Reduction of coverage due to surface steps

K. Morawetz[1,2], C. Olbrich[3], S. Gemming[4], M. Schreiber[1]

[1] Institute of Physics, Chemnitz University of Technology, Reichenhainer Str. 70, 09107 Chemnitz, Germany
[2] Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Str. 38, 01187 Dresden, Germany
[3] Jacobs University Bremen gGmbH Postfach 750561, 28725 Bremen Germany
[4] Forschungszentrum Rossendorf, PF 51 01 19, 01314 Dresden, Germany

CHEMNITZ UNIVERSITY OF TECHNOLOGY

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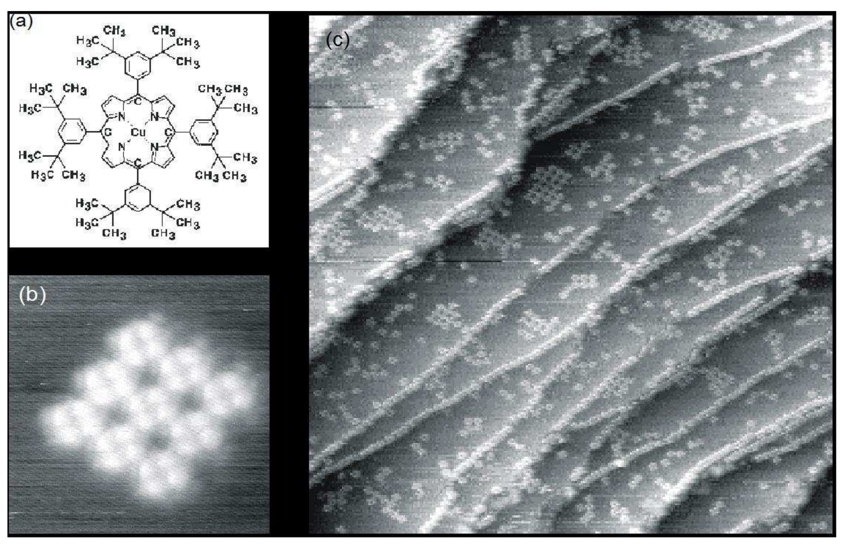
Experimental motivation

Adsorbed molecules

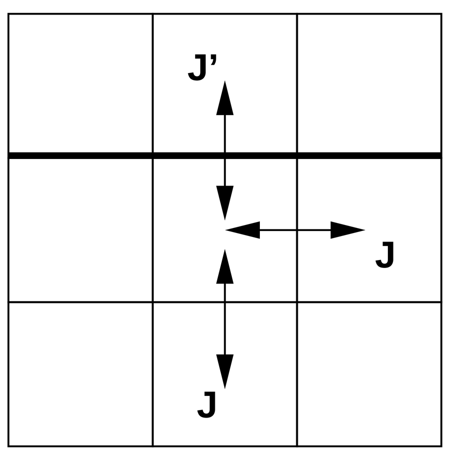


Cu-tetra(3,5 di-t-butylphenyl) porphyrin: Cu-TBPP — Cu(110)

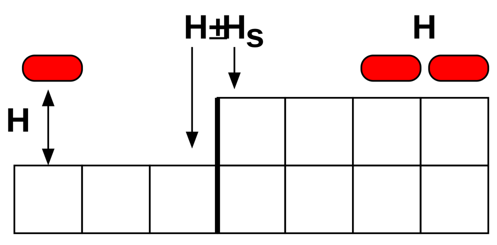
Loppacher et al., Appl. Phys. A72, S105-S108 (2001)

Modified 2D Ising model

Lateral interaction:



Vertical interaction:



•external magnetic field synonym for coupling of molecules with background
•adjacent sites to step magnetic field augmented by $H_s > 0$, Ehrlich-Schwoebel barrier: impedes diffusion of adsorbates across surface steps
•Total energy:

$$E_{tot} = \sum_{ij} J\, s_i s_j + \sum_{[ij]} J'\, s_i s_j + \sum_{i} H\, s_i + \sum_{[i]} (H \pm H_s) s_i$$

$s_i = 1$: occupied; $s_i = -1$: empty

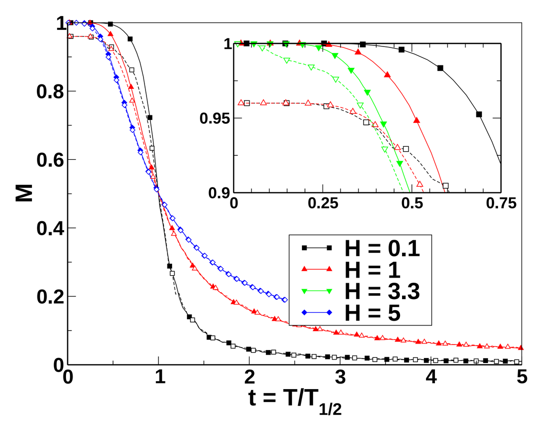
$\Downarrow$

Monte-Carlo simulation with Metropolis algorithm

$\Downarrow$

Pattern analysis with Hoshen-Kopelmann algorithm

Reduction of mean spin due to step



$J = 1$, $J' = 1/\sqrt{2}$, $H_s = -5$
with step: dotted lines
without step: solid lines

- Reduction of mean spin for low tempertures only for low external field due to the step
- Occurence of an critical external field
- For $T > T_{1/2}$ same behaviour with step as without

Ground state discussion

• Normal mean spin s, at the step $s^{\pm}$ according to Ehrlich-Schwoebel barrier
• Spin flip at s^- energetically favored over flip at s
• Site energy difference $\Delta E^- = E^-(\uparrow\downarrow) - E^-(\uparrow\uparrow)$ between parallel and antiparallel alignment of site s^- with respect to neighbor sites:

$$\begin{aligned}\Delta E^- &= [-J's^+s^- - Jss^- - 2J(s^-)^2 - \mu_0(H-H_s)s^-] \\ &- [-J's^+s^+ - Jss^+ - 2J(s^+)^2 - \mu_0(H-H_s)s^+] \\ &= J(2(s^+)^2 - 2(s^-)^2 + ss^+ - ss^-) + J'((s^+)^2 - s^+s^-) \\ &\quad -\mu_0 H(s^- - s^+) + \mu_0 H_s(s^- - s^+)\end{aligned}$$

• Equating $s^+ = s$ and $s^- = -s$ energy difference $\Delta E^- = -\mu_0 H + \mu_0 H_s - Js - J's$
• Anti-parallel orientation is favorable if $\Delta E^- < 0$
• Condition for transition ($T = 0$):

$$\mu_0 H + J + J' < \mu_0 H_s$$

Effective two-spin mean-field model

• Spins s on attractive sites and fictitious spins s' at the repulsive sites with $H - H_s$ along step
• Mean field of the normal spins

$$\mu_0 \tilde{H} = \mu_0 H + \frac{N-3}{N-1} 4Js + \frac{6s+2s'}{N-1} J \equiv \mu_0 H + jJs.$$

$jJs = 4Js + 2Jc/(N-1) \approx 4Js$ with error of $1/(N-1)$
• Effective spins consisting of linear combination of J, J'
Second transition occur at temperature $T_c' \sim J'$: effective model

$$\mu_0 \tilde{H}' = \mu_0 H - \mu_0 H_s + (J+J')s + 2J's',$$

Partition function

$$\begin{aligned}s' &= \tanh\left[\beta(\mu_0(H-H_s) + (J+J')s + 2J's')\right] \\ s &= \tanh\left[\beta(\mu_0 H + jJs)\right],\end{aligned}$$

Total mean spin

$$M = \left(1 - \frac{1}{N}\right) s + \frac{1}{N} s'$$

approaches reduced value $M = 1 - 2/N$ ($T = 0$) at correct condition

Exact results for defect lines

Ising 2D exact solution [4]
Different surface defects studied within Ising models [5]
Phase transitions in inhomogeneous systems [3],
Without fields H and H_s Ising square lattice with ladder defect [3] has exact critical exponent [6]

$$\beta_{\mathrm{H}=0} = \frac{1}{2\pi^2} \arccos^2\left(-\tanh\frac{2(J-J')}{k_B T}\right).$$

We investigate influence of Ehrlich-Schwoebel barrier and magnetic field

Failure of standard mean-field model

• $N \times N$ spins and periodic boundary conditions
• $N-2$ normal Ising spins, s, and 1 spin at each side of the step $s^{\pm}$
• $N-4$ normal spins see a mean-field $\tilde{H}$ consisting of the external field H and the interaction with 4 neighbors, remaining two of $N-2$ normal spins interact only 3 normal spins and one spin at step

$$\mu_0 \tilde{H} = \mu_0 H + \frac{N-4}{N-2} 4Js + \frac{2}{N-2}(3Js + J\frac{s^+ + s^-}{2}).$$

spins along step have mean-field

$$\mu_0 \tilde{H}^{\pm} = \mu_0 H \pm \mu_0 H_s + 2Js^{\pm} + J's^{\mp} + Js.$$

• Partition function
$z = [2\cosh(\beta\mu_0\tilde{H})]^{(N-2)N} [2\cosh(\beta\mu_0\tilde{H}^+)]^N [2\cosh(\beta\mu_0\tilde{H}^-)]^N$
• Self-consistent mean-field equations (Bragg-Williams) for open surface defects [1, 2] $s = \tanh(\beta\mu_0\tilde{H})$ and $s^{\pm} = \tanh(\beta\mu_0\tilde{H}^{\pm})$.
• Total mean spin

$$M = \left(1 - \frac{2}{N}\right) s + \frac{1}{N}(s^+ + s^-)$$

approaches the reduced value $M = 1 - \frac{2}{N}$ for $T \to 0$ if $\tilde{H} > 0$ and ($\tilde{H}^+ \gtrless 0$ and $\tilde{H}^- \lessgtr 0$)
• Reduction happens if and only if

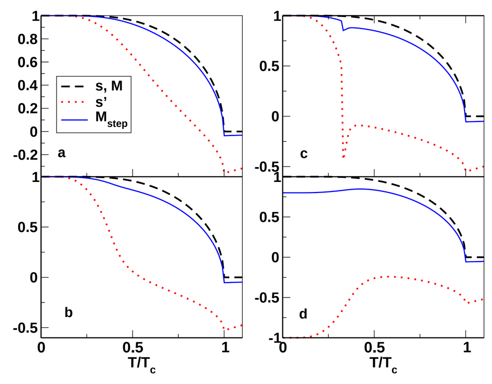
$$\mu_0 H + 3J + J' < \mu_0 H_s$$

Local mean-field model fails due to nonlocal correlations [3]

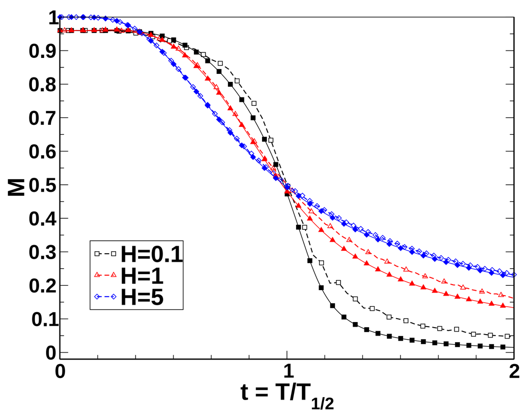
References

[1] D. L. Mills, *Phys. Rev. B* **3**, 3887 (1971).
[2] K. Binder, P. C. Hohenberg, *Phys. Rev. B* **9**, 2194 (1974).
[3] F. Igloi, I. Peschel, L. Turban, *Adv. in Phys.* **42**, 683 (1993).
[4] L. Onsager, *Phys. Rev.* **65**, 117 (1944).
[5] M. C. Chung, M. Kaulke, I. Peschel, M. Pleimling, W. Selke, *Eur. Phys. J. B* **18**, 655 (2000).
[6] R. Z. Bariev, *Soviet Phys. JETP* **50**, 613 (1979).

Effective mean-field model results



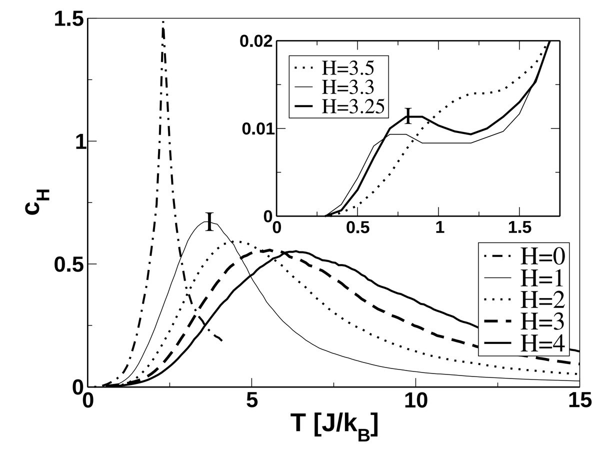
Calculation details:
$N = 10$, $J = 0.1$, $J' = 0.07$, $H = 0$
different Schwoebel barriers:
$\mu_0 H_s = 0.4, 0.3, 0.2, -0.3$ (a-d)



Magnetization of Ising model (open symbols and dashed line) with step together with effective meanfield model (closed symbols and solid lines)

good agreement with the simulation for $T < T_{1/2}$

Specific heat for Ising model with a step



Besides the $H = 0$ curve all curves are regressions and the error bars indicate the corresponding maximal ones

Scaling properties for effective mean-field model

Spontaneous magnetization	$M \sim \lvert T - T_C\rvert^{\beta}$
Magnetic field dependence	$H \sim \lvert M_{T=T_C}\rvert^{\delta}$
Susceptibility	$\chi \sim \lvert T - T_C\rvert^{-\gamma}$
Specific heat	$c_H \sim \lvert T - T_C\rvert^{-\alpha}$

2D Ising model phase transition with critical behaviour fulfilling scaling relations $\alpha + 2\beta + \gamma \geq 2$ and $\alpha + \beta(1+\delta) \geq 2$

	α	β	γ	δ	$\alpha+2\beta+\gamma$	$\alpha+\beta(1+\delta)$
2D Ising (exact)	0	1/8	7/4	15	2	2
2D Ising (Weiss)	0	1/2	1	3	2	2
T_c $N\neq\infty$	3/2	0	1	0	2.5	1.5
T_c $N=\infty$	0	1/2	1	3	2	2
T_c' $N\neq\infty$ ($H_s=0$)	2 (0)	0 (0)	1 (1)	0 (1)	3 (1)	2 (2)
T_c' $N=\infty$ ($H_s=0$)	0 (0)	0 (0)	0 (0)	0 (1)	0 (0)	0 (0)

Critical exponents for the two transitions in mean-field approximation. The value in brackets give the results without Ehrlich-Schwoebel barrier.

Summary

- Occurrence of a second transition besides Ising phase transition due to finite surface step
- Local mean-field model describe qualitatively main features, but fail quantitatively due to nonlocal correlations
- Effective two-spin mean-field model constructed K. Morawetz, C. Olbrich, S. Gemming, M. Schreiber, cond-mat/0608013
- Diverging heat capacity appears here due to the spatial distortion besides Ising phase transition
- Signals with large heat capacities are not conclusive for the identification of phase transitions in finite systems
- Simulating the surface coverage with molecules including a step the present model is able to describe main equilibrium features for fabrication of nanowires
Ch. Loppacher, U. Zerweck, L. M. Eng, S. Gemming, G. Seifert, C. Olbrich, K. Morawetz, M. Schreiber, Nanotechnology 17 (2006) 1568-1573