phenomenological droplet model. Moreover, \mathcal{C}_c increases with J , the ferromagnetic binding.

The approximate $1/|\alpha|$ -dependence of \mathcal{C}_c for various γ is shown in the following table which has been obtained by computer experiments (based on a relatively small statistics of 10 samples).

γ	$ \alpha $	\mathcal{C}_c (measured)
0.99	0.8	12
	0.6	12
0.97	0.4	20
	0.25	30, 5
	0.17	36
0.95	0.4	12
	0.2	28
	0.1	48
	0.05	88
0.9	0.4	12
	0.2	20
	0.1	36

C. Long time phenomenology

The property of a distribution to be initially concentrated on configurations of admissible droplets is in general not strictly conserved in the course of time. At non-zero temperature, filaments can for instance be created by sequential superposition of protuberances. The probability of such occurrences is, however, of the order $(1 - \gamma)^2 = 0(\exp(-8\beta J))$ and can be neglected within our low temperature approximation.

We can therefore admit that the rate equations (14) and (15) (which were derived only for an initially admissible droplet) remain valid at all times, provided that one interprets $n = n(t)$, $\mathcal{C} = \mathcal{C}(t)$, ... as the average bulk, length ... at time t .

The system (14), (15) is a part of an infinite hierarchy of equations of motion and can in general not be integrated. There is, however, one instance in which an exact solution exists, namely if $\gamma = 1$, $\alpha = 0$. In this case (16) implies $n'(t) = -4\tau^{-1}$ which is confirmed by the computer experiments.

At zero temperature and field, the bulk of a droplet therefore decreases linearly in time, irrespective of its shape. We observe in fact that the states concentrated on pure (\pm)-configurations are both stationary and attract respectively the states of (\mp)-droplets.

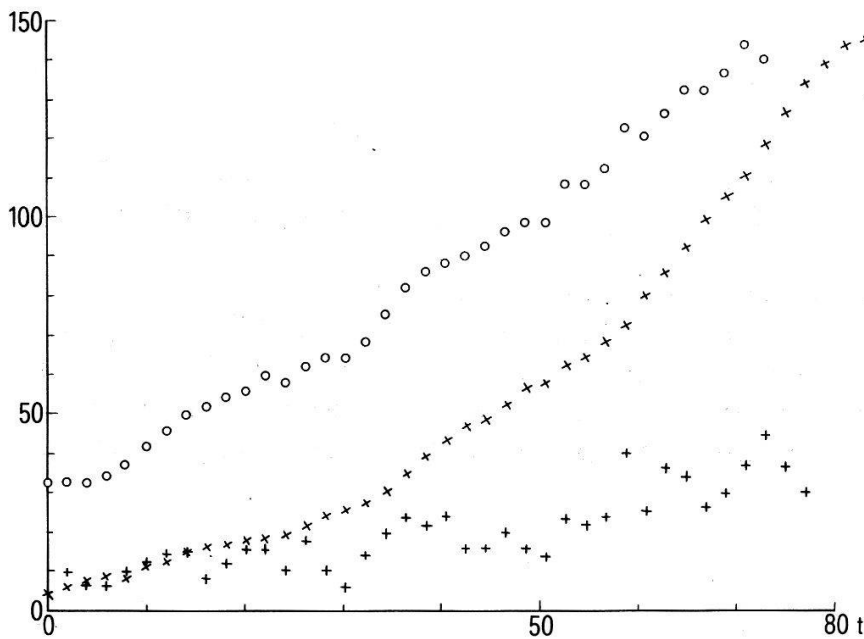


Figure 6

Plot of $\mathcal{C}(t)$, $v(t)$, $\frac{1}{10}n(t)$.

Notation: $\circ = \mathcal{C}$, $+$ = v , $\times = n/10$

Parameters: $\alpha = -0.6$, $\gamma = 0.99$, initial shape: diamond.

For finite temperature a discussion of the system (14), (15) must rely on a combination of computer 'insight' and heuristics.

Our computer films of highly supercritical droplets at various low temperatures and fields exhibit two characteristic features: If one disregards a short initial period (during which an arbitrary droplet reaches its typical shape) and a final period (starting at the moment where the droplets rap completely around the torus)

- (a) $\mathcal{C}(t)$ increases approximately linearly
- (b) the ratio $\lambda = v/\mathcal{C}$ remains approximately constant in time (Fig. 6)

(We note that, in the initial period of adjustment, \mathcal{C} remains constant, v decreases and n increases, as expected for a diamond. The fluctuations observed for the \mathcal{C} and v are essentially caused by the presence of isolated protuberances).

Property (a) may be explained by the following argument: for highly supercritical droplets, the effect of erosion by 2-processes may be neglected. The long time growth of \mathcal{C} (and n) is then due solely to the adjunction of entire new columns and rows to the bulk of the droplet, each leading to an increase $\Delta\mathcal{C} = 2$.

If the average time needed to the formation of a full row by a 2-process is proportional to the initial density of protuberances, then adjunction of new rows is to occur linearly in time.

The linearity of $\mathcal{C}(t)$ is compatible with the assumption that

$$n(t) = \kappa\mathcal{C}^2(t), \quad v(t) = \lambda\mathcal{C}(t)$$

with $\kappa > 0$ and $0 < \lambda < 1$ independent of time. Equation (16) then implies

$$\frac{d\mathcal{C}(t)}{dt} = a + b\mathcal{C}^{-1}(t)$$

with

$$a = \frac{|\alpha| \lambda (\gamma - \frac{1}{2}) + (1 - \gamma)}{2\kappa} > 0$$

and thus $\mathcal{C}(t) \sim at$ for t large. (Note that the second equation (17) is only compatible with this model if p_- and p_+ also increase linearly with time at well defined rates).

The computer simulation: the computer algorithm proceeds as follows. A random spin is chosen out of the $N \times N$ lattice, and this spin flips with probability $[1 - \text{th}(2\beta J(2-s))][1 - \omega\alpha]^{1/2}(1 + |\alpha|)$ for $s \neq 0$. To avoid the creation of inadmissible droplets, the probability of 0-processes is taken strictly equal to zero by setting $\text{th} 4\beta J = 0$. The time unit corresponds to $N \times N$ steps of the algorithm.

V. General formalism

A. Dynamics of spin correlations

The formal solution of the equation of motion (3) can be expressed in terms of a linear semigroup $\exp(Gt)$ with generator G defined by the right hand side of (3), i.e.

$$\frac{d\rho_t(\omega)}{dt} = (G\rho_t)(\omega), \quad \rho_t(\omega) = (\exp(Gt)\rho)(\omega) \quad (21)$$

If we define the dual action G^* of G on functions of spin configurations by

$$\sum_{\omega} (G\rho)(\omega) f(\omega) = \sum_{\omega} \rho(\omega) (G^*f)(\omega)$$

then G^* is

$$(G^*f)(\omega) = \sum_x W(\omega^x | \omega) (f(\omega^x) - f(\omega)) \quad (22)$$

The average $\langle f \rangle_t = \sum_{\omega} \rho_t(\omega) f(\omega)$ of f at time t , now satisfies the differential equation

$$\frac{d\langle f \rangle_t}{dt} = \sum_{\omega} (G\rho_t)(\omega) f(\omega) = \sum_{\omega} \rho_t(\omega) (G^*f)(\omega) = \langle G^*f \rangle_t \quad (23)$$

We remark that the equations of motion are invariant under lattice translations τ_x and rotations R by ninety degrees. Indeed, denoting with the same symbol τ_x an x -translation of a configuration ω or of a function $f(\omega)$,

$$(\tau_x \omega)(y) = \omega(x + y), \quad (\tau_x f)(\omega) = f(\tau_x \omega)$$

(4) and (6) imply

$$W(\tau_z \omega^x | \tau_z \omega) = W(\omega^x | \omega), \quad \tau_z \omega^x = (\tau_z \omega)^{x-z}$$

and

$$(\tau_z G^*f)(\omega) = (G^* \tau_z f)(\omega), \quad z \in \Lambda \quad (24)$$

Similar relations hold for a $\pi/2$ -rotation R .

We consider the translation and rotation invariant correlation functions which are averages in a state ρ of polynomials of spin functions of the form

$$\begin{aligned} \langle y_1 \cdots y_n \rangle(\omega) &= \left(\frac{1}{N^2} \sum_x \tau_x \right) \left(\frac{1}{4} \sum_{k=0}^3 R^k \right) \omega(y_1) \cdots \omega(y_n) \\ &= \frac{1}{4N^2} \sum_x \sum_{k=0}^3 \omega(x + R^k y_1) \cdots \omega(x + R^k y_n) \end{aligned} \quad (25)$$

where y_1, \dots, y_n is a given set of distinct points in Λ for $n = 1, 2, \dots$

The following graphical notation is useful. We denote the one point function (the magnetization density) by

$$\langle y_1 = 0 \rangle(\omega) = \langle \bullet \rangle(\omega),$$

the two point functions by

$$\langle y_1 = 0, y_2 = e_1 \rangle(\omega) = \langle \bullet \rightarrow \rangle(\omega)$$

$$\langle y_1 = e_1, y_2 = e_2 \rangle(\omega) = \langle \uparrow _ \rangle(\omega)$$

$$\langle y_1 = e_1, y_2 = -e_1 \rangle(\omega) = \langle \bullet + \bullet \rangle(\omega),$$

the three point functions by

$$\langle y_1 = 0, y_2 = e_1, y_3 = e_2 \rangle(\omega) = \langle \uparrow _ \rangle(\omega)$$

$$\langle y_1 = 0, y_2 = e_1, y_3 = -e_1 \rangle(\omega) = \langle \bullet \cdots \bullet \rangle(\omega)$$

$$\langle y_1 = e_1, y_2 = e_2, y_3 = -e_1 \rangle(\omega) = \langle \bullet \uparrow \bullet \rangle(\omega) \text{ etc. } \dots$$

For the same correlation functions of a state ρ_t at time t , we write in the same way

$$\langle \uparrow _ \rangle_t = \sum_{\omega} \rho_t(\omega) \langle \uparrow _ \rangle(\omega) \text{ etc.}$$

(Notice that the orientation of a symbol has no importance since all quantities are rotation invariant.)

The master equation (3) induces a set of coupled differential equations for the correlations. This is due to the fact that G^* leaves the class of functions of the form (25) invariant. This can be seen as follows:

Fact 1. $W(\omega^x | \omega)$ is a polynomial of third degree in the spin functions, as implied from the explicit formula [17]

$$\text{th}(\beta J \mu(x)) = \frac{1}{4}(C_+ \mu(x) - C_- \nu(x)) \quad (26)$$

with

$$\begin{aligned} \nu(x) &= \sum_{k=0}^3 \omega(x + R^k e_1) \omega(x + R^k e_2) \omega(x - R^k e_1) \\ C_{\pm} &= \text{th} 2\beta J \pm \frac{1}{2} \text{th} 4\beta J \end{aligned} \quad (27)$$

Fact 2. $G^* f$ is of order $0(1)$. Indeed the sum in the right hand side of (22) runs over N^2 terms, but $f(\omega^x) - f(\omega)$ is $0(1/N^2)$ It follows from (4), with

$y_1 \neq y_2 \neq \dots \neq y_n$, that

$$\prod_i^n \omega^x(y_i) = \prod_i^n \omega(y_i) - 2\omega(x) \sum_j^n \delta_{x,y_j} \prod_{i \neq j}^n \omega(y_i)$$

and

$$\frac{1}{N^2} \sum_y \left(\prod_i^n \omega^x(y + y_i) - \prod_i^n \omega(y + y_i) \right) = -\frac{2\omega(x)}{N^2} \sum_j^n \prod_{i \neq j}^n \omega(y_i - y_j + x) = 0 \left(\frac{1}{N^2} \right) \quad (28)$$

Fact 3. By the covariance (24), G^*f is translation and rotation invariant whenever f has the same property.

Using (6), (26) and (28) in (22) and (23) one finds, by straight forward algebra, the equations for the lowest order correlations

$$\tau \frac{d\langle \cdot \rangle}{dt} = -2(1 - C_+) \langle \cdot \rangle_t - 2C_- \langle \cdot \downarrow \cdot \rangle_t + 2\alpha(1 - C_+ \langle \cdot \rightarrow \cdot \rangle_t + C_- \langle \cdot \uparrow \cdot \rangle_t) \quad (29)$$

$$\begin{aligned} \tau \frac{d\langle \cdot \rightarrow \cdot \rangle_t}{dt} = & C_+ - 4\langle \cdot \rightarrow \cdot \rangle_t + (C_+ - C_-)(2\langle \cdot \downarrow \cdot \rangle_t + \langle \cdot \uparrow \cdot \rangle_t) - C_- \langle \cdot \uparrow \cdot \rangle_t \\ & + \alpha \{ (4 - C_+) \langle \cdot \rangle_t - (C_+ - C_-)(2\langle \cdot \downarrow \cdot \rangle_t + \langle \cdot \rightarrow \cdot \rangle_t) + C_- \langle \cdot \uparrow \cdot \rangle_t \} \end{aligned} \quad (30)$$

The full hierarchy would give a complete description of the time evolution of a translation and rotation invariant state equivalent to that obtained from the master equation (3).

B. Low temperature approximation

Since our specific goal is to understand the evolution of individual droplets, we consider only the low temperature domain in which the concept of droplet makes sense.

It is appropriate to introduce new functions $[y_1 \dots y_n](\omega)$ on spin configurations defined by

$$\langle y_1 \dots y_n \rangle(\omega) = 1 - \frac{2}{N^2} [y_1 \dots y_n](\omega) \quad (31)$$

with corresponding averages $[y_1 \dots y_n]_t$.

$$[\cdot](\omega) = \sum_x \frac{1 - \omega(x)}{2}$$

is simply the number of (-1) spins in the configuration ω . More generally,

$$[y_1 \dots y_n](\omega) = \frac{1}{4} \sum_{k=0}^3 \sum_x \left(\frac{1 - \omega(x + R^k y_1) \dots \omega(x + R^k y_n)}{2} \right) \quad (32)$$

is the number of lattice sites at which $\omega(x + y_1) \dots \omega(x + y_n) = -1$, averaged over rotations.

Substitution of (31) in (29) and (30) yields

$$\begin{aligned} \tau \frac{d[\cdot]_t}{dt} = & N^2(1-\alpha)(1-C_+ + C_-) - 2(1-C_+)[\cdot]_t - 2C_-[\cdot\uparrow\cdot]_t \\ & + 2\alpha(C_+[\cdot\leftrightarrow]_t - C_-[\cdot\uparrow\cdot]_t) \end{aligned} \quad (33)$$

$$\begin{aligned} \tau \frac{d[\cdot\leftrightarrow]_t}{dt} = & 2N^2(1-\alpha)(1-C_+ + C_-) - 4[\cdot\leftrightarrow]_t + (C_+ - C_-)(2[\cdot\downarrow\cdot]_t + [\cdot\leftrightarrow]_t) \\ & - C_-[\cdot\uparrow\cdot]_t + \alpha\{(4-C_+)[\cdot]_t - (C_+ - C_-)(2[\cdot\downarrow\cdot]_t + [\cdot\leftrightarrow]_t) + C_-[\cdot\uparrow\cdot]_t\} \end{aligned} \quad (34)$$

As in section II, we consider the low temperature domain in which the 0-processes can be neglected versus the 1-processes. This amounts to retain only the 0th and 1st order terms in the low temperature expansion of C_{\pm} , i.e.

$$\begin{aligned} C_+ &= \gamma + \frac{1}{2} = \frac{3}{2} - 2 \exp(-4\beta J) + 0(\exp(-8\beta J)) \\ C_- &= \gamma - \frac{1}{2} = \frac{1}{2} - 2 \exp(-4\beta J) + 0(\exp(-8\beta J)) \end{aligned} \quad (35)$$

In particular, we neglect the first terms in (33), (34) which are proportional to $N^2(1 - \text{th } 4\beta J)$, representing the contribution of the spin flips in the bulk to the rate of change of $[\cdot]_t$ and $[\cdot\leftrightarrow]_t$. With (35) we find

$$\begin{aligned} \tau \frac{d[\cdot]_t}{dt} = & -([\cdot\uparrow\cdot]_t - [\cdot]_t) + \alpha(3[\cdot\leftrightarrow]_t - [\cdot\uparrow\cdot]_t) \\ & + 2(1-\gamma)\{[\cdot\uparrow\cdot]_t - [\cdot]_t + \alpha([\cdot\leftrightarrow]_t - [\cdot\uparrow\cdot]_t)\} \end{aligned} \quad (36)$$

$$\begin{aligned} \tau \frac{d[\cdot\leftrightarrow]_t}{dt} = & -(4[\cdot\leftrightarrow]_t - 2[\cdot\downarrow\cdot]_t - [\cdot\leftrightarrow]_t + \frac{1}{2}[\cdot\uparrow\cdot]_t) \\ & + \alpha(\frac{5}{2}[\cdot]_t - 2[\cdot\downarrow\cdot]_t - [\cdot\leftrightarrow]_t + \frac{1}{2}[\cdot\uparrow\cdot]_t) \\ & + (1-\gamma)\{[\cdot\uparrow\cdot]_t + \alpha([\cdot]_t - [\cdot\uparrow\cdot]_t)\} \end{aligned} \quad (37)$$

Equations (36) and (37) are general and hold rigorously for arbitrary states in the low temperature limit.

C. The geometry of droplets

In order to derive the dynamics of droplets characterized by the rate equations (14) and (15), we must express the correlation functions in (36) and (37) in terms of the geometry of single admissible droplets. This connection is established by the following

Lemma. For admissible $(-)$ droplets the following relations hold between n , \mathcal{C} , v , p_+ , p_- and the correlation functions:

$$\begin{aligned} n &= [\cdot] \\ \mathcal{C} &= 2[\cdot\leftrightarrow] \\ v &= 3[\cdot\leftrightarrow] - [\cdot\uparrow\cdot] \\ p_+ + p_- &= 2[\cdot\leftrightarrow] - [\cdot\downarrow\cdot] - \frac{1}{2}[\cdot\leftrightarrow] + \frac{1}{4}[\cdot\uparrow\cdot] \\ p_+ - p_- &= \frac{5}{4}[\cdot] - [\cdot\downarrow\cdot] - \frac{1}{2}[\cdot\leftrightarrow] + \frac{1}{4}[\cdot\uparrow\cdot] \end{aligned}$$

Moreover,

$$[\cdot\uparrow\cdot] - [\cdot] = 4$$

$$p_+ + p_- = \frac{1}{2}[\cdot\uparrow\cdot] - 2[\cdot\leftrightarrow] + v$$

$$p_+ - p_- = \frac{1}{2}([\cdot\uparrow\cdot] - [\cdot]) + 4$$

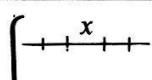
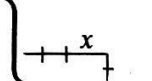
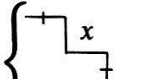
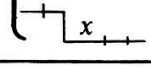
Insertion of these relations into (36), (37) yields (14), (15).

Proof of the lemma. We begin with the following observations.

- (1) The results $[\cdot](\omega) = n$ and $[\cdot\leftrightarrow](\omega) = \mathcal{C}/2$ are immediate.
- (2) In order to calculate any of the other functions, say $[\cdot\uparrow\cdot](\omega)$, we count the number of sites y where the figure $\cdot\uparrow\cdot = \omega(y)\omega(y+e_1)\omega(y+e_2)\omega(y+e_2)\omega(y-e_2)$ centered at y equals -1 , and average over rotations.
- (3) The value of the figure at y depends only on the next neighbours of y .
- (4) An even figure equals 1 except at sites adjacent to the boundary, an odd figure equals 1 except at sites adjacent to the boundary and at the other sites inside the droplet where it equals necessarily -1 .
- (5) In view of (4) and the fact that the combinations of odd correlations in the lemma are zero when all figures are -1 , it is sufficient to evaluate the figures only at the sites adjacent to the boundary.
- (6) The admissibility condition insures that the value of a figure at a site adjacent to the boundary is uniquely determined by the neighbouring configuration of the boundary itself.

The proof follows now by induction. One lists all the relevant modifications of the boundary due to individual s -processes $s = 1, 2, 3$, and one calculates for each of them the variation Δ of the expressions in the lemma and of the geometrical quantities. The validity of the formulae is readily checked on the simplest admissible droplets, for instance the 2×2 square.

The basic changes are of four types given graphically below. The segment represents a piece of boundary and x is the place where the flip $\omega(x) \rightarrow -\omega(x)$ occurs. Here x is external to the droplet (i.e. $\omega(x) = 1$). The cases where x is adjacent to the boundary but inside the droplet are obtained by spin reversal symmetry.

Process	Δn	$\Delta \mathcal{C}$	Δv
$s = 1$  	1	2	4
	1	2	2
$s = 2$  	1	0	-2
	1	0	0

Each of the above cases gives rise to four different situations depending on the creation or destruction of protuberances; for the first one we have e.g.

	Δp_+	Δp_-
	0	1
	1	1
	1	1
	2	1

One obtains in this way sixteen different cases.

In order to compute for these the variations Δ of the expressions in the lemma, it is according to (5) and (6) sufficient to consider the values of the figures adjacent to the boundary before and after the flip at x . We give a few examples.

Figures	Local modification of the boundary	Differences
		$3 - 3 = 0$
		$\frac{10}{4} - \frac{6}{4} = 1$
		$\frac{14}{4} - \frac{18}{4} = -1$
		$1 - 3 = -2$

Here a number equals the average over rotations of the numbers of -1 found in evaluating the figure at y , for instance

$$[\cdot\cdot\cdot] = \frac{1}{4} \sum_k (1 - \omega(y)\omega(y + R^k e_1)\omega(y + R^k e_2)\omega(y - R^k e_1))$$

Hence

$$\Delta(3[\cdot\cdot\cdot])(\omega) - [\cdot\cdot\cdot](\omega) = 3 + 1 = 4 = \Delta v$$

$$\Delta([\cdot\cdot\cdot])(\omega) - [\cdot](\omega) = -2 = 2(\Delta p_+ - \Delta p_-)$$

The reader can check the remaining cases.

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Appendix

There is a one to one correspondance between the process of erosion of corners of a square droplet in the zero temperature limit and the following one dimensional asymmetric random walk with exclusion.

Consider the space of infinite sequences of 1 and 0 (occupied and unoccupied particle sites) with configurations $\{\nu(i) = 0, 1, i \in \mathbb{Z}\}$ and denote the configuration obtained by the exchange of occupation of the i and $i+1$ sites by

$$\nu^{i,i+1}(k) = \begin{cases} \nu(k), & k \neq i, j \\ \nu(i+1), & k = i \\ \nu(i), & k = i+1 \end{cases}$$

A Markov process is defined by giving a transition rate $w(\nu^{i,i+1} | \nu)$ between the ν and the $\nu^{i,i+1}$ configuration

$$w(\nu^{i,i+1} | \nu) = \begin{cases} \frac{1}{2}(1+\alpha) & \text{if } \nu(i) = 1, \nu(i+1) = 0 \\ \frac{1}{2}(1-\alpha) & \text{if } \nu(i) = 0, \nu(i+1) = 1 \\ 0 & \text{otherwise} \end{cases}$$

A configuration of a left bottom of an eroded corner is a non-increasing step function $m(n) = \lim_{\varepsilon \rightarrow 0} m(n - \varepsilon)$ on the positive integers.

The correspondance between the erosion process and the random walk is established by associating the initial particle configuration $\nu_0 = \{\nu(i) = 1, i \leq 0; \nu(i) = 0; i > 0\}$ to the non-eroded corner and identifying $m(n)$ with the displacement to the right of the n^{th} particle from its initial position in ν_0 .

The 2-process with (1) \rightarrow (-1) flip ((-1) \rightarrow (1) flip) has probability $\frac{1}{2}(1+\alpha)$ ($\frac{1}{2}(1-\alpha)$) corresponding to the move of a particle to the left (resp. to the right).

The absence of 1-processes corresponds to exclusion. The full asymmetric case with $\alpha = 1$ is treated in [18]: it describes the shrinkage of a (-1) droplet in a positive external field. The case of interest for the study of the typical shape of a (-1) droplet in a negative fields corresponds to the partially asymmetric situation $-1 \leq \alpha < 0$.

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