Uphill diffusion in the 2d, n.n. Ising model

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Introduction.

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In the literature "uphill diffusion" has several meanings.

Here we refer to one in particular which occurs when **a diffusive fluid is in a stationary state with a boundary driven current and undergoes a phase transition.**

We then say that:

there is uphill diffusion if the current brings mass from the region with the low density phase to the one with the larger density.

Introduction.

To verify the presence of uphill diffusion I consider the nearest-neighbor Ising model with ferromagnetic interaction in a square $[0, L]^2 \cap \mathbb{Z}^2$ with

- Kawasaki dynamics at $\beta > \beta_c$.
- Periodic boundary conditions in the vertical direction.
- Independent spin flips on the first and last columns that force a magnetization $m_+ > 0$ on the right column and $m_- = -m_+$ on the left column (which simulate two reservoirs).

Precise definitions are given later.

Introduction.

We have fixed $\beta = 1$ ($> \beta_c \approx 0,440686$ by Onsager)

and run computer simulations with $L = 50, 100$

for various values of m_+ in $(1 - \delta, 1]$, $\delta > 0$ small, and $m_- = -m_+$

Fact.

As m_+ decreases from $m_+ = 1$ the current is first negative and past a critical value it becomes positive.

Introduction.

Downhill and uphill diffusions.

We conclude from the simulations that:

- If m_+ (the magnetization of the right reservoir $\mathcal{R}_+$) **is supercritical** and m_- (the magnetization of the left reservoir $\mathcal{R}_-$) $= -m_+$ then the magnetization flows from the plus to the minus phase (from $\mathcal{R}_+$ to $\mathcal{R}_-$) so that the **current is negative** (in agreement with the Fick's law) and we see **downhill diffusion.**

- Instead if m_+ **is subcritical** the magnetization flows from the minus to the plus phases (from $\mathcal{R}_-$ to $\mathcal{R}_+$) thus the **current is positive** and **it goes uphill**.

Introduction.

The value of the critical magnetization (at $\beta = 1$) obtained from the computer simulations is $m_{\text{crit}} = 0.99926$ which is very close to the equilibrium spontaneous magnetization $m_\beta = 0.9992757$ (by Onsager) of the Ising model.

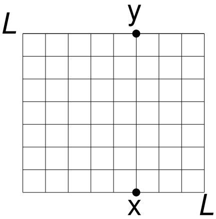
I will present in this talk heuristic arguments which indicate that *in the limit $L \to \infty$ there is no uphill diffusion which is instead present at finite L*. Therefore the claim is:

There is uphill diffusion but it is a finite size effect.

The model : nearest-neighbor Ising spin system

$\sigma(x) = \pm 1$ in the square $x \in [0, L]^2 \cap \mathbb{Z}^2$. $\beta = 1 > \beta_c \approx 0, 44$

$\mathcal{E}$ = set of n.n. bonds $< x, y >$ including $x =< i, 1 >$ and $y =< i, L >$ (vertical periodicity).



The ferromagnetic hamiltonian we consider is

$$H(\sigma) = H_0(\sigma) + H_b(\sigma)$$

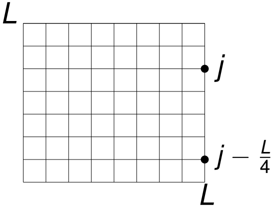
where

$$H_0(\sigma) = 2 \sum_{<x,y>\in\mathcal{E}} \mathbf{1}_{\sigma(x)\neq\sigma(y)}$$

$$H_b(\sigma) = 2\sum_{j=1}^{L} \mathbf{1}_{\sigma(1,j)\neq\sigma(1,j-\frac{L}{4})} + 2\sum_{j=1}^{L} \mathbf{1}_{\sigma(L,j)\neq\sigma(L,j-\frac{L}{4})}$$

where $j - \frac{L}{4}$ stands for j minus the integer part of $\frac{L}{4}$ modulo L.

It will become clear later why we have added the boundary interaction $H_b(\sigma)$ which describes a delocalized spin-spin interaction on the first and last columns:



Kawasaki dynamics + spin flips.

This is the continuous time Markov process with the following rates. The spins in a bond $< x, y > \in \mathcal{E}$ exchange values at rate

$$c(x, y; \sigma) = \mathbf{1}_{\sigma(x) \neq \sigma(y)} \begin{cases} 1 & \text{if } \Delta H(\sigma) = H(\sigma^{x,y}) - H(\sigma) \leq 0 \\ e^{-\beta \Delta H(\sigma)} & \text{otherwise} \end{cases}$$

The spins $\sigma(x)$ at the boundaries flip at rate $c_\pm$:

$$c_-(x; \sigma) = \frac{1 - \sigma(x) m_-}{2} \quad \text{if } x = (1, j)$$

$$c_+(x; \sigma) = \frac{1 - \sigma(x) m_+}{2} \quad \text{if } x = (L, j)$$

We have done computer simulations using both the *classical Metropolis Monte Carlo method* and the *kinetic Monte Carlo method*.

While the first algorithm is better suited to describe stationary states the second, which mimics a continuous time dynamics, is useful in the description of transient regimes.
The two dynamics have the same invariant measure.

P. Kratzer, *Monte Carlo and Kinetic Monte Carlo Methods – A Tutorial, Multiscale Simulation Methods in Molecular Sciences*, Forschungszentrum Jülich, NIC Series, Vol. 42, pp. 51-76, 2009

Hydrodynamic limit.

Let $\beta < \beta_c$.

In the limit $L \to \infty$ the empirical magnetization in the stationary state converges to the macroscopic magnetization profile $m(r)$, $r \in [0,1]^2$.

By the vertical symmetry $m(r)$ is a function $m(r_1)$ of the x-coordinate r_1 and $m(r_1)$, $r_1 \in (0,1)$, is the unique solution of:

$$j = -D(m)\frac{dm}{dr_1} = \text{const.}, \qquad m(0) = m_-, \quad m(1) = m_+$$

with $D(m) > 0$ given by the Green Kubo formula.

$\beta < \beta_c$: Fick law.

$m(r_1)$, $r_1 \in (0, 1)$ is the unique solution of

$$j = -D(m)\frac{dm}{dr_1} = \text{const.}, \qquad m(0) = m_-, \quad m(1) = m_+$$

The statement should follow from:

- Varadhan, Yau: *Diffusive limit of lattice gas with mixing conditions* (1997)
- Spohn, Yau: *Bulk Diffusivity of Lattice Gases Close to Criticality* (1995)
- Eyink, Lebowitz, Spohn: *Hydrodynamics of stationary non-equilibrium states for some stochastic lattice gas models* (1990)

$\beta > \beta_c$.

The analysis is much more complex when $\beta > \beta_c$ because of phase-coexistence with regions (interfaces) where the magnetization profile is not slowly varying.

However if the system is in only one phase the problem disappears and the hydrodynamic limit should still be described by a diffusion with D strictly positive.

Indeed:

▶ Spohn and Yau have proved that for $\beta > \beta_c$ the Green-Kubo diffusion coefficient

$$D(m) > 0 \quad \text{if } |m| > m_\beta, \qquad D(m) = 0 \text{ otherwise}$$

$\beta > \beta_c$, finite volume effects.

Thus **if both** m_+ **and** m_- **are** $\geq m_\beta$ **(or both** $\leq -m_\beta$**)** we expect in the hydrodynamic limit that the **Fick law is satisfied** as when $\beta < \beta_c$.

▶ In a finite $L \times L$ box the evolution is then well approximated by the regular diffusion described above, provided there is a single phase.

Here comes the key point we will use to explain the phenomenon of uphill diffusion:

If L is finite (and large) the plus stable region is slightly larger than $[m_\beta, 1]$ *and extends to* $(m_\beta - cL^{-2/3}, 1]$ *(see next slide).*

▶ *Conjecture*: profiles with values in $(m_\beta - cL^{-2/3}, 1]$ are well described by a regular diffusion.

Finite size effect: stable regions are larger.

Consider the canonical Gibbs measure μ with magnetization m in the torus $[0, L]^2 \cap \mathbb{Z}^2$: (see for instance Biskup, Chayes, Kotecký, 2003)

- *If $m \in (m_\beta - cL^{-2/3}, m_\beta)$, c small enough, then μ is supported by configurations with "small" contours (of size $\leq \log L$).*

  There is no phase separation.

- *Instead if $m = m_\beta - cL^{-2/3}$ with c large there is a droplet of size $L^{2/3}$.*

Thus:

- $(-m_\beta, -m_\beta + cL^{-2/3})$ is the *minus metastable region*
- $(m_\beta - cL^{-2/3}, m_\beta)$ is the *plus metastable region*

The conjecture I stated before is that if both m_- and m_+ are in the plus metastable region (or both in the minus metastable region) then the profile is well described by a regular diffusion.

In our case we want m_+ plus-stable/metastable and $m_- = -m_+$ minus-stable/metastable, there is then necessarily an interface and the above considerations do not apply.

$\beta > \beta_c$: equilibrium.

To understand what happens we start from equilibrium considering the canonical Gibbs measure with Hamiltonian $H_0 + H_b$ and total magnetization $m = 0$.

This is the Wulff problem first studied by Dobrushin, Kotecký, Shlosman.

Wulff shape.

It is proved that the typical configurations have the following structure:

there is a vertical strip centered at $L/2$ of macroscopically infinitesimal thickness: to the right of the strip the magnetization is essentially m_β and to the left $-m_\beta$; or viceversa.

Without the additional hamiltonian H_b the magnetization in the last column differs from m_β (or $-m_\beta$) and this is why we have added H_b.

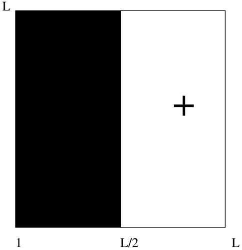
We have used ideas in:

▶ Bodineau and Presutti (2003) where they study surface tension and Wulff shape without spin flip symmetry.

Finite volume corrections.

If L is finite the magnetization of the last column is not exactly equal to m_β (or to $-m_\beta$).

We thus compute its value by running simulations of the Kawasaki dynamics with hamiltonian $H_0 + H_b$ starting from a configuration with:
$\sigma(x) = -1$, $x = (i, j)$, $i \leq \frac{L}{2}$ and $\sigma(x) = 1$ elsewhere.



We have computed the time asymptotic magnetization m^* in the last column. When $L = 50$ we get:

$$m^* = \frac{1}{T} \sum_{t=t_0}^{t_0+T} \sum_{j=1}^{L} \sigma_t(L, j) \approx 0.99926, \qquad t_0 = 10^9, T = 10^5$$

which is very close to the equilibrium magnetization

$$m_\beta = 0.9992757 \text{ (Onsager)}$$

Critical value of m_+.

Observe that m^* must be very close to the critical value m_{crit} of m_+ because:

if $m_+ = m^*$ *(and* $m_- = -m^*$*) then the reservoirs are trying to impose a magnetization which is already there so that their influence is negligible.*

Therefore the current in the presence of the reservoirs is essentially the current without reservoirs which is 0.

We have thus explained the relation between the critical value of m_+ and the equilibrium spontaneous magnetization m_β.

We have not made many progresses in the analysis of the model and we have thus looked for simplifications.

We have tried two of them.

▶ The first one is to study the regime where $\beta \to \infty$ and time is scaled appropriately.

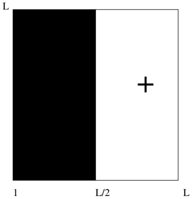
Analogous regime has been considered by:

- den Hollander, Nardi, Olivieri, Scoppola (2000), (2003)
- A. Bovier · F. den Hollander · F.R. Nardi (2006)

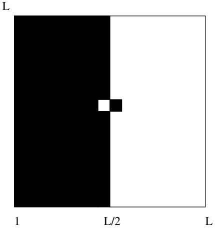
to identify size and shape of the critical droplet and the time of its creation in the limit as $\beta \to \infty$.

The $\beta \to \infty$ limit: $m_+ = 1$.

We start from the initial configuration:



The first change occurs on the time scale $e^{12\beta}$ when the spins in a bond across the interface exchange their values.
At this time we have a - in a set of +'s and a + in a set of -'s.



The $\beta \to \infty$ limit: $m_+ = 1$

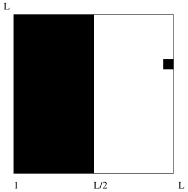
With probability 1 as $\beta \to \infty$ the discrepancies die and the initial state is re-stablished on the time scale $e^{4\beta}$

This can happen in two ways:

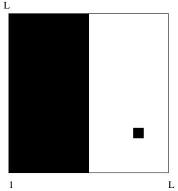
- the discrepancies are killed by the reservoirs (the - is absorbed by the right reservoir and the + by the left reservoir)
- the discrepancies meet at the interface and swhich back.

The $\beta \to \infty$ limit: $m_+ = 1 - e^{-a\beta}$, $a \in (0, 4)$

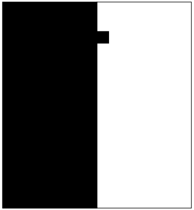
The first change is when either a - is created on the last column or a + on the first column.



The discrepancy is either re-absorbed by the reservoir or moves inside. In the latter case it moves as a random walk with intensity 1 till it either reaches the interface or it is absorbed by the reservoir.



If it reaches the interface it "sticks"



because the intensity for detaching is $e^{-4\beta}$ and so this will not be seen on the time scale $e^{a\beta}$.

We have therefore a two-sided kind of *diffusion-limited aggregation.*

The analysis of this process and of what happens on longer times scale becomes too complicated and we did not make many progresses.

Pinning the interface.

▶ A second type of simplifications which is more successful than the previous one deals with a main difficulty, namely to control location and size of the interface.

To this end we add to the Hamiltonian H the energy due to a space dependent magnetic field

$$h(x) = \varepsilon \mathbf{1}_{i > \frac{L}{2}} - \varepsilon \mathbf{1}_{i \leq \frac{L}{2}}, \qquad x = (i, j)$$

The corresponding Gibbs measure is a sort of Dobrushin state with a localized interface and magnetization approching $m_{\beta,\varepsilon}$ when moving to the right of the interface.

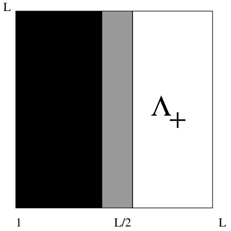
$m_{\beta,\varepsilon} > m_\beta$ (m_β is the spontaneous magnetization for H.)

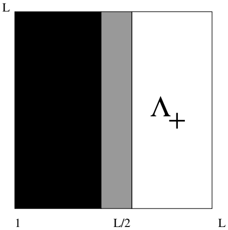
Reservoirs and an external magnetic field

- We now consider the process with the reservoirs and with m_+ "+ stable or metastable" and $m_- = -m_+$.

It is natural to conjecture that as in equilibrium, in the stationary state there is a vertical interface localized around the middle of the box.

Consider a "small (macroscopically infinitesimal) vertical strip" around the interface and call $\Lambda_\pm$ the regions to the right and left of the strip. We may suppose that the magnetization at the end of the strip are $\approx \pm m_{\beta,\varepsilon}$.





Notice that in Λ_+ the magnetic field is constant so that the bond exchange rates are the same as when $\varepsilon = 0$. The reservoirs spin flips are also independent of ε.

Then in the region Λ_+ we are in set up where the Fick law approximately holds with boundary values $m_{\beta,\varepsilon}$ and m_+ provided that $m_+ > m_\beta - cL^{-2/3}$, c small.

Then the sign of the current j depends on the difference $m_{\beta,\varepsilon} - m_+$ and $j > 0$ if the difference is positive.

Reservoirs with an external magnetic field.

- If $m_+ \in [m_\beta, m_{\beta,\varepsilon})$ in the hydrodynamic limit the stationary macroscopic magnetization density $m(r_1)$, $r_1 \in (0,1)$ satisfies:

$$j = -D(m)\frac{dm}{dr_1} = \text{const.}, \qquad r_1 \in (0, \frac{1}{2}) \cup (\frac{1}{2}, 1)$$

$$m(0) = m_-, \; m(\frac{1}{2}) = -m_{\beta,\varepsilon}, \quad m(\frac{1}{2}) = m_{\beta,\varepsilon}, \; m(1) = m_+$$

▶ In the hydrodynamic limit we get a positive current hence uphill diffusion.

This is in contrast to what claimed in the introduction but it is not a contradiction because the effect is due to the external magnetic field.

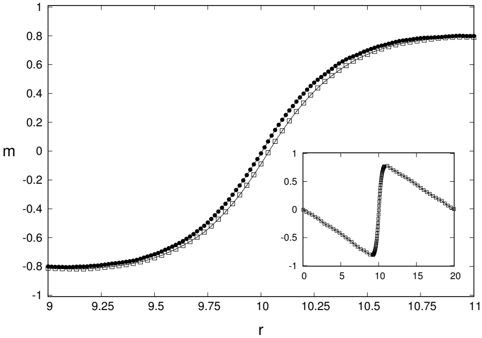
Indeed the magnetic field produces a positive current even when there is no phase transition with a magnetization jump in the middle.

Consider for instance the case $H = 0$. The Kawasaky dynamics is the well known exclusion process except for the bonds which cross the interface: in that case the exchange $+ \Rightarrow -$ is favored with respect to the opposite one due to h.

A 1d version of this is system has been studied by:

• Bodineau, Derrida, Lebowitz, (2010) *A diffusive system driven by a battery or by a smoothly varying field.*

We have considered a similar model but not in a circle and with reservoirs at the ends. (Colangeli, DM, Presutti (2017)).



At the bottom right corner we plot the magnetization profile.

The profile in the large picture is an enlarging of the profile around the middle .

In macroscopic units the transition region becomes a discontinuity point.

Uphill diffusion.

In these cases we have uphill diffusion namely a current which goes uphill from smaller to larger values of the magnetization in the region where the magnetic field changes sign.

This phenomenon was first observed by:

- L.S. Darken (1949) *Diffusion of carbon in austenite with a discontinuity in composition* Transactions of the American Institute of Mining, Metallurgical and Petroleum Engineers **180**

Uphill diffusion: Darken experiment

Quoting Darken: *In order to demonstrate the existence of such diffusion in metals a series of four weld-diffusion experiments was made. In these measurements pairs of steel of virtually the same carbon content, were weld at the end and held at* 1050° *C for about 2 weeks. Subsequent analysis showed that the carbon had diffused so as to produce an inequality of carbon content on the two sides of the weld.... The uphill diffusion of carbon is most clear in fig.2.... The large difference is occasioned by the high silicon content of one side.... Thus silicon decreases and.... gives a pronounced migration of carbon to the high carbon side.*

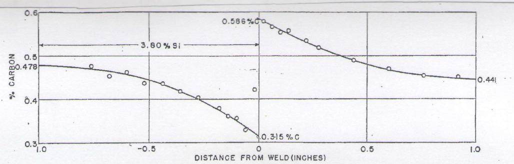


FIG 2—CARBON DISTRIBUTION IN WELDED SPECIMEN NO. 2 AFTER 13 DAYS AT 1050°C.

Uphill diffusion

The relation with the previous models is that the role of silicon is represented by our magnetic field which has a discontinuity at the middle and the carbon particles are then represented by the spins values +1 (-1= empty site).

For recent survey on uphill diffusion see for instance:

Rajamani Krishna, *Uphill diffusion in multicomponent mixtures* Chem. Soc. Rev., **44**, 2812–2836 (2015)

Thus the positive current found by adding the external magnetic field may be produced by the magnetic field itself and vanishes when it is removed. ($m_+ \in [m_\beta, m_{\beta.\varepsilon})$)

Here enter into play the finite volume effects:

if we take $m_+ \in (m_\beta - cL^{-2/3}, m_\beta)$ then the difference $m_{\beta,\varepsilon} - m_+$ is not only positive but uniformly positive as $\varepsilon \to 0$.

Thus when $m_+ = m_\beta - cL^{-2/3}$ we have a positive current uniformly in ε which scales like $L^{-1}L^{-2/3}$.

This might look as the desired explanation of the uphill diffusion phenomenon, but it is not quite so.

In fact the simulation show that $\varepsilon = 0$ is quite different than $\varepsilon > 0$ if $m_+ \in (m_\beta - cL^{-2/3}, m_\beta)$.

We see that the magnetization profile when $\varepsilon = 0$ moves away from the middle of the domain and goes to the boundary.

Nonetheless the current we measure is still positive.

Simulations

Below I show the computer simulations.

Recall that we have argued that $m_{\text{crit}} \approx 0.99926$

- Start with the interface at $\frac{L}{4}$ and with $m_+ = 0.99936$
- Start with the interface at $\frac{L}{4}$ and with $m_+ = 0.998$
- For smaller values of m_+ strange phenomena appear. The simulation refers to $m_+ = 0.9$