Surprises, Computational Methods, and Results for Metastable Phenomena and Homogeneous Nucleation and Growth

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Supported by NSF, DOE, and HPC^2 C

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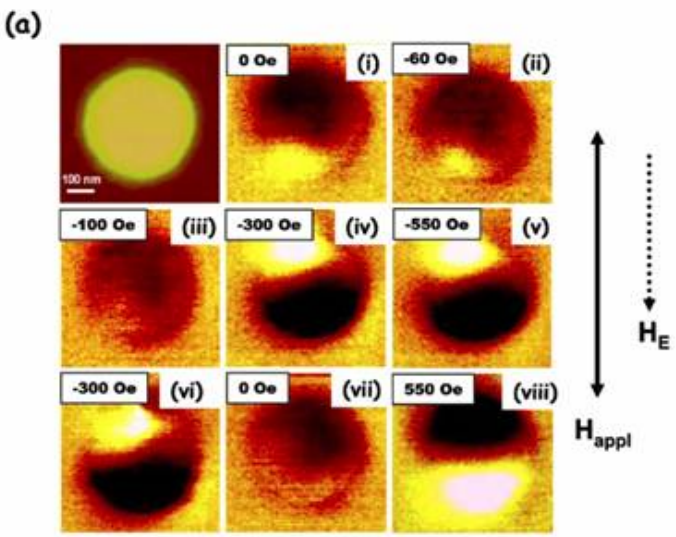
Zoltan Toroczkai (Notre Dame)

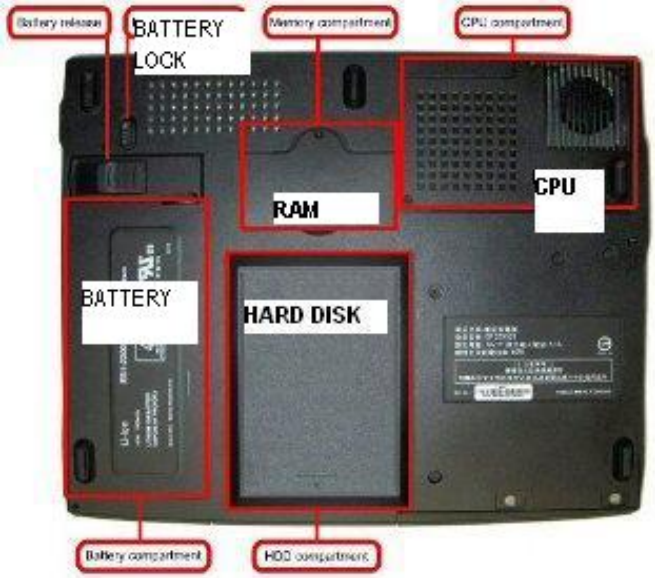
Supported by NSF and DOE

Questions:

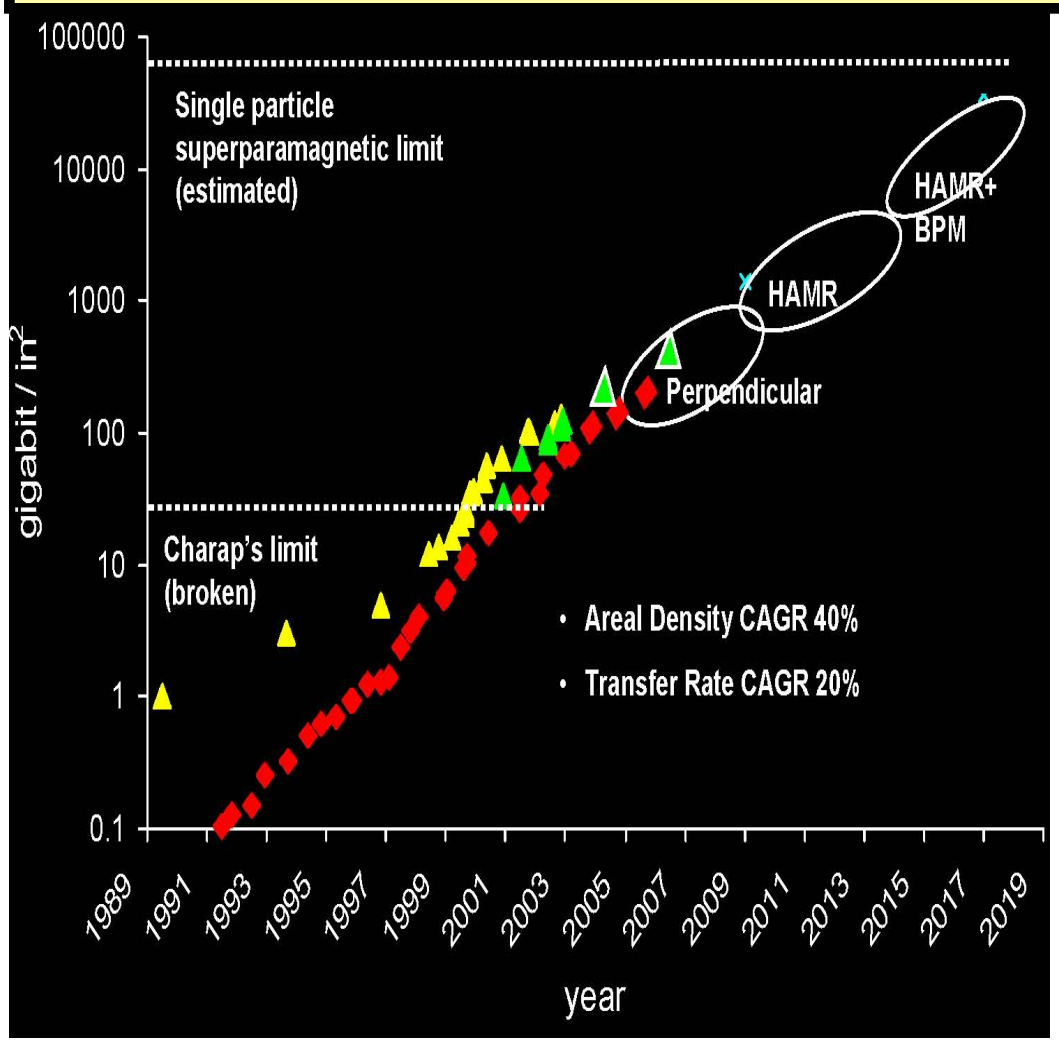
How many of you used magnetic nanoparticles today?

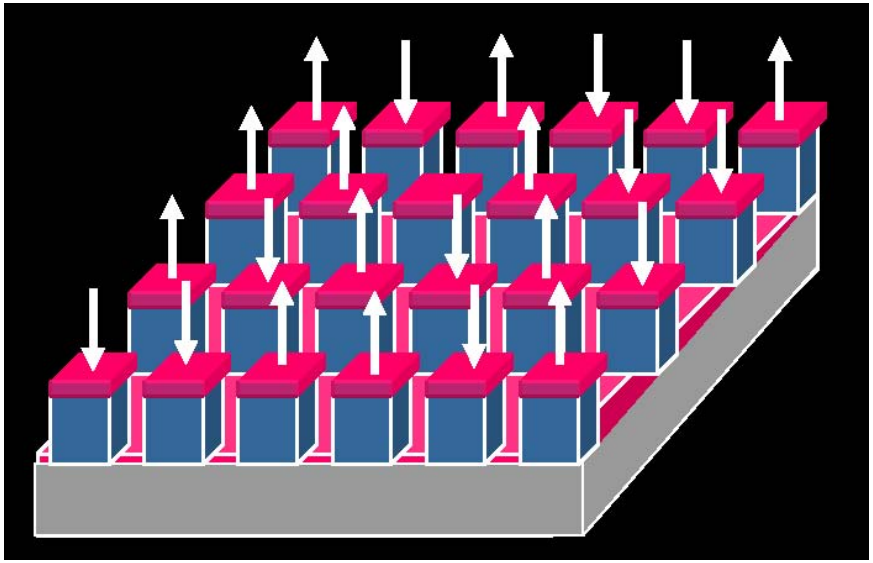


How many of you used a computer today?



Motivation

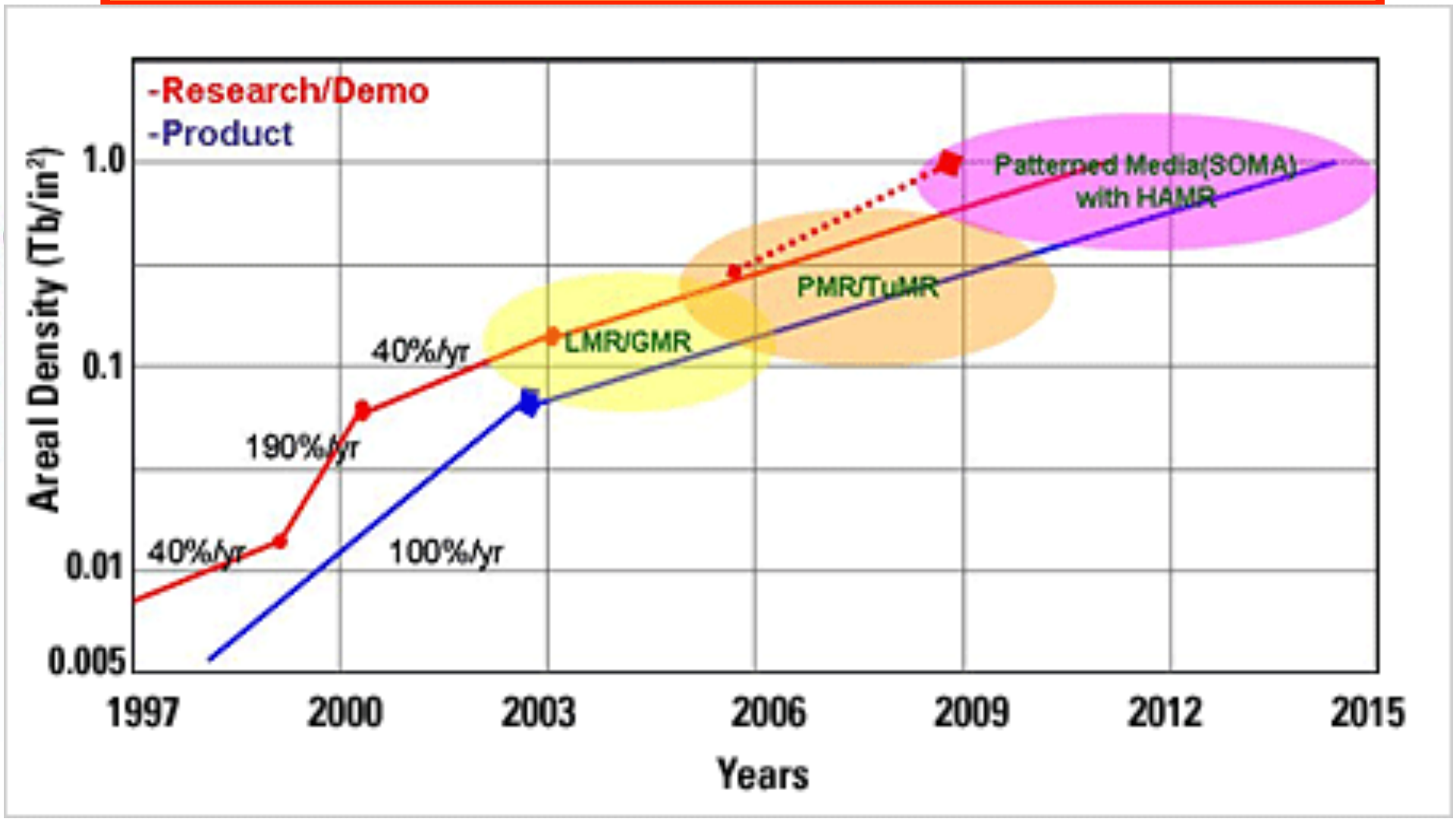




Magnetic recording

Dynamics of nanoscale magnets

Motivation: Increasing magnetic data recording density

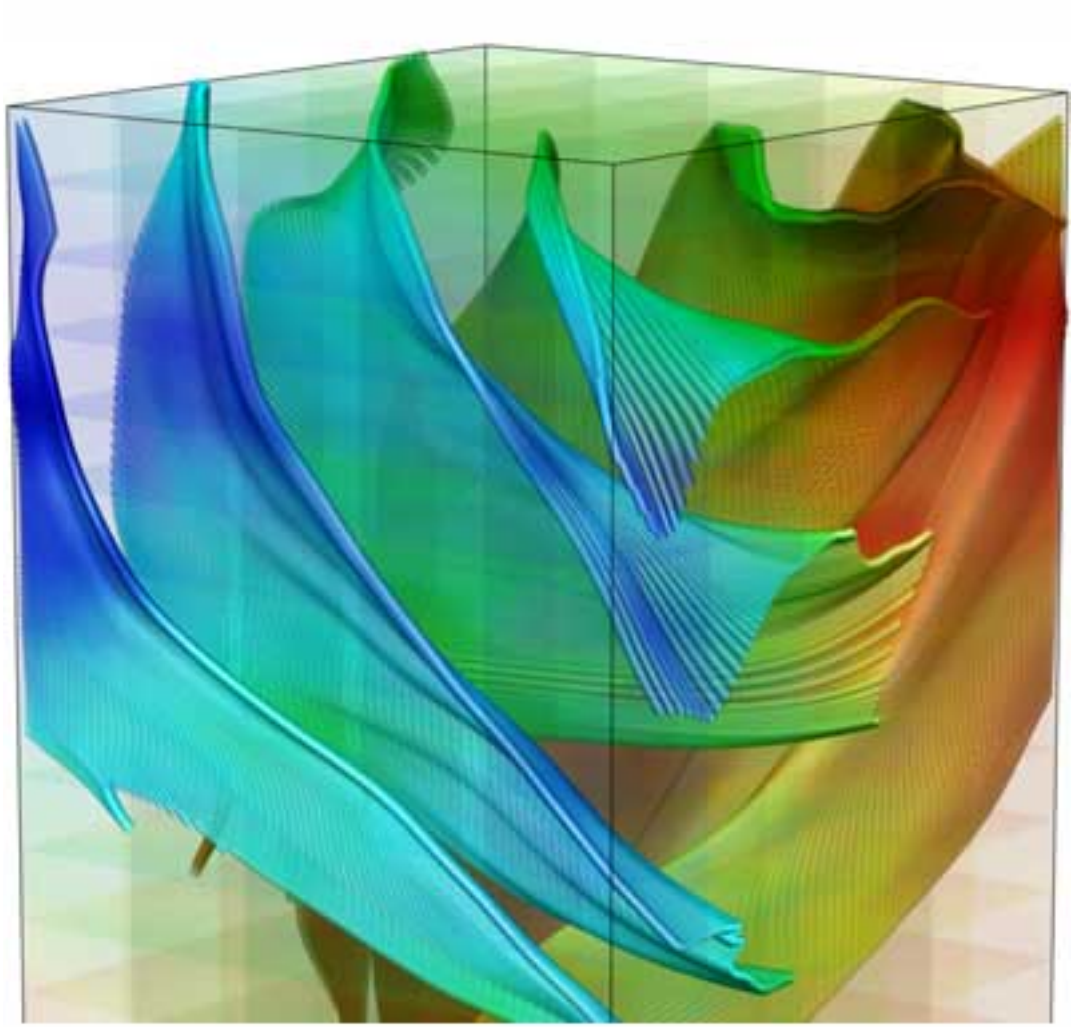


Dynamics of nanoscale magnetic particles

Physical Review B

January 2009

Kaleidoscope



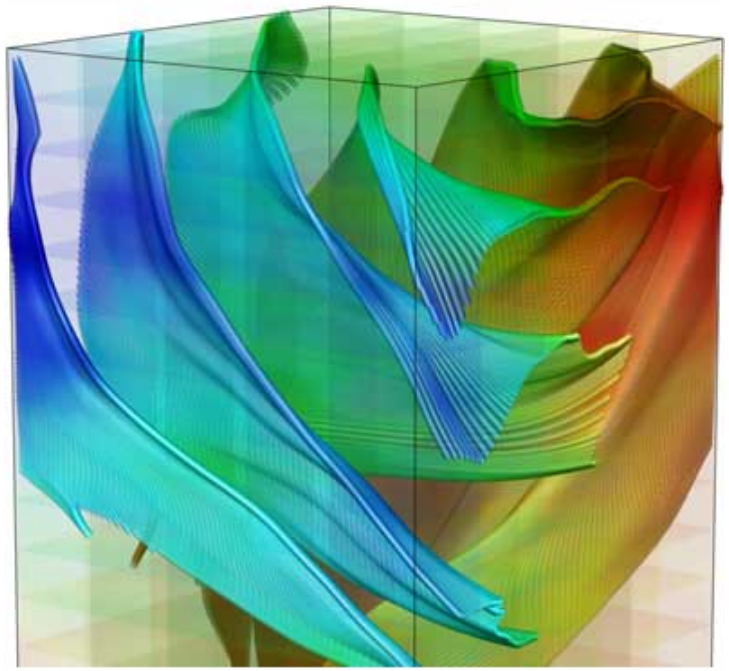
Typical end-cap configuration for a metastable Fe nanopillar.

10 x 10 x 150 nm^3

Phys. Rev. B. **79**, 024429 (2009) [9 pages]

S.H. Thompson, G. Brown, A.D. Kuhnle, P.A. Rikvold, MAN

This is
HETEROGENEOUS
nucleation and growth
--- Too Hard



10 x 10 x 150 nm^3

HOMOGENEOUS NUCLEATION / GROWTH / METASTABILITY

- Homogeneous nucleation and growth is complicated!
- Algorithms can be devised to efficiently study nucleation & growth
- Can changing dynamic change nucleation rate exponential?
- How does hysteresis loop area depend on the f for oscillating field?
- What is a dynamic phase transition? Experimental realization?

Model

2D Ising Hamiltonian on $L \times L$ square lattice: s_i=+1 or s_i=-1

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} s_i s_j - H(t) \sum_{i}^{L^2} s_i$$

Dimensionless magnetization:

$$m = L^{-2} \sum_{i} s_i$$

Temperature $T < T_c \Rightarrow m$ for H=0 takes one of two degenerate equilibrium values:

$$m(T < T_c,\ H = 0) \ = \ \pm m_{\rm eq}(T)$$

Homogeneous Nucleation and Growth

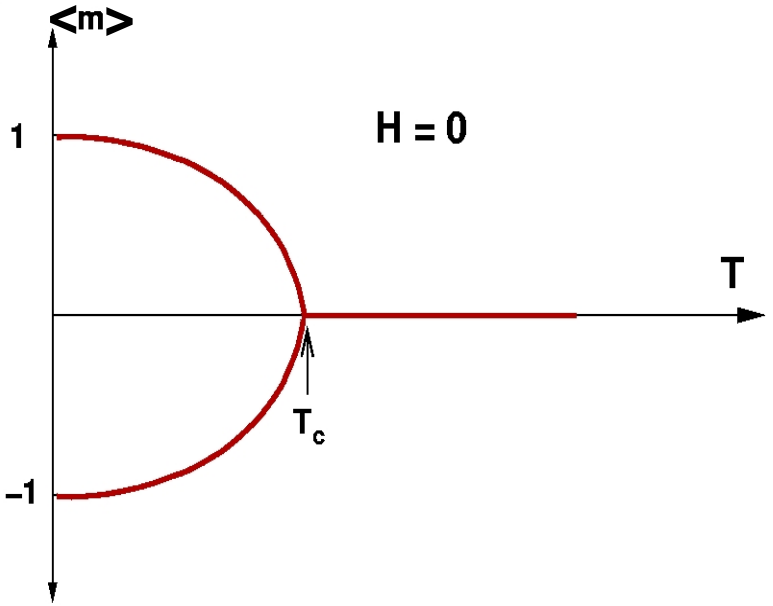
Equilibrium Phase Transition

- Curie transition in ferromagnet is example of *equilibrium* second-order phase transition

- Two-dimensional Ising model with energy

$$E = -J \sum_{<i,j>} S_i S_j - H \sum_i S_i$$

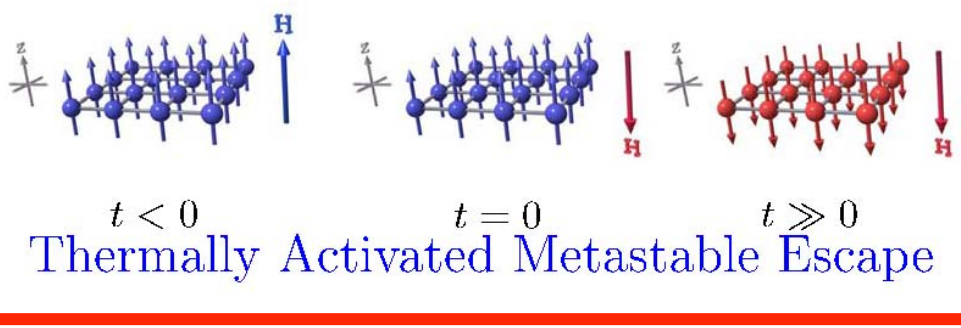
shows this phase transition analytically at H = 0

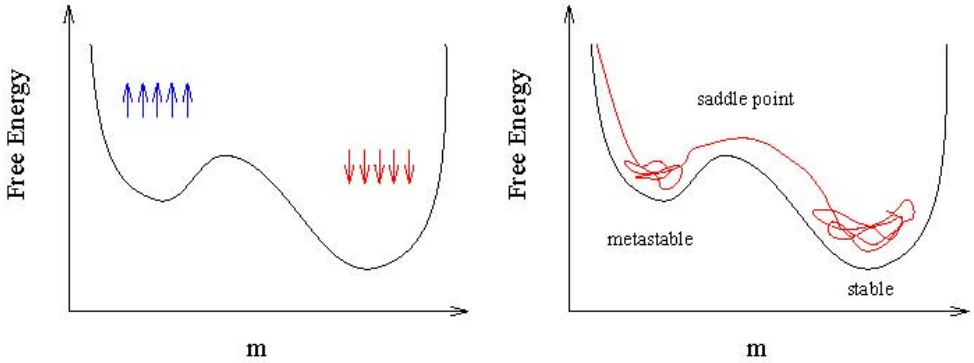


- Transition can be seen in static Monte Carlo simulation of (finite-size) Ising lattices

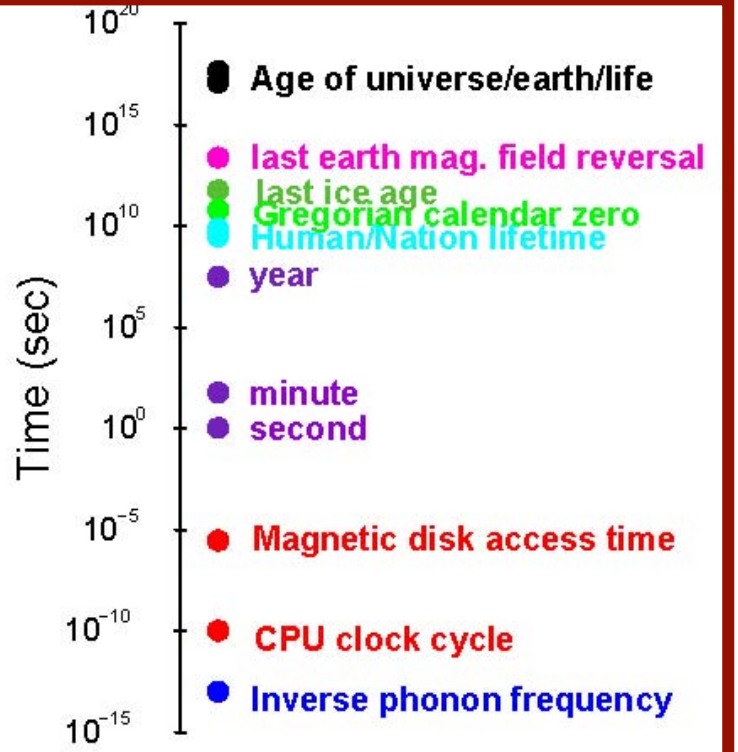
Simulation GOAL:

Faster-than-real-time Simulations



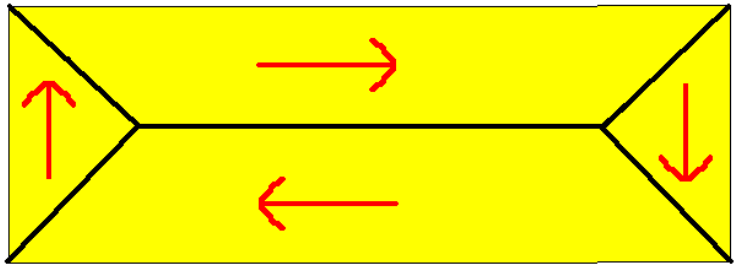


Spanning disparate time scales

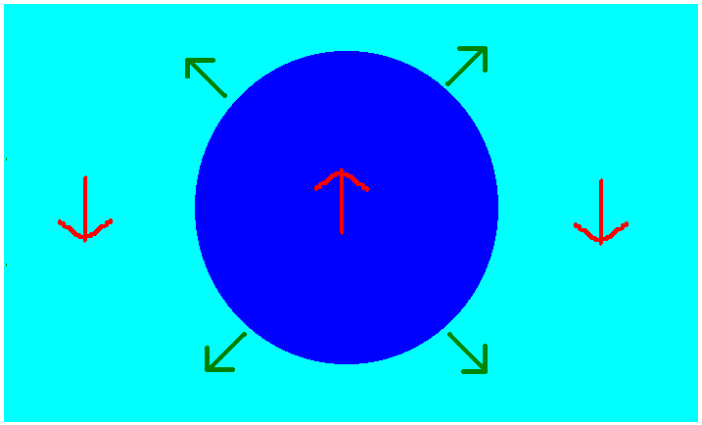


- ω^{-1} = inverse phonon frequency
- bit read/write time
- data stability time

Droplets versus domains



Domains are equilibrium structures that minimize the magnetic energy. Nanometer sized particles can be single-domain.

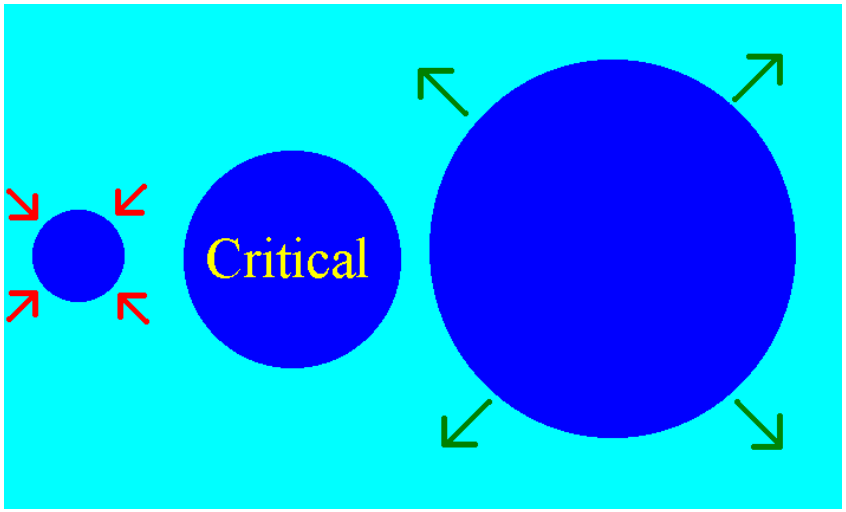


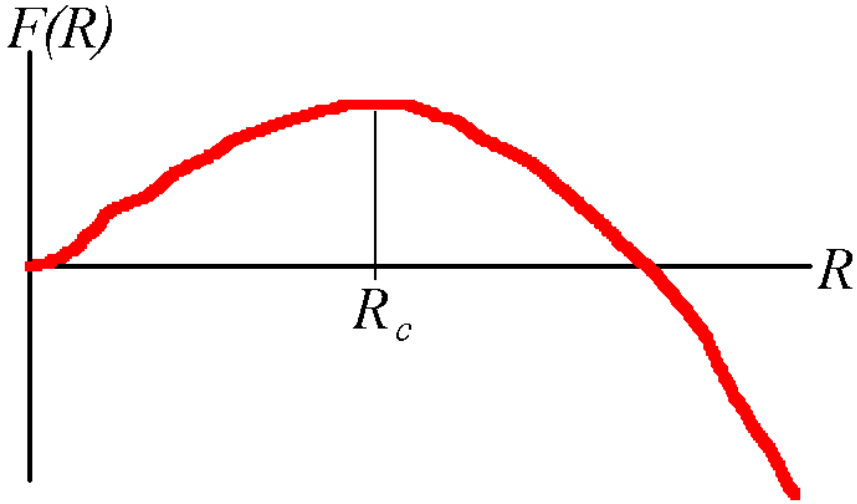
Droplets are nonequilibrium structures that only exist during the process of magnetization switching.

Classical Nucleation theory of metastable decay

Relevant fluctuations are *compact droplets* of radius R and volume $\Omega_d R^d$ with free energy

$$F(R) \approx \Omega_d^{(d-1)/d} \sigma_0(T) R^{d-1} - |H| 2 m_s(T) \Omega_d R^d$$





$\sigma_0(T)$: Droplet surface tension.

$m_s(T)$: Spontaneous magnetization.

$F(R)$ is maximum for the *critical radius* $R_c \approx \frac{(d-1)\sigma_0(T)}{2 m_s(T) |H|}$

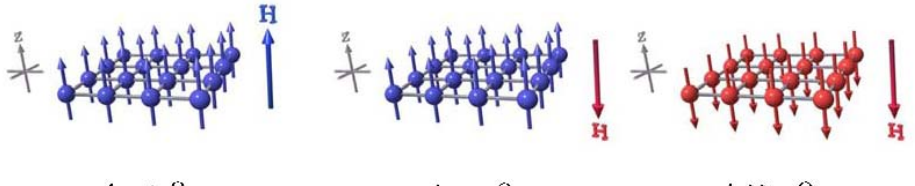
Nucleation rate: $\Gamma(T,H) \propto |H|^K \exp\left[-\frac{\beta \Xi(T)}{|H|^{d-1}}\right]$

$\Xi(T)$ and K known for d=2 Ising model. *No* sharp spinodal!

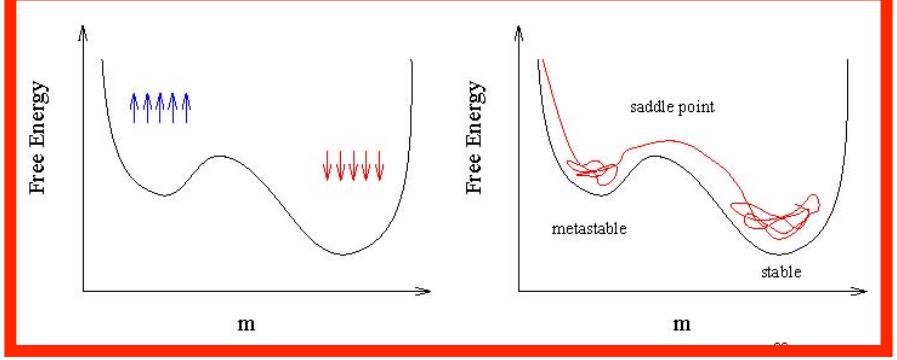
Monte Carlo dynamic: Field Reversal

Ising model

- Square lattice, $L \times L$
- Lattice sites have spins $s = \pm 1$
- Periodic boundary conditions
- Nearest-neighbor interactions, strength $J > 0$
- Interaction with external magnetic field $H < 0$
- Start with all $s = +1$
- Measure τ, average first time when $\sum_i s_i = 0$



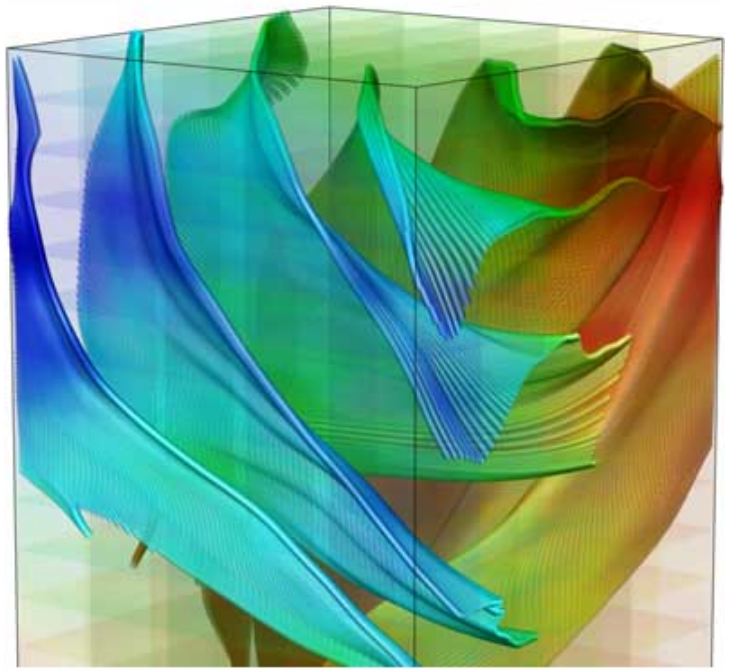
Thermally Activated Metastable Escape



- Randomly choose a lattice point
- Calculate energy change ΔE if spin s_i changes
- Calculate transition probability

  $W_{s_i \to -s_i} = [1 + \exp(\Delta E / k_B T)]^{-1}$ fermion: Martin '77

  $W_{s_i \to -s_i} = \left| \Delta E \, [1 - \exp(\Delta E / k_B T)]^{-1} \right|$ phonon: Park '01
- Calculate a random number r
- Flip spin $s_i \to -s_i$ if $r \leq W$
- Repeat $\sim 10^{30}$ times!!!!!!



$10 \times 10 \times 150\ nm^3$

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HOMOGENEOUS Nucleation and Growth Theories

- Activated Barrier Crossing
  - Van't Hoff (1884); Arrhenius (1889); Kramers (1940)
  - Lifetime $\langle \tau(T,H,L) \rangle \propto 1/[L^d I(T,H)]$
  - Nucleation rate per unit [illegible]
    $I(T,H) \propto \omega_0 \exp(-E_{\rm activation}/k_{\rm B}T)$
- Néel-Brown (1949, 1959); magnetization reversal
  - $\langle \tau \rangle \propto \exp\left(\frac{\Delta}{L^d k_{\rm B} T}\right)$
- Imaginary part of Free energy; Langer (1968, 1969)
  - Analytic continutation
  - $I(T,H) = \frac{\kappa}{\pi k_{\rm B} T} \left| {\rm Im}(\mathcal{F}_{\rm ms}) \right|$
  - $\left| {\rm Im}(\mathcal{F}_{\rm ms}) \right| = B(T) |H|^b \exp\left[- \frac{\Xi(T)}{k_{\rm B} T |H|^{d-1}} \right]$
- KJMA theory
  - Overlapping, independent, non-interacting droplets
  - Kolmogorov (1937)
  - Johnson & Mehl (1939)
  - Avrami (1939, 1940, 1941)
  - $\langle \tau \rangle \propto \left[\Omega v^d I(T,H) \right]^{-1/(d+1)}$ L^0
  - $v \approx |H| \nu$ Allen-Cahn

$L^?$

Anders Celsius (1701-1744)

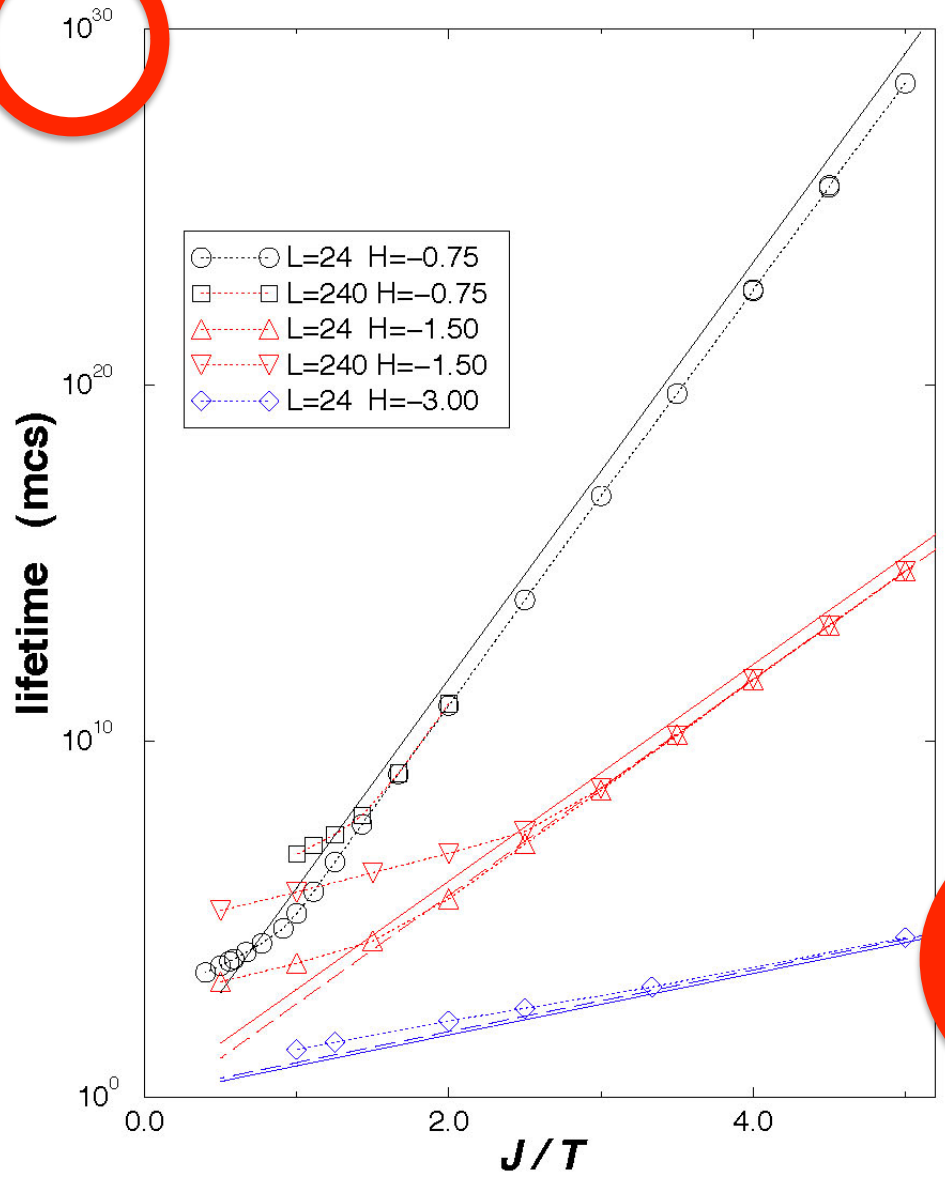
Hermann von Helmholtz (1821-1894)

Svante Arrhenius (1859-1927)

James Clerk Maxwell (1831-1879)

HOMOGENEOUS Nucleation and Growth

d=2 Ising ferromagnet : "Low Enough T, L fixed"



Arrhenius result

Classical nucleation

(low T)

$< \tau > = \tilde{A} \exp(\beta\Gamma)$

$\beta = 1/k_B T$

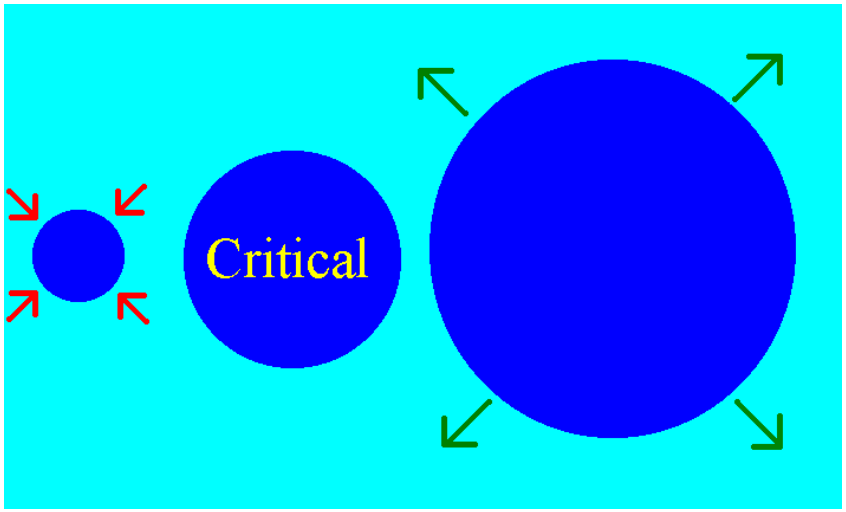
$T \to 0$ exact result $\quad \langle \tau \rangle = \exp(\Gamma/T)$

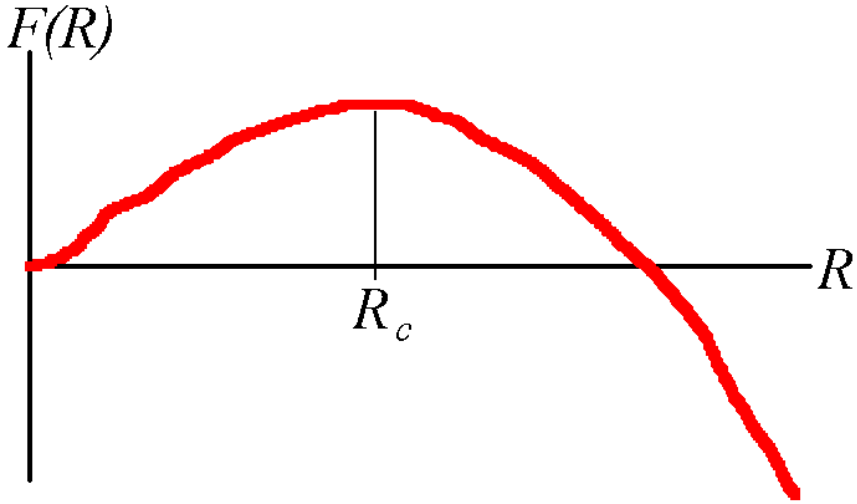
- $\Gamma(H, J) = 8J\ell_c - 2|H|(\ell_c^2 - \ell_c + 1)$
- $\ell_c = \lceil 2J/|H| \rceil$
- $\lceil x \rceil$ = smallest integer not less than x

Nucleation theory of metastable decay

Relevant fluctuations are *compact droplets* of radius R and volume $\Omega_d R^d$ with free energy

$$F(R) \approx \Omega_d^{(d-1)/d} \sigma_0(T) R^{d-1} - |H| 2 m_{\rm s}(T) \Omega_d R^d$$





$\sigma_0(T)$: Droplet surface tension.

$m_{\rm s}(T)$: Spontaneous magnetization.

$F(R)$ is maximum for the *critical radius* $R_{\rm c} \approx \frac{(d-1)\sigma_0(T)}{2 m_{\rm s}(T)|H|}$

Nucleation rate: $\Gamma(T,H) \propto |H|^K \exp\left[-\frac{\beta \Xi(T)}{|H|^{d-1}}\right]$

$\Xi(T)$ and K known for d=2 Ising model. *No* sharp spinodal!

KJMA theory of metastable decay

Following sudden field reversal, critical droplets nucleate at constant rate per unit volume,

$$I(T,H) = B(T)|H|^K \exp\left[-\frac{\beta\Xi(T)}{|H|^{d-1}}\right]$$

Ξ and $K = 3$ exactly known for 2D Ising model.

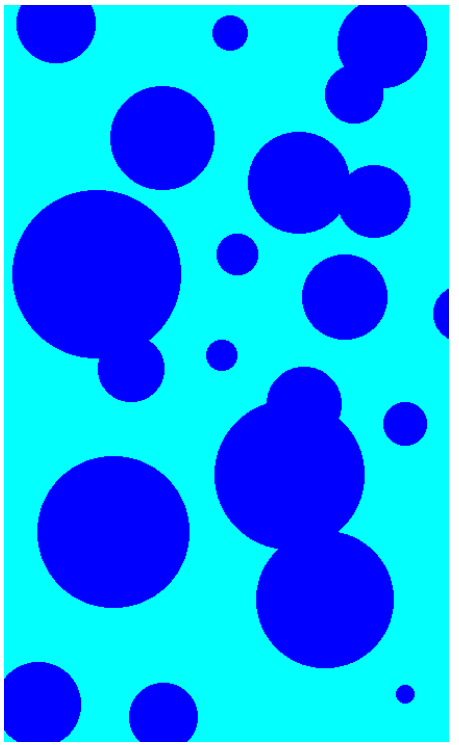
Large supercritical droplets grow at constant velocity v (Lifshitz-Allen-Cahn approximation):

$$v \quad \propto \quad |H|$$

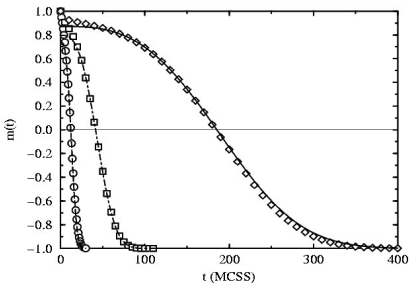
Droplet Growth and Finite-Size Effects in the Ising Model

KJMA (Avrami) theory. (Kolmogorov, Johnson-Mehl, Avrami, 1939-42)
Large supercritical droplets grow at approximately constant speed (Allen-Cahn approximation):

$$\begin{aligned} v_\perp &= (d-1)\nu\left(R_c^{-1} - R^{-1}\right) \\ &\xrightarrow{R\to\infty} (d-1)\nu R_c^{-1} \equiv v_0 \propto |H| \end{aligned}$$



Time evolution of magnetization
(randomly placed, freely overlapping droplets):



$$\begin{aligned} m(t) &\approx 2\exp\left[-\Gamma\int_0^t \Omega_d (v_0 s)^d \mathrm{d}s\right] - 1 \\ &= 2\exp\left[-\frac{\Omega_d}{d+1}\left(\frac{t}{t_0}\right)^{d+1}\right] - 1 \end{aligned}$$

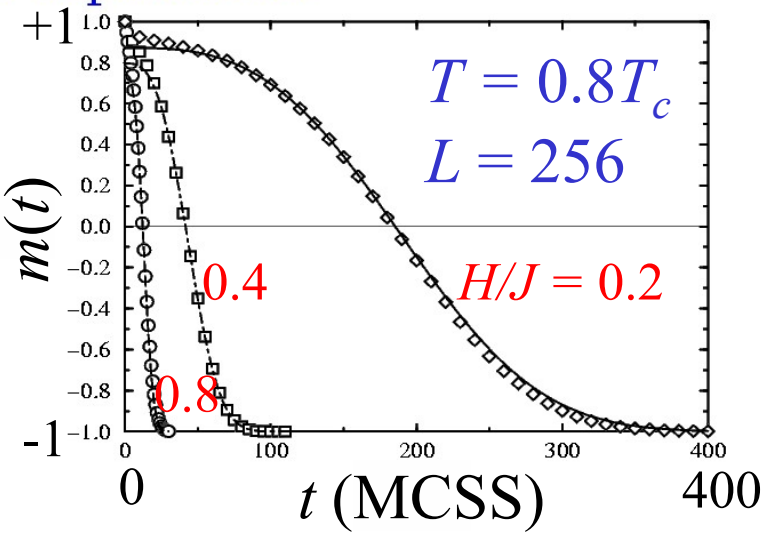
where $t_0 = (v_0^d\Gamma)^{-1/(d+1)}$ is the *average time of free growth*.

Time evolution of magnetization in KJMA theory (randomly placed, freely overlapping droplets):

$$m(t) \approx m_{\rm eq}(T)\left\{2\exp\left[-I\int_0^t \Omega_d (vs)^d {\rm d}s\right]-1\right\}$$

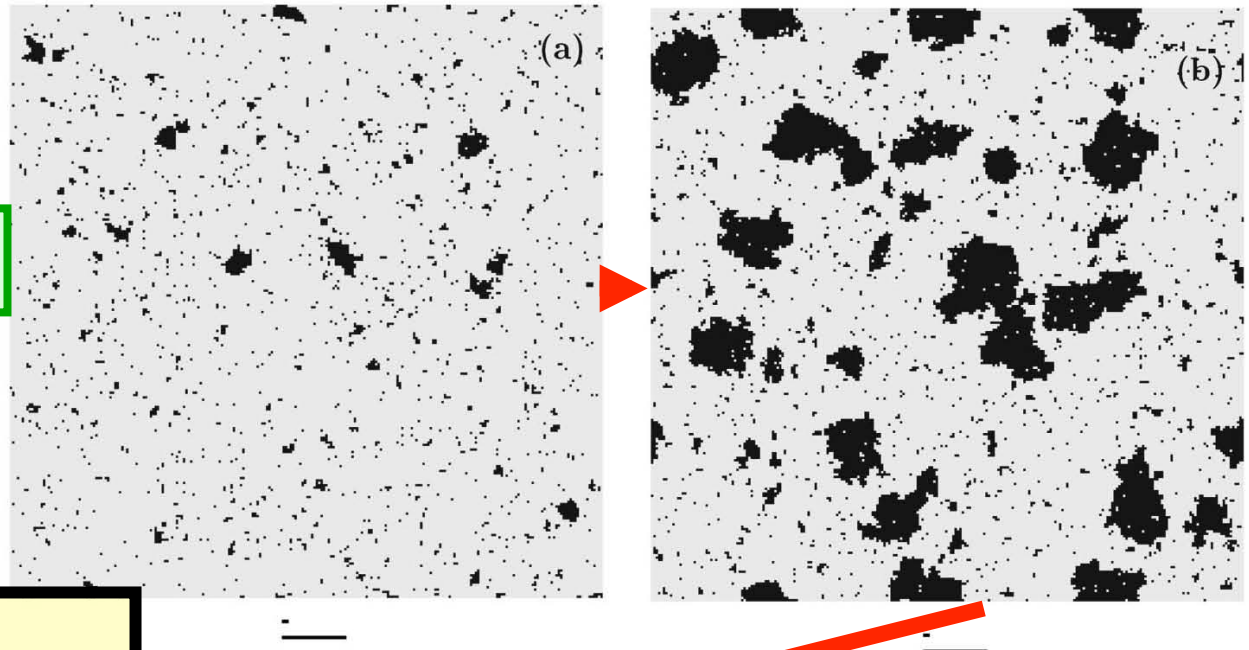
$$= m_{\rm eq}(T)\left\{2\exp\left[-\frac{\Omega_d}{d+1}\left(\frac{t}{\tau}\right)^{d+1}\right]-1\right\}$$

$\langle\tau\rangle = (v^d I)^{-\frac{1}{d+1}}$ is average metastable lifetime. $R_0 \approx v\langle\tau\rangle$ is average droplet separation.



1999

t=80 MCSS



t=260 MCSS

L=250

T=0.8 Tc

|H|=0.15 J

$< \tau >$=392 MCSS

$R_c \approx$ 2.5

$R_0 \approx$25.0

t=390 MCSS≈$<\tau>$

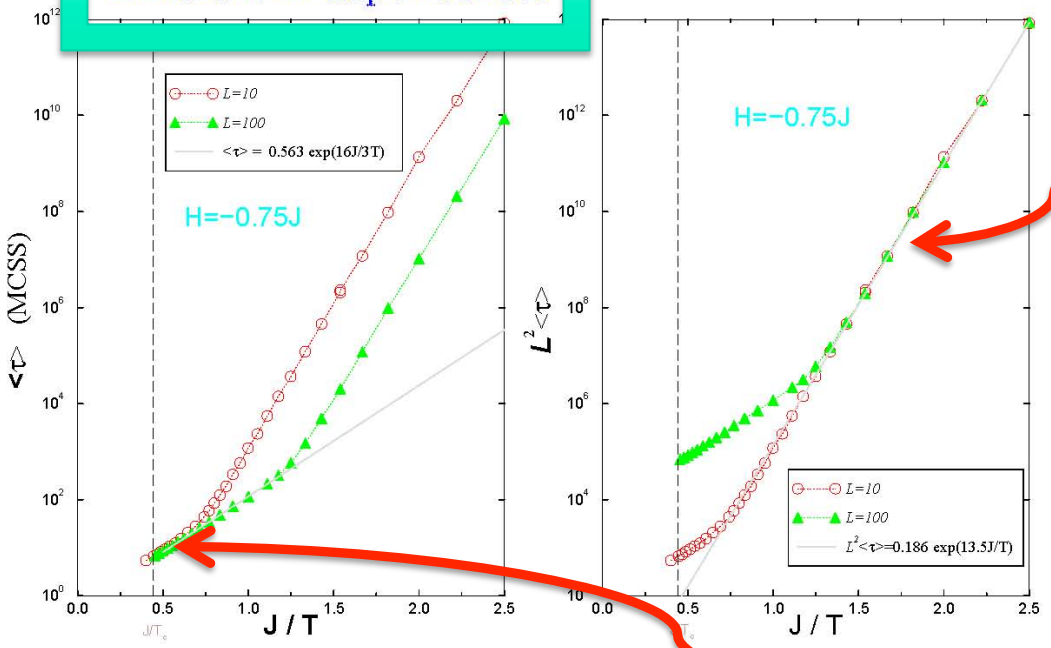
HOMOGENEOUS Nucleation and Growth

d=2 Ising ferromagnet

- T dependence of lifetime

Slopes differ by $\sim \frac{1}{d+1}$

Different L dependences



- 4 length scales
  - a = lattice spacing
  - R_c = critical droplet radius
  - L = linear system size
  - R_0 = typical distance supercritical droplets travel

Same data plotted

SD : Single Droplet

Arrhenius

$\langle\tau\rangle \approx L^d$

$a < R_c < L < R_0$

MD : Multi-Droplet

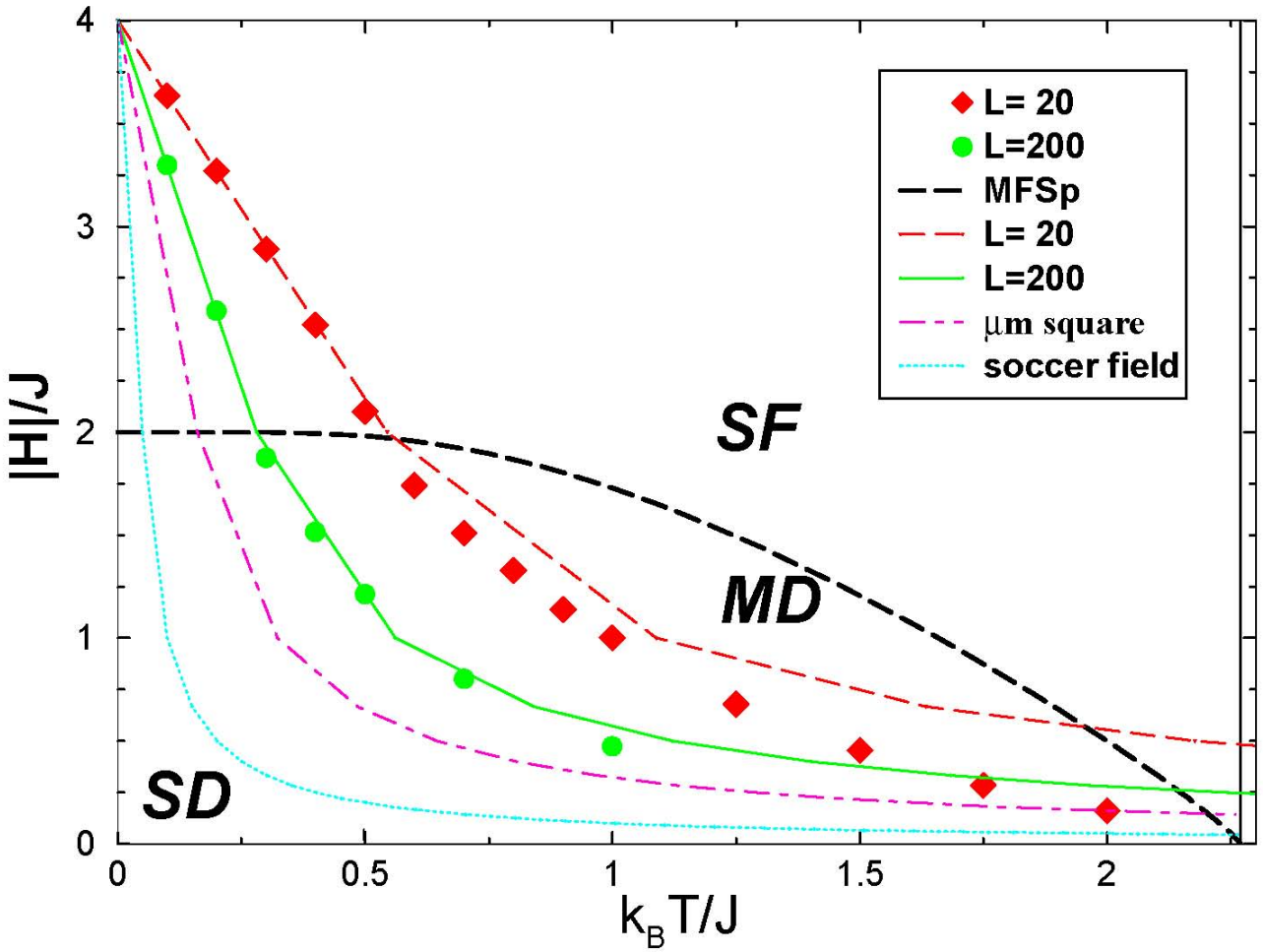
KJMA

$\langle\tau\rangle \approx L^0$

$a < R_c < R_0 < L$

HOMOGENEOUS Nucleation and Growth "Phase Diagram"

d=2 Ising ferromagnet

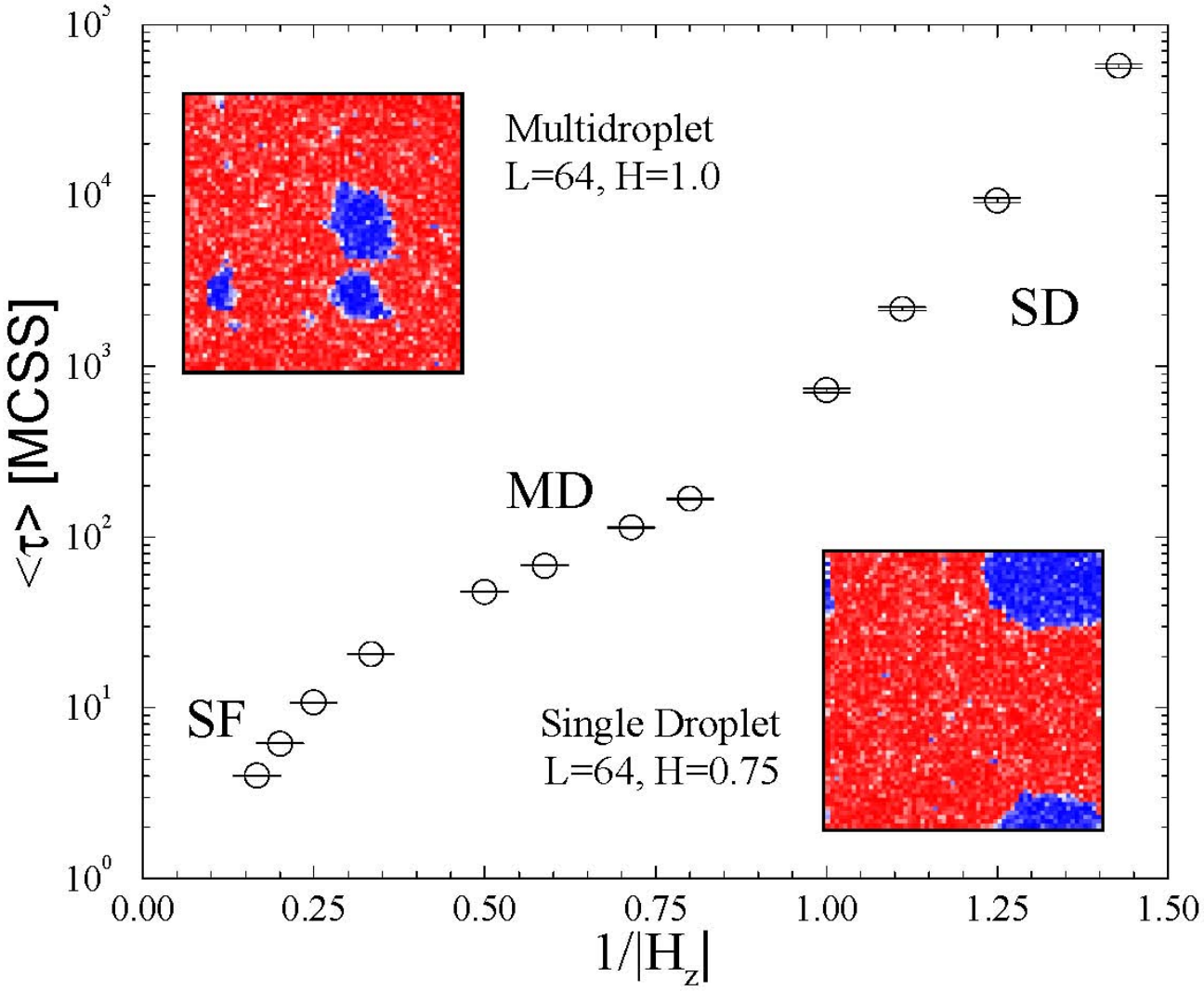


MD : Multi-Droplet
KJMA
$a < R_c < R_0 < L$

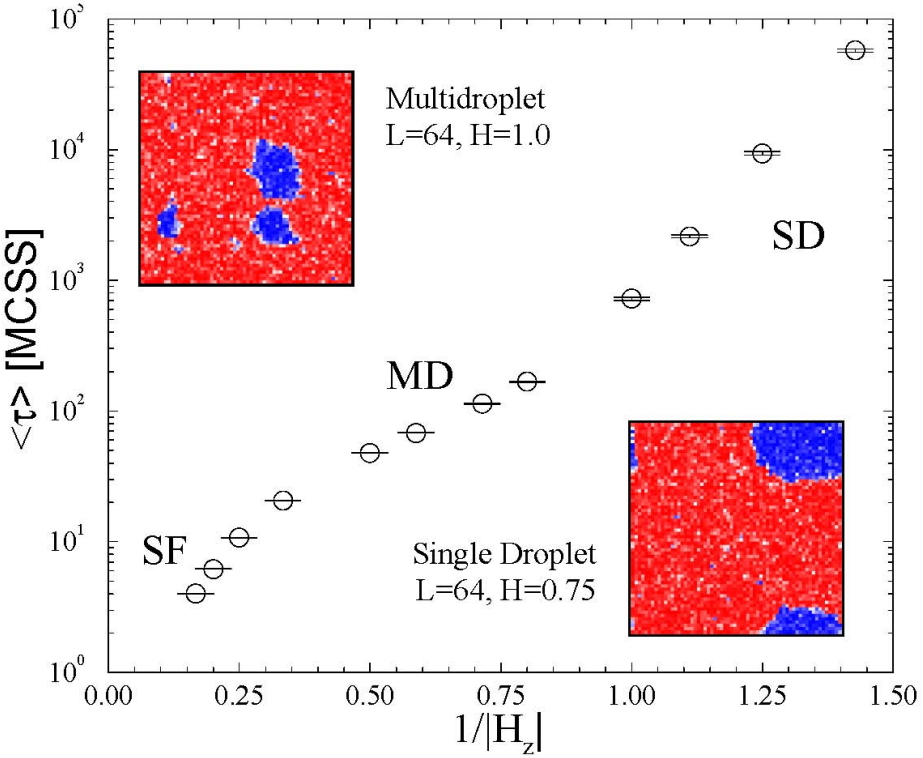
SD : Single Droplet
Arrhenius
$a < R_c < L < R_0$

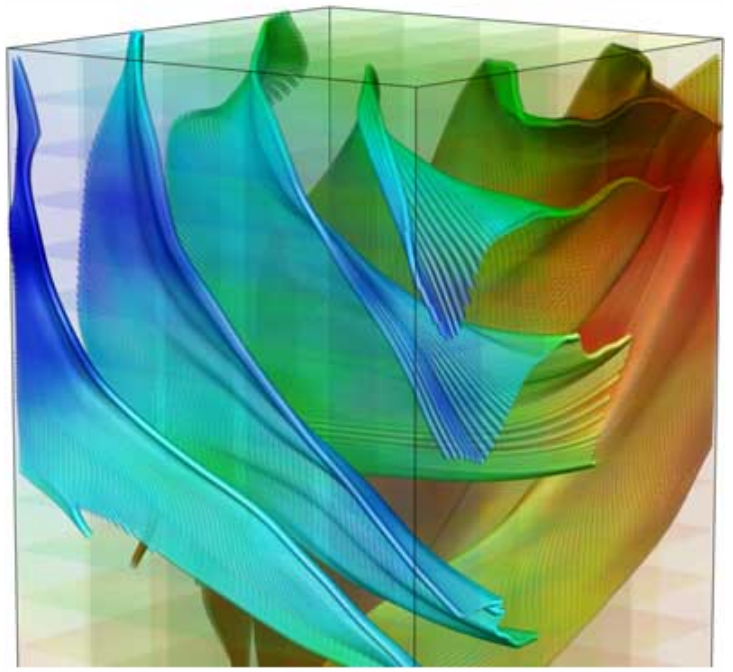
HOMOGENEOUS Nucleation and Growth
For d=2 anisotropic classical Heisenberg model

Also shows different regimes for decay



$J_x=J_y=J_z/2=1$





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HOMOGENEOUS Nucleation and Growth

MCAMC : Monte Carlo with Absorbing Markov Chains

d=2 Ising ferromagnet

PHYSICAL REVIEW
LETTERS

VOLUME 74 | 2 JANUARY 1995 | NUMBER 1

Monte Carlo Algorithms with Absorbing Markov Chains: Fast Local Algorithms for Slow Dynamics

M. A. Novotny
Supercomputer Computations Research Institute, Florida State University, Tallahassee, Florida 32306-4052
(Received 3 June 1994)

A class of Monte Carlo algorithms which incorporate absorbing Markov chains is presented. In a particular limit, the lowest order of these algorithms reduces to the n-fold way algorithm. These algorithms are applied to study the escape from the metastable state in the two-dimensional square-lattice nearest-neighbor Ising ferromagnet in an unfavorable applied field, and the agreement with theoretical predictions is very good. It is demonstrated that the higher-order algorithms can be many orders of magnitude faster than either the traditional Monte Carlo or n-fold way algorithms.

PACS numbers: 02.70.Lq, 05.50.+q, 64.60.My, 75.40.Mg

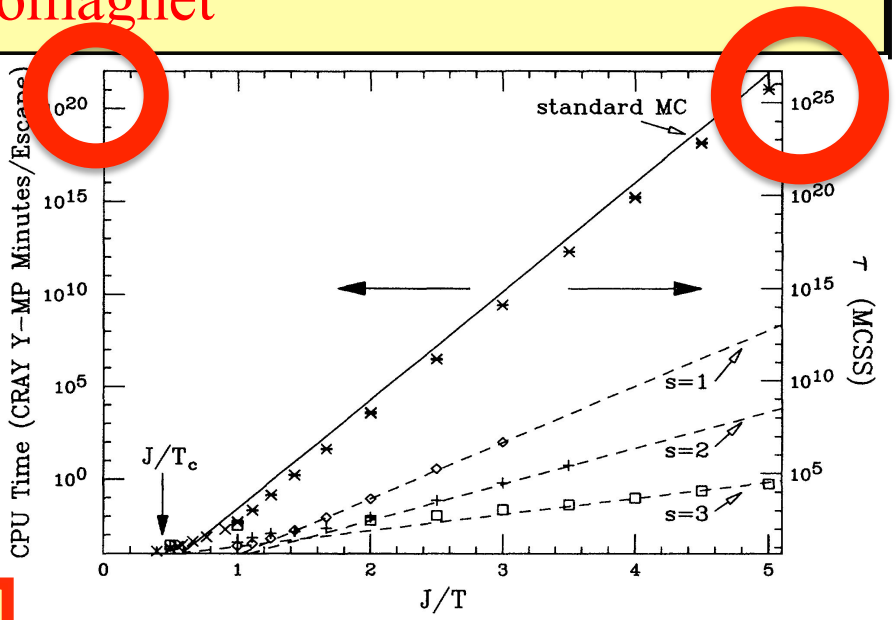
Monte Carlo (MC) methods [
sible tools for nonperturbative
fields, including materials scien
chemistry, biology, engineering
methods are used for two func
poses: to calculate time-indepe
and to simulate time series (dy
case, the slow relaxation observ
sitions (critical slowing down)
is merely a nuisance that has b
ber of new MC algorithms, inc
[2], vertex algorithms [3], multi
and hybrid MC algorithms [5].
many orders of magnitude faster
ods. However, they all replace t
with a different dynamic. Conse
gorithms may be very efficient i
quantities, information about th
MC dynamic cannot be obtain
stances where the kinetics, rather
physical importance. Recently,
strained cluster-flipping algorithms have been proposed in order to obtain information about the long-wavelength kinetics of a system [6]. However, in such methods the local dynamic is modified, and universality arguments must be made to relate the results to the dynamic of the original system.

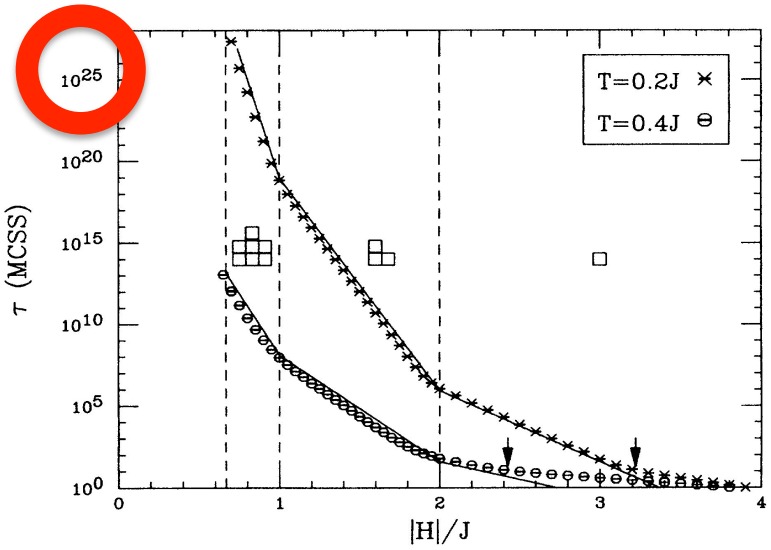
the probability distribution vector $\vec{v}^T$ is then given by $\vec{v}^T(m+1) = \vec{v}^T(m)\mathbf{M}$. An AMC is one in wh
one state has the property that transitions ou
are forbidden.

The Markov matrix associated with
MC with q absorbing states and s transient states is
by [8]

0031-9007/95/74(1)/1(5)$06.00

Spanning Disparate Time Scales





Square Ising Absorbing Markov Chains

q=10 absorbing states; s=1 transient state; N spins

initial vector $\vec{v}_I$; $\vec{e}$ has all elements unity

$$\mathbf{M} = \begin{pmatrix} \mathbf{I}_{q\times q} & \mathbf{0}_{q\times s} \\ \mathbf{R}_{s\times q} & \mathbf{T}_{s\times s} \end{pmatrix}$$

Probability in transient state s after m time steps: $\vec{v}_I^T \mathbf{T}^m \vec{e}$

$$\vec{v}_I^T \mathbf{T}^m \vec{e} < r \leq \vec{v}_I^T \mathbf{T}^{m-1} \vec{e}$$

c_i = # spins in current configuration in class i

$p(i)$ = probability of flipping a spin in class i

Define $Q_j = \sum_{i=1}^{j} c_i p(i)$, $\quad Q_0 = 0$

$$\mathbf{R}_{1\times 10} = \Big(c_1 p(1),\ c_2 p(2),\ \cdots,\ c_{10} p(10) \Big)/N$$

$$\mathbf{T}_{1\times 1} = 1 - Q_{10}/N$$

Time increment m is $m > \ln(r)/\ln(1-Q_{10}/N) > m-1$.

spin in class j is chosen to flip if $\tilde{r}$ satisfies $Q_{j-1} < \tilde{r} Q_{10} < Q_j$

For small Q_{10}/N then $1/\ln(1-Q_{10}/N) \approx -N/Q_{10}$

Δt is a continuous time step; $m \approx \Delta t = -N\ln(r)/Q_{10}$

PD: Geometrical $\rightarrow$ Exponential

Rejection-free

n-fold way

Discrete time step

Rejection-free

n-fold way

Bortz, Kalos, Lebowitz

1975

Higher s MCAMC

q absorbing states; s transient states;

initial vector $\vec{v}_I$; $\vec{e}$ has all elements unity

Novotny *PRL* pg.1, 1995

$$\mathbf{M} = \begin{pmatrix} \mathbf{I}_{q\times q} & \mathbf{0}_{q\times s} \\ \mathbf{R}_{s\times q} & \mathbf{T}_{s\times s} \end{pmatrix}$$

Probability in transient states s after m time steps: $\vec{v}_I^T \mathbf{T}^m \vec{e}$

$$\vec{v}_I^T \mathbf{T}^m \vec{e} < r \leq \vec{v}_I^T \mathbf{T}^{m-1} \vec{e}. \qquad (1)$$

$$\vec{v}_I^T \; \mathbf{T}^{m-1}{}_{s\times s} \, \mathbf{R}_{s\times q} \qquad (2)$$

vector of unnormalized probabilities of exiting to the q states

Eq. (1) and (2) are the **only** equations necessary to utilize in a Monte Carlo simulation Absorbing Markov Chains

s=3 MCAMC

$$\mathbf{T} = \frac{1}{N} \begin{pmatrix} N-2p_7-6p_2-(N-8)p_1 & 2p_7 & 0 \\ 4p_2 & N-p_6-4p_2-(N-5)p_1 & p_6 \\ 0 & Np_1 & N(1-p_1) \end{pmatrix}$$

$$\mathbf{R} = \frac{1}{N} \begin{pmatrix} (N-8)p_1 & 2p_2 & 4p_2 & 0 \\ 0 & 0 & 0 & (N-5)p_1 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

Rejection-free Efficiency for Particles

$$V(r) = \begin{cases} \left(\frac{\sigma}{r}\right)^p - \left(\frac{\sigma}{r_0}\right)^p & r \le r_0 \\ 0 & r \ge r_0 \end{cases} \qquad U_i(\vec{x}) = \sum_{N_{\rm nn}} V(r) = \sum_{k=1}^{N_{\rm nn}} \frac{\sigma^p}{|\vec{x} - \vec{x}_k|^p}$$

$$\langle \exp\left[-\beta \Delta E\right] \rangle = \frac{\Gamma\left(\frac{d}{2}+1\right)}{\pi^{\frac{d}{2}}\, r_{\rm choose}^d} \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} d^d x\, \Theta_{\rm cage}\, \exp\left\{-\beta\left[U_i(\vec{x}) - U_i(\vec{0})\right]\right\}$$

Use Laplace Saddle Point Integration Approximation

$$\langle t_{\rm wait} \rangle \approx \frac{1}{\langle \exp\left[-\beta \Delta E\right] \rangle} \sim \frac{r_{\rm choose}^d\, \rho^{\frac{p+2}{2}}}{T^{\frac{d}{2}}}$$

Marta L. Guerra, M.A. Novotny, Hiroshi Watanabe, and Nobuyasu Ito
Phys Rev E, 2009

Example: PROJECTIVE DYNAMICS
Divide system into two bins

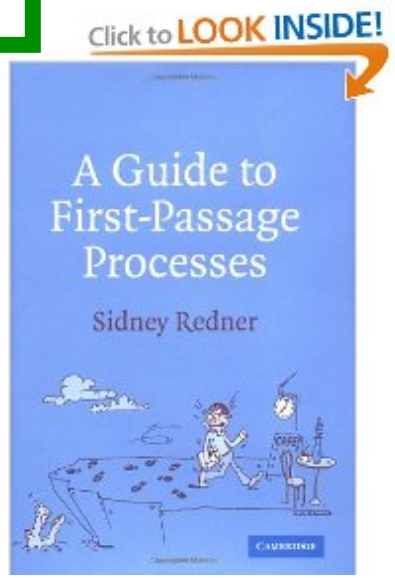
Transient State → **Absorbing State**

Growing rate g

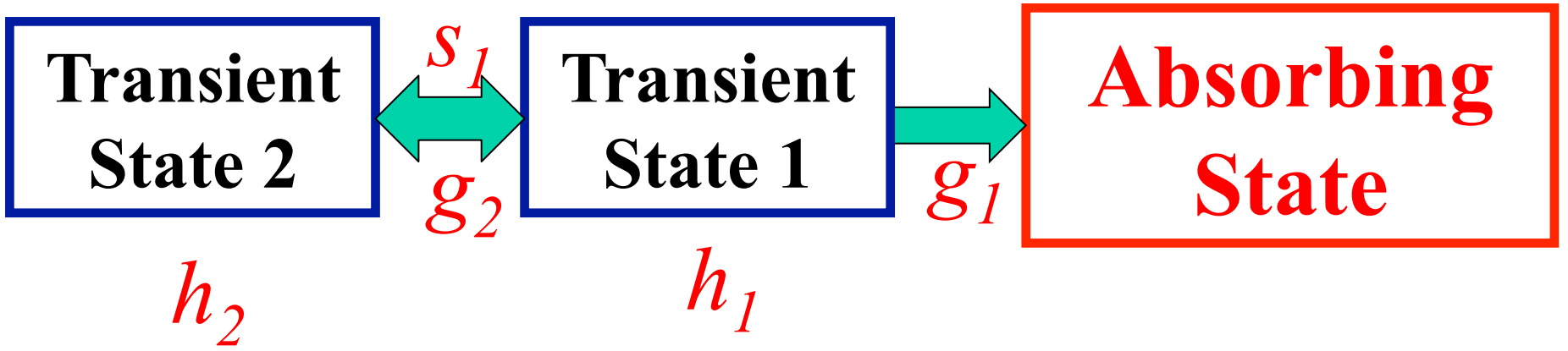
Occupancy time in transient state: h

$$\text{MFPT} = \langle \tau \rangle = 1/g = h$$

***Any* Dynamic**
***Any* System**



Generalize Example: PROJECTIVE DYNAMICS
Divide system into three bins

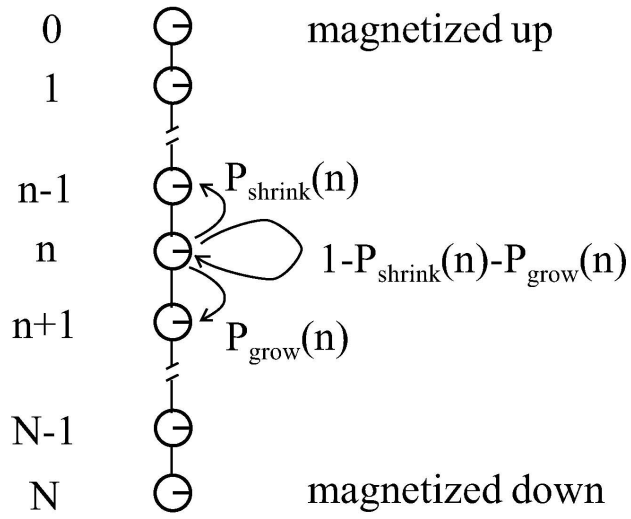


$$\text{MFPT} = \langle\tau\rangle = h_1 + h_2$$

***Any* Dynamic**
***Any* System**

Want h_i the same as for original system

Projection into one dimension



Residence Time

Markov-chain recursion relation

$h(i)=[1+h(i\text{-}1)\,P_{shrink}(i\text{-}1)]/P_{grow}(i)$

$h(1)=1/P_{grow}(1)$

Assume Boltzmann weight

$h(i) \propto \exp(-F(i)/k_B T)$

Free-energy barrier

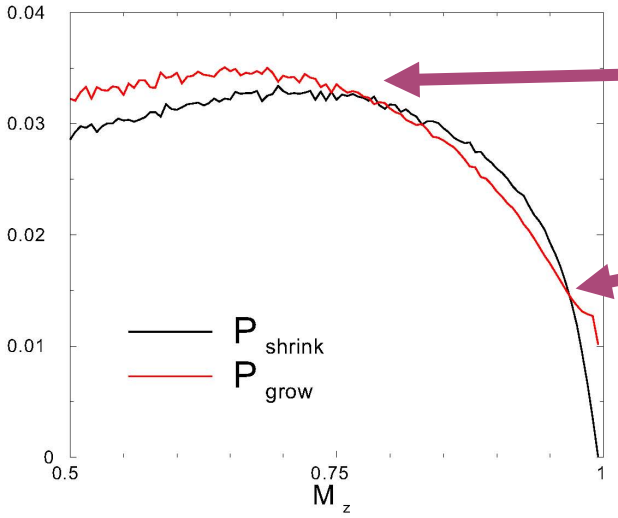
$\Delta F = k_B T \ln[\, h(i_m)/h(i_s)\,]$

i_m = bin of metastable configuration

i_s = bin of saddle point configuration

Find the saddle point

Crossing of growing and shrinking probabilities



H=1000 Oe

T= 50 K

Projective Dynamics

Saddle point

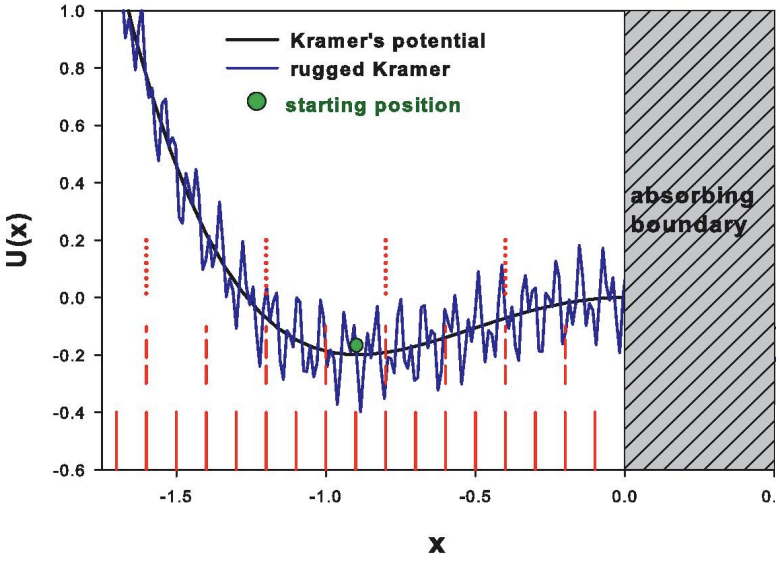
Metastable minimum

VOLUME 80, NUMBER 15 PHYSICAL REVIEW LETTERS 13 APRIL 1998

Projection Method for Statics and Dynamics of Lattice Spin Systems

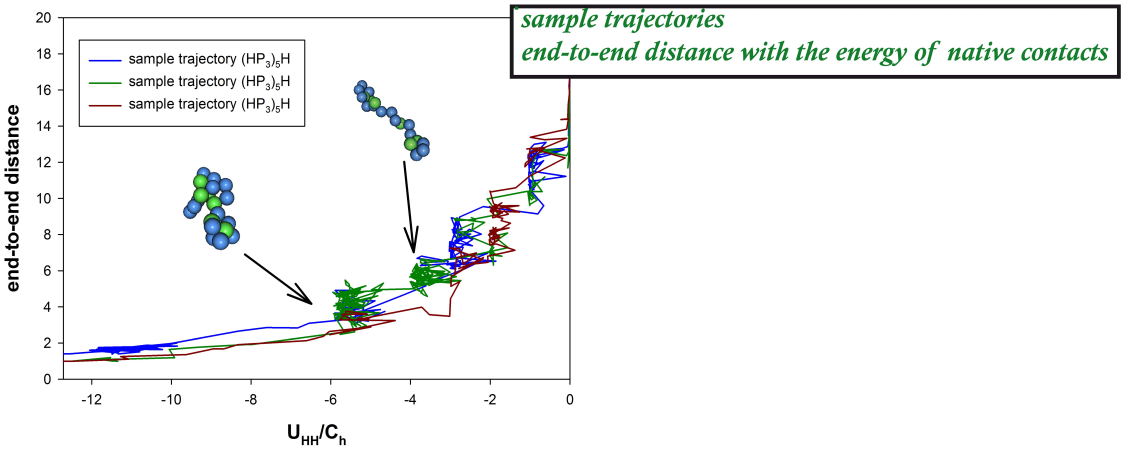
M. Kolesik,[1,2] M. A. Novotny,[1] and P. A. Rikvold[1,3]

Projective Dynamics:
Does the 'binning' matter?
For continuous degrees of freedom?



Katja Biswas (née Schäfer)

J. Phys. A: Math. Theor. **44** 345004 2011



Projective Dynamics: Main Theorem

Given:

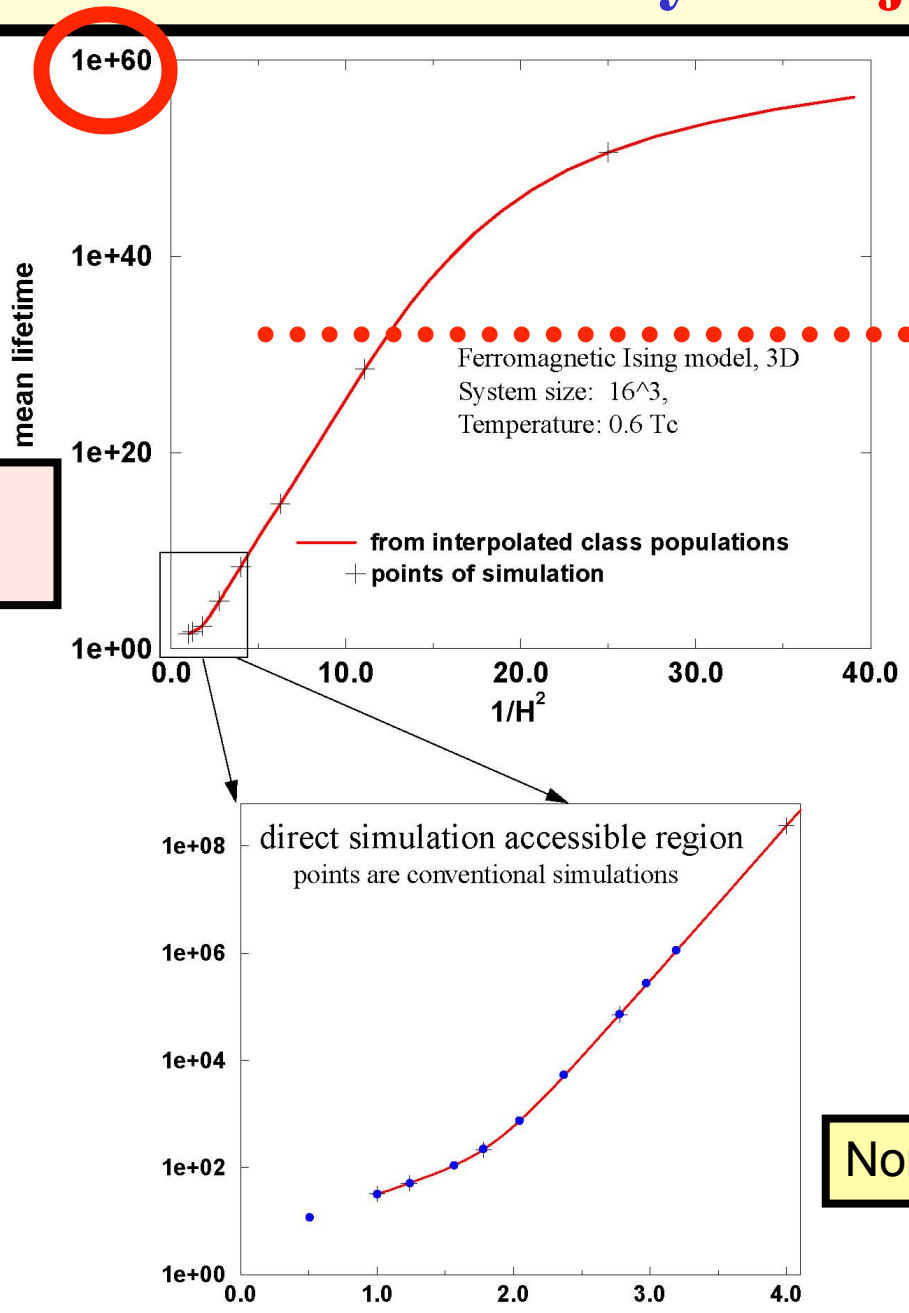
- Absorbed from only one bin
- Time *dt* such that only $i \rightarrow i+1$ or $i \rightarrow i-1$ or $i \rightarrow I$
  - *Generalized in second paper (in prep, PhD dissertation)*
- Binning constant in time
- Growing and shrinking rates those of system

Then:

- Same MFPT as original system
- Same h_i as original system
  - *Not same $h_i(t)$*

Biswas (Schäfer) and Novotny; J Phys A: Math Theor 2011

Field Reversal Metastability: Projective Dynamics



Faster than real time simulation

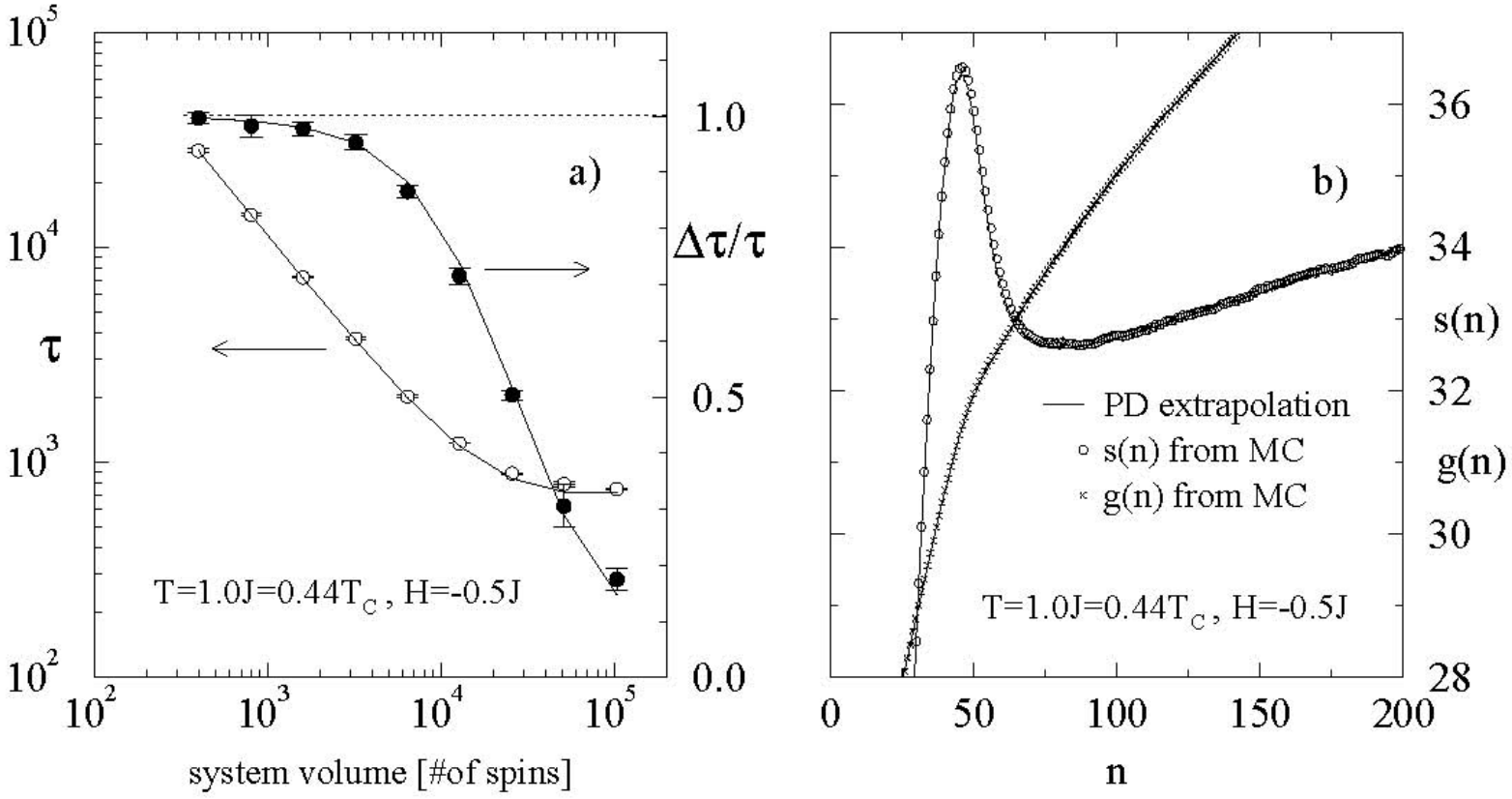
Age of universe in femtoseconds

Normal MC simulation

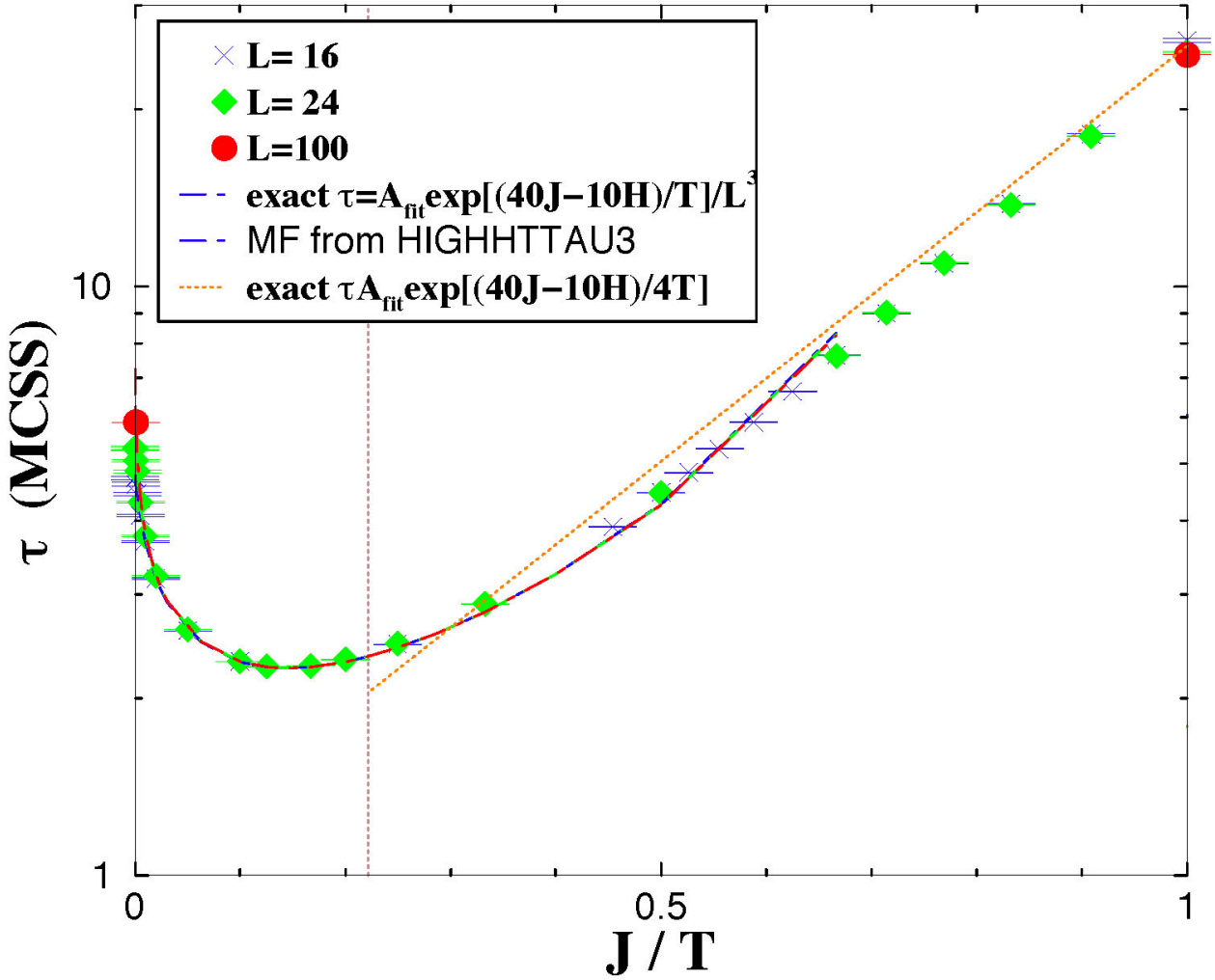
Projective Dynamics – to large L – Ising

Assuming two 'independent' systems

$$g(2V,n) \approx \frac{\sum_{i=0}^{n} h(V,n-i)h(V,i)\,[g(V,n-i)+g(V,i)]}{\sum_{i=0}^{n} h(V,n-i)h(V,i)}$$



MFPT: 3d Ising
H=2.7
Projective Dynamics: no simulations



Nucleation and Growth

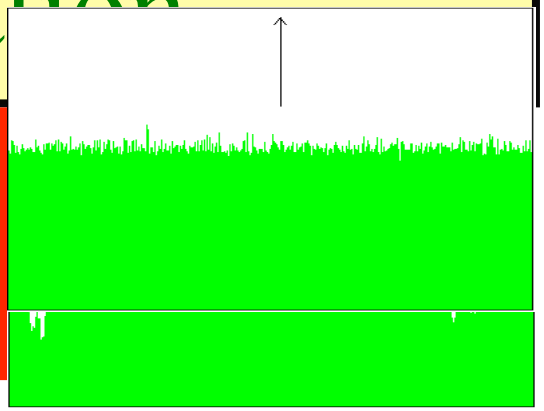
Monte Carlo (entire class of similar algorithms)

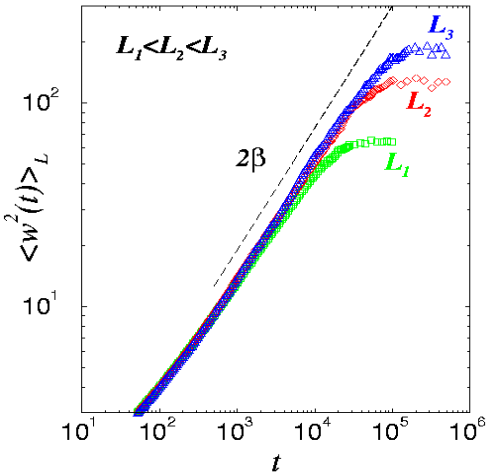
Non-trivial parallelization

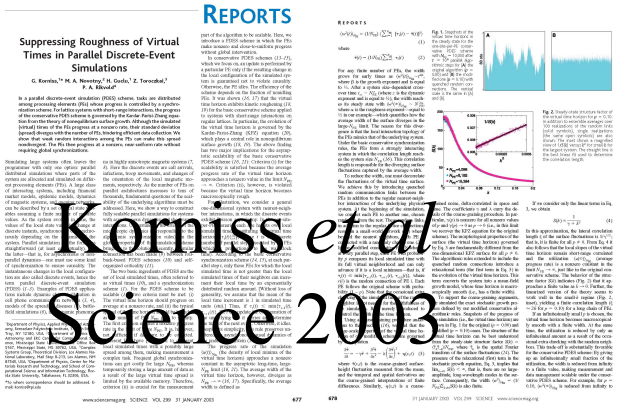
VOLUME 84, NUMBER 6 PHYSICAL REVIEW LETTERS 7 FEBRUARY 2000

From Massively Parallel Algorithms and Fluctuating Time Horizons to Nonequilibrium Surface Growth

G. Korniss,[1] Z. Toroczkai,[2,3] M. A. Novotny,[1] and P. A. Rikvold[1,4]



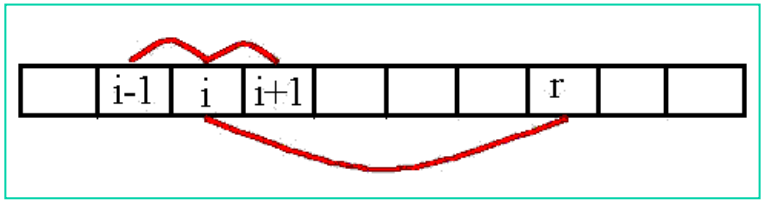


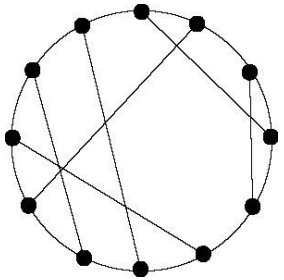


Korniss *et al* Science 2003

$N \to \infty$

$w \sim const.$ PE number





All short-ranged discrete event simulations can be made PERFECTLY SCALABLE

(patented architecture)

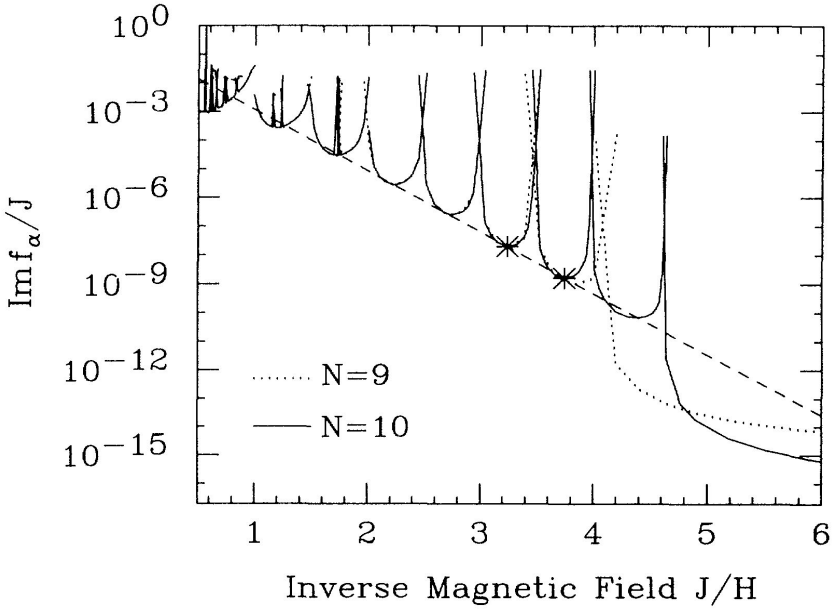
HOMOGENEOUS Nucleation and Growth
Imaginary part of Free Energy
d=2 Ising ferromagnet

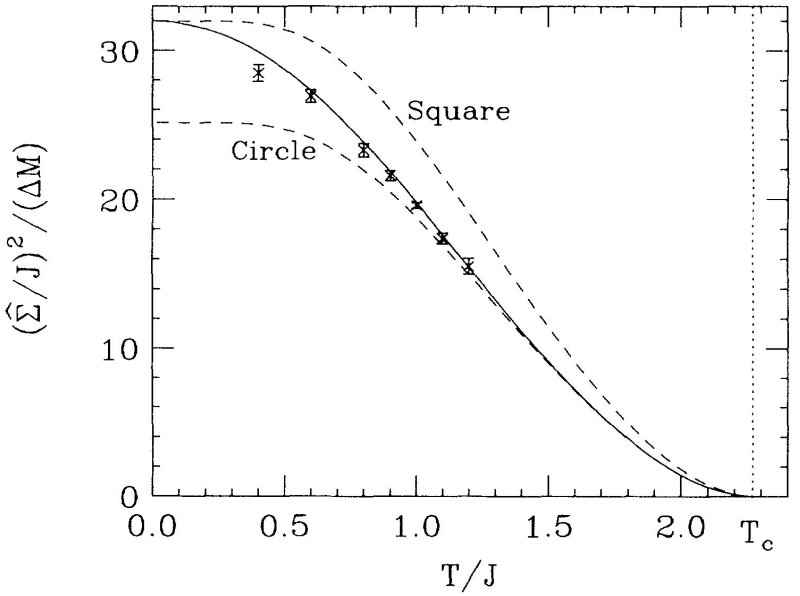
VOLUME 71, NUMBER 24 PHYSICAL REVIEW LETTERS 13 DECEMBER 1993

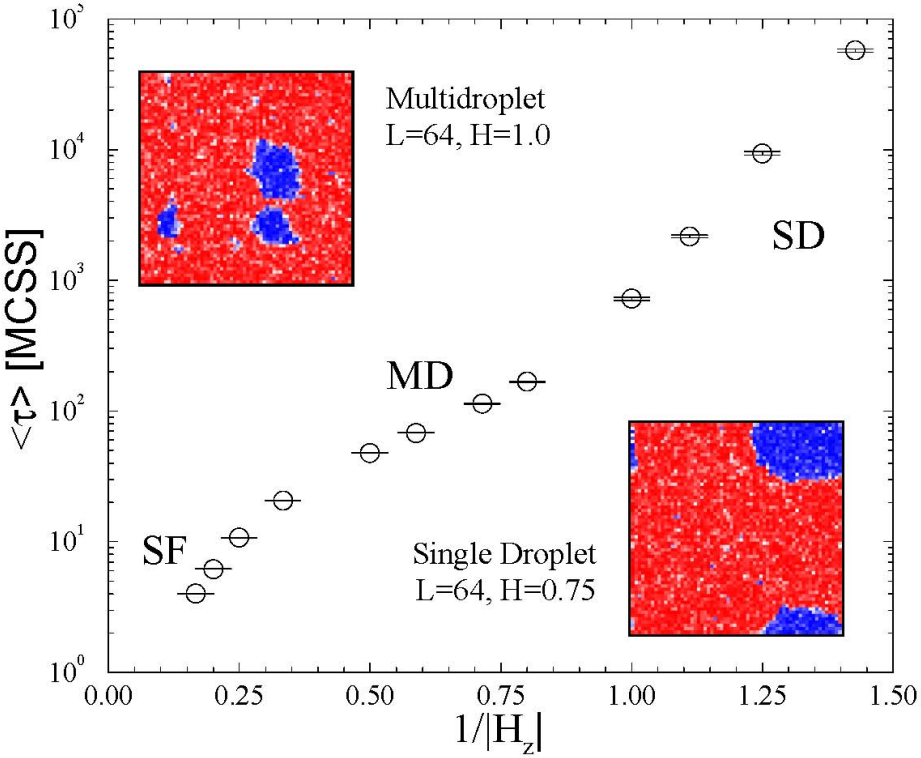
Numerical Transfer-Matrix Study of Metastability in the $d=2$ Ising Model

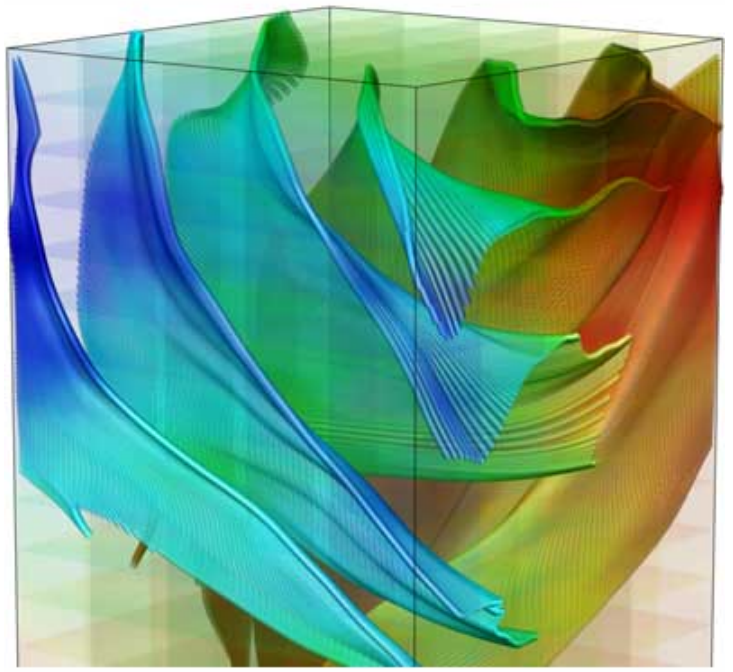
C. C. A. Günther,[1,2] Per Arne Rikvold,[1,2,3] and M. A. Novotny[1]

$$\mathrm{Im} f_{\mathrm{ms}} = B|H|^{b} \exp\left(-\frac{1}{T}\,\frac{\hat{\Sigma}^{2}}{4|H|\Delta M}\,[1+O(H^{2})]\right)$$









$10 \times 10 \times 150\ nm^3$

HOMOGENEOUS NUCLEATION / GROWTH / METASTABILITY

- Homogeneous nucleation and growth is complicated!
- Algorithms can be devised to efficiently study nucleation & growth
- Can changing dynamic change nucleation rate exponential?
- How does hysteresis loop area depend on the f for oscillating field?
- What is a dynamic phase transition? Experimental realization?

What dynamic to use?

Dynamic Quantum Model → Dynamic MC

Hamiltonian: $\mathcal{H} = \mathcal{H}_{\rm sp} + \mathcal{H}_{\rm ph} + \mathcal{H}_{\rm sp-ph}$

$$\mathcal{H}_{\rm sp} = -J \sum_{<i,j>} \sigma_i^z \sigma_j^z - H_z \sum_i \sigma_i^z$$

$$\mathcal{H}_{\rm ph} = \sum_q \hbar\omega_q c_q^\dagger c_q$$

$$\mathcal{H}_{\rm sp-ph} = \lambda \sum_{j=1}^{N_s} \sum_q \sqrt{\frac{\hbar}{2NM\omega_q}} (iq\sigma_j^x)(c_q^\dagger - c_q) \times \exp\left(i\vec{q}\cdot\vec{R}_j\right)$$

local strain $\varepsilon(0) = \frac{1}{\sqrt{N}} \sum_{\vec{q}} i\vec{q}\cdot\vec{u}_{\vec{q}}$

$\mathcal{H}_{\rm sp-ph} \sim \sigma^x \varepsilon(0)$

Generalized Master Equation:

$$\frac{d\rho(t)_{m'm}}{dt} = \frac{i}{\hbar}[\rho(t), \mathcal{H}_{\rm sp}]_{m'm} - \gamma_{m'm}\rho(t)_{m'm} + \delta_{m'm} \sum_{n\neq m} \rho(t)_{nn} W_{mn}$$

$$\gamma_{m'm} = \frac{W_m + W_{m'}}{2}, \quad W_m = \sum_{k\neq m} W_{km}$$

Dynamic Quantum Model → Dynamic MC

Integrate out heat bath ···

Transition Rate from l to k-th eigenstate of $\mathcal{H}_{\rm sp}$:

$$W_{k,l} = \frac{2\pi}{\hbar} \sum_{j,q,n_q} |\langle n_q+1, k|\mathcal{H}_{\rm sp-ph}|n_q, l\rangle|^2 \langle n_q|\rho_{\rm ph}|n_q\rangle \delta(\hbar\omega_q - (E_l - E_k))$$

when $E_l > E_k$

With 2D phonon heat bath:

$$W_{k,l} = \frac{\lambda^2}{2\rho\hbar^3 c^4} \left| \frac{(E_l - E_k)^2}{1 - e^{-\beta(E_l - E_k)}} \right|$$

λ: spin-phonon coupling constant

ρ: mass density of the unit cell

c: sound velocity

E_k: k-th energy eigenvalue of the spin system

Satisfies detailed balance.

General case derived using spin-lattice technique

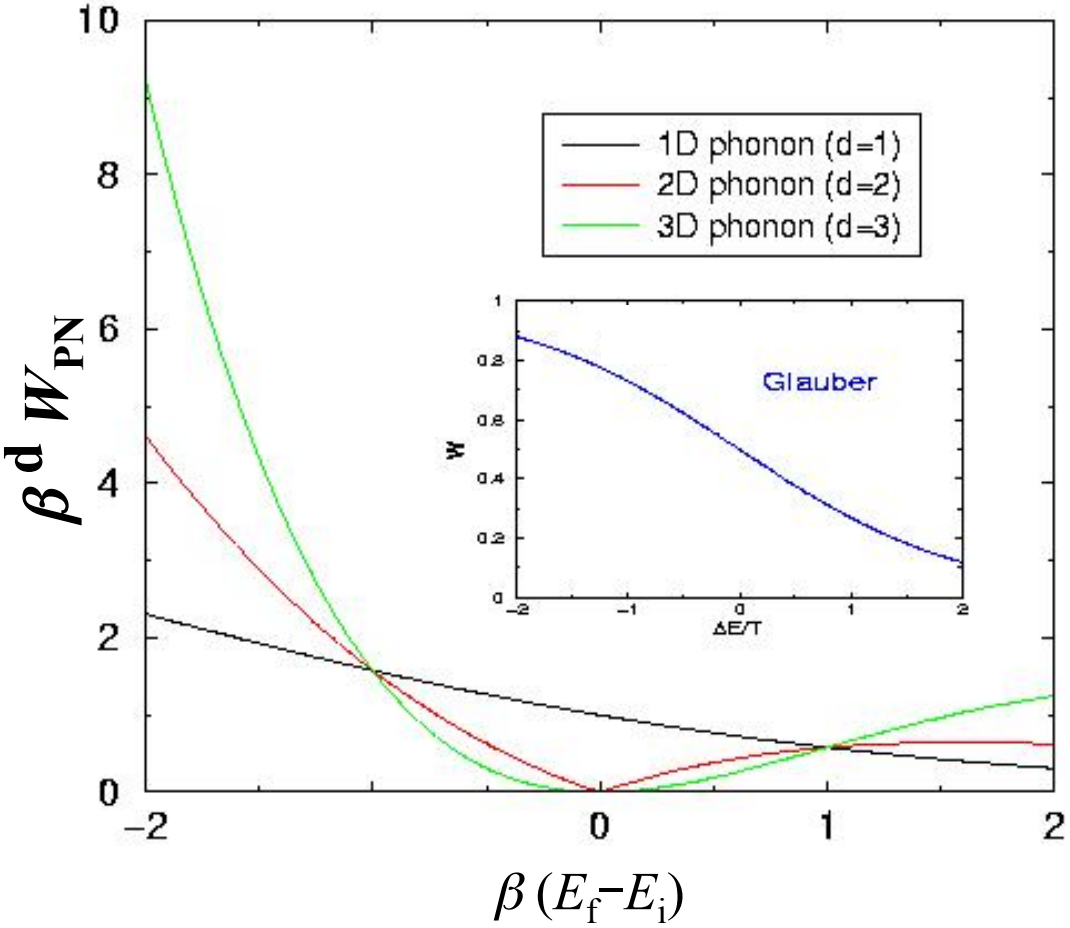
L or *J* conservation

Phonon dynamic for spins

$$W_{\mathrm{PN}}(\mathrm{i}\rightarrow\mathrm{f}) = \nu\left|\frac{(E_{\mathrm{f}}-E_{\mathrm{i}})^{d}}{\exp\left[\beta(E_{\mathrm{f}}-E_{\mathrm{i}})\right]-1}\right|$$

Found by integrating out bath degrees of freedom in quantum density matrix.

W_{PN} vanishes for
$E_{\mathrm{f}}-E_{\mathrm{i}} = 0$
if *d=2 or 3*



K. Park, M.A. Novotny, P.A. Rikvold, Phys. Rev. E **66**, 056101 (2002).

Does the dynamic matter?

Γ is ***not*** universal

depends on dynamic for all *H* at low *T*

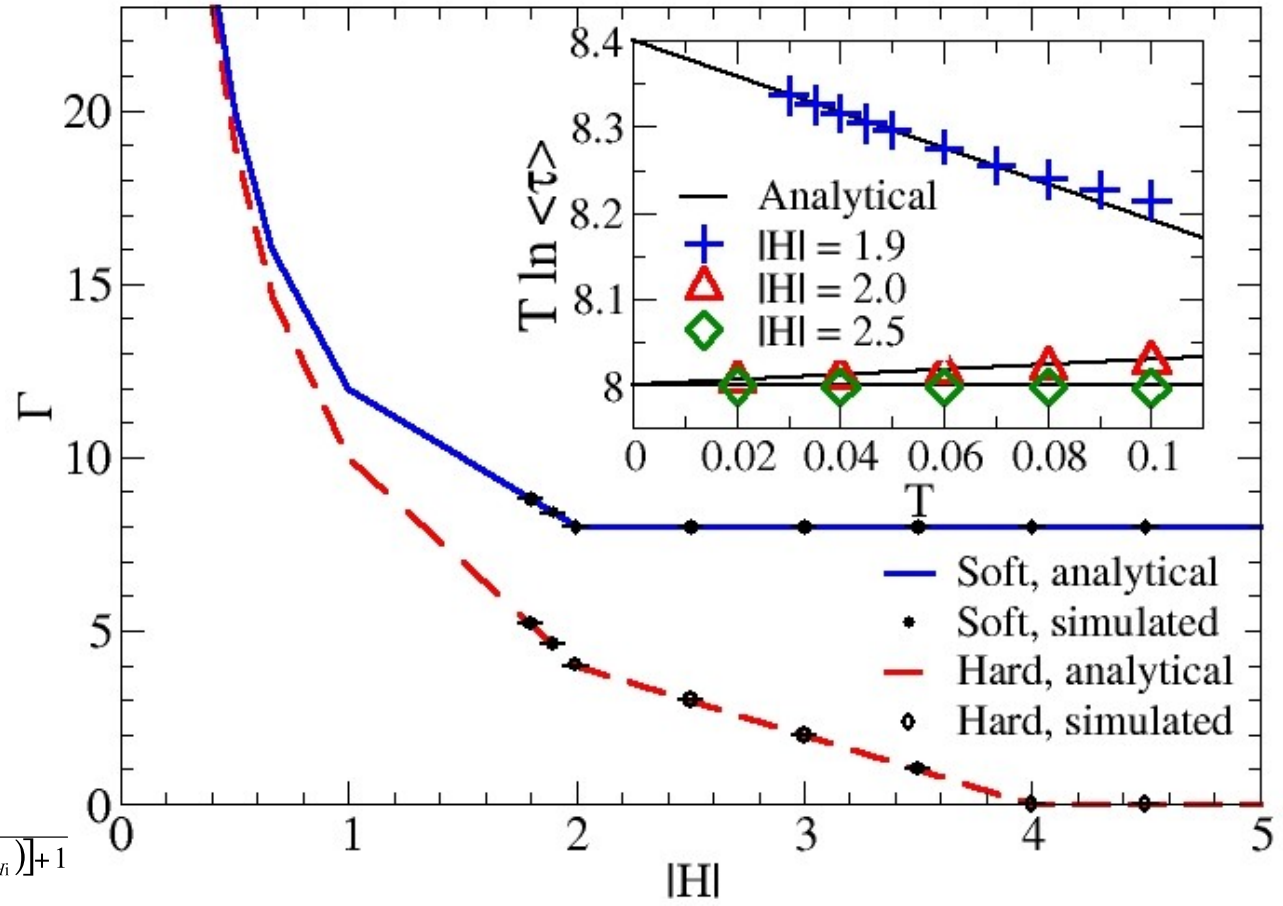
$< \tau > = \tilde{A} \exp(\beta\Gamma)$

$\beta = 1/k_B T$

Compare:

Hard Glauber

Soft Glauber



$$W_{SG}(i \to f) = \nu \frac{1}{\exp[\beta(E_{Jf} - E_{Ji})]+1} \times \frac{1}{\exp[\beta(E_{Hf} - E_{Hi})]+1}$$

K. Park, et al., Phys. Rev. Lett. **92**, 015701 (2004).

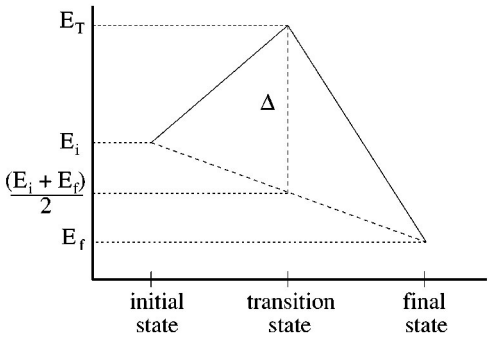
Does the dynamic matter?

Γ is ***not*** universal depends on dynamic for all H at low T !

$$< \tau > = \tilde{A} \exp(\beta\Gamma)$$

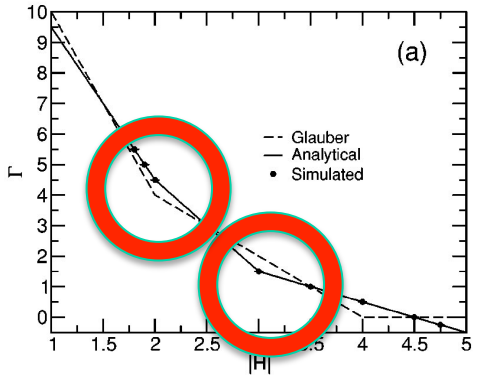
$$\beta = 1/k_B T$$

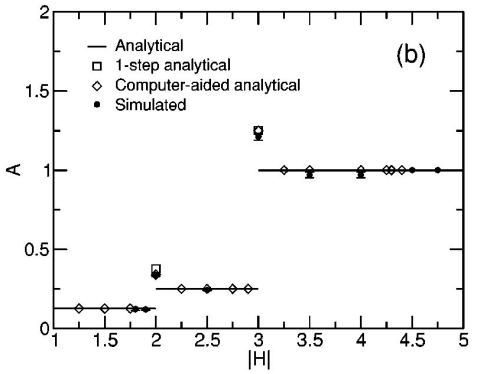
Compare:
Hard Glauber
TDA, OSD



Transition Dynamic Approximation

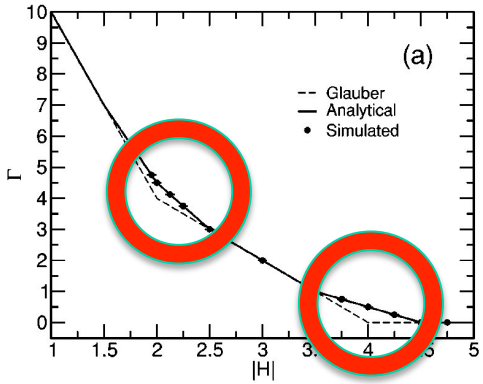
$$W_{OSD} = \exp[-\beta(E_T - E_i)]$$
$$= \exp(-\beta\Delta)\exp\left(-\beta\frac{E_f - E_i}{2}\right)$$

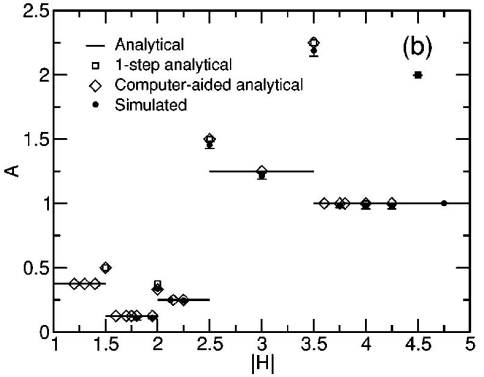




One-Step Dynamic

$$W_{TDA} = \frac{1}{1+\exp[\beta(E_T - E_i)]}\frac{1}{1+\exp[\beta(E_f - E_T)]}$$



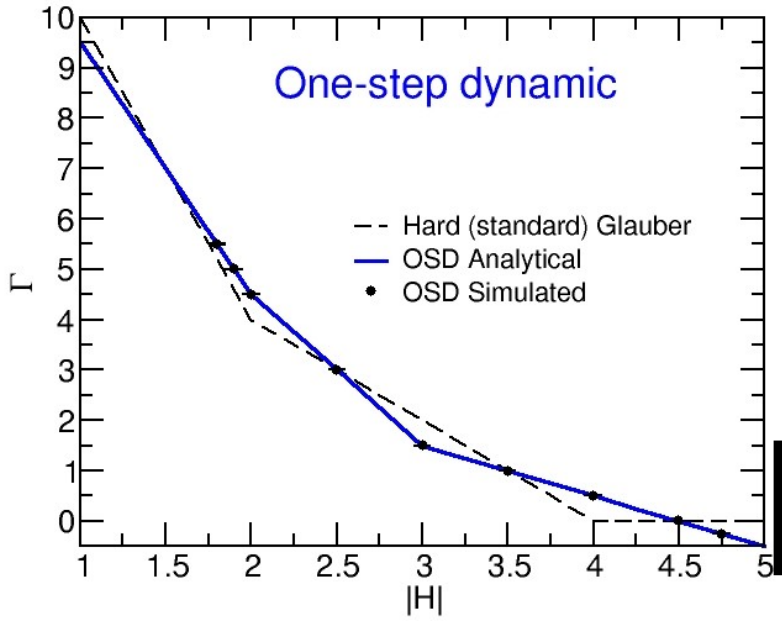


G. Buendia, et al., J. Chem. Phys. **121**, 4193 (2004).

Does the dynamic matter?

Why does dynamic matter so much?

At low enough T
Nucleating Droplet = Transition State
TPT probability of reactive trajectory at nucleating droplet = 0

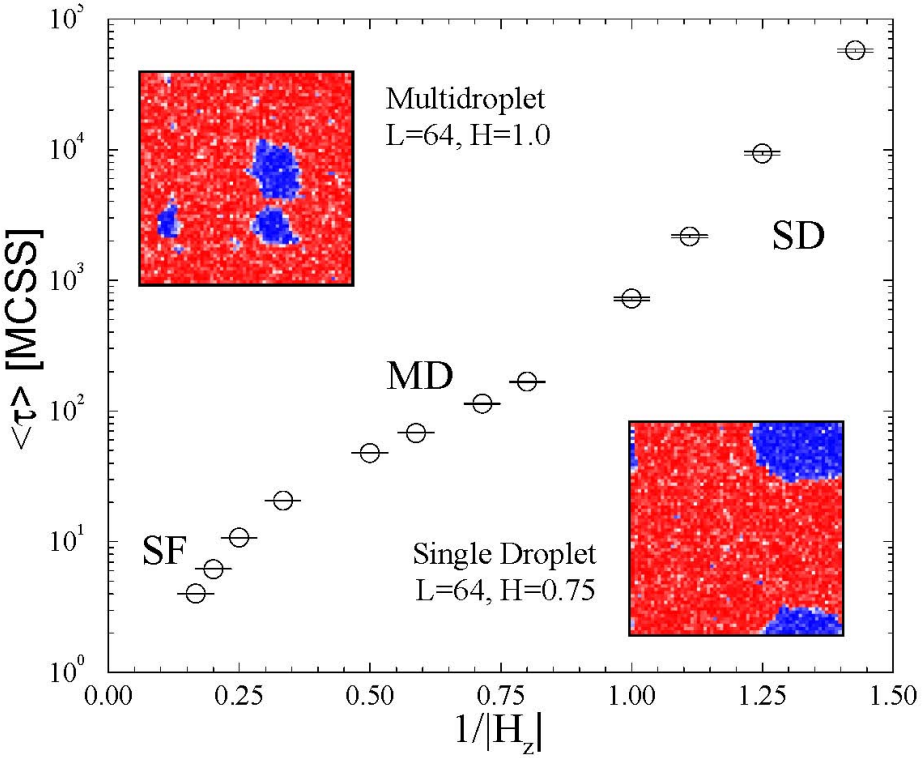


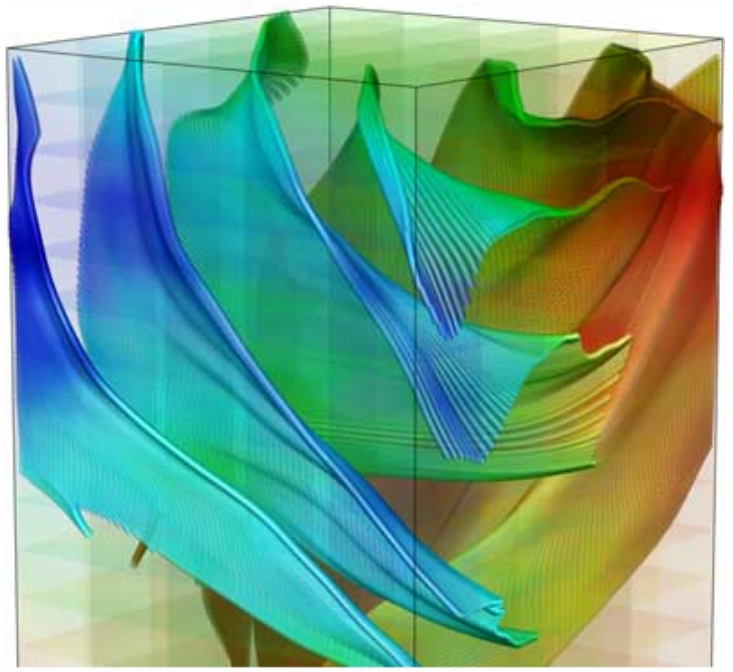
Transition Tube

Transition State

G.M. Buendía, et al., *J. Chem. Phys. 2005*

Same states, energies, saddle





10 x 10 x 150 nm^3

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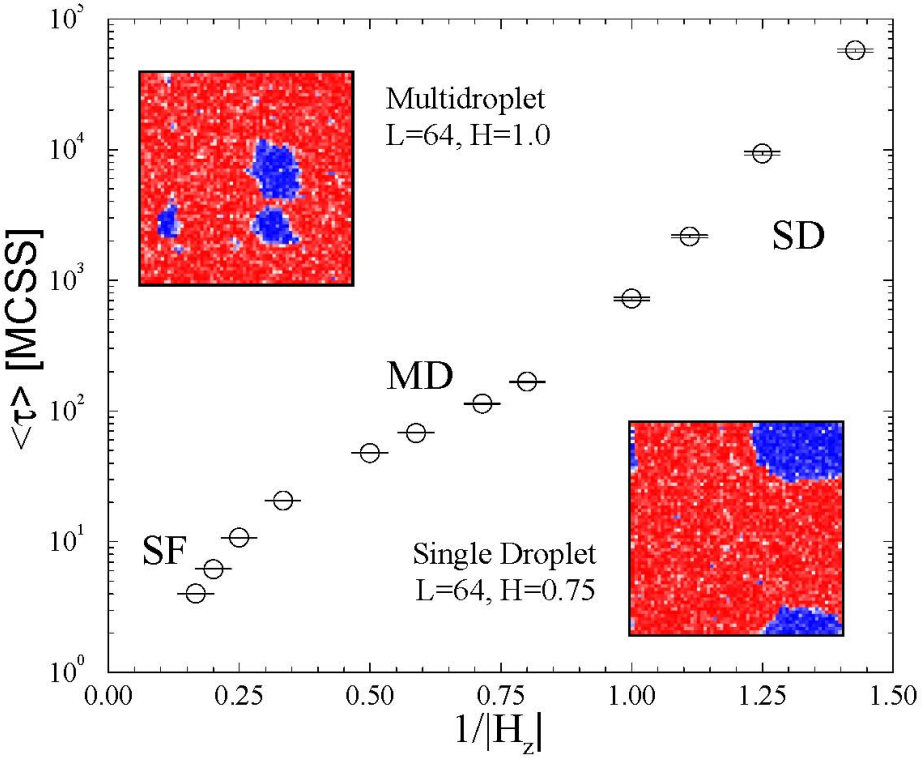
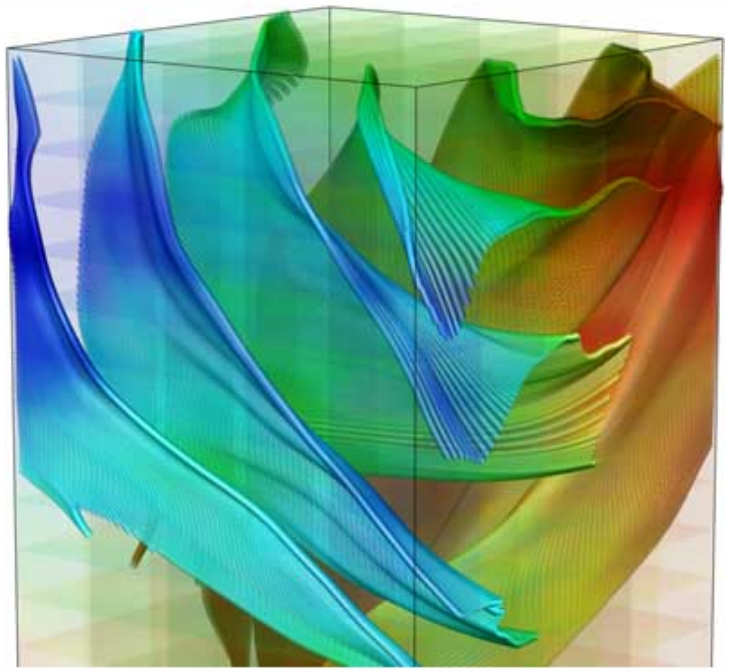
Surprises, Computational Methods, and Results for Metastable Phenomena and Homogeneous Nucleation and Growth

Mark A. Novotny

Dept. of Physics and Astronomy
HPC^2 Center for Computational Sciences (CCS)
Mississippi State University

Part 2: Oscillating Fields: Hysteresis; Dynamic Phase Transition





10 x 10 x 150 nm^3

HOMOGENEOUS NUCLEATION / GROWTH / METASTABILITY

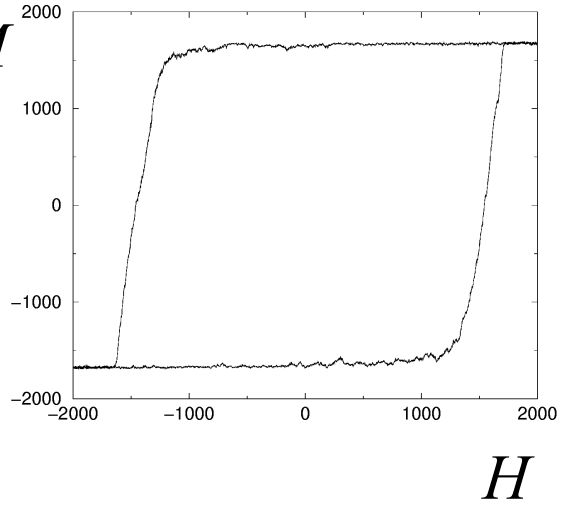
- Homogeneous nucleation and growth is complicated!
- Algorithms can be devised to efficiently study nucleation & growth
- Can changing dynamic change nucleation rate exponential?
- How does hysteresis loop area depend on the f for oscillating field?
- What is a dynamic phase transition? Experimental realization?

Hysteresis

Average loop areas

Apply oscillating magnetic field:

$$H(t) = H_0 \sin(\omega t)$$



- Loop area $A = -\oint m \, \mathrm{d}H$ important
  - Energy dissipation per cycle
  - Electric transformers and motors, ...
  - Engineering interest for more than a century: Ewing 1881; Warburg 1881; Steinmetz 1892
  - Frequency dependence: $A \propto H_0^a \omega^b$??
  - Frequency dependence: $A = A_0 + const.[\omega^2(H_0^2 - H_{\mathrm{sp}}^2)]^{1/3}$??
  - Frequency dependence: $A \propto -\ln(H_0 \omega)$??

Average loop area, $\langle A \rangle$ *d=2* Ising PRE 1998

$$I\left(H(t),T\right) \approx B(T)\left|H(t)\right|^{K} \exp\left[-\frac{\Xi_0(T)}{\left|H(t)\right|^{d-1}}\right]$$

$$\begin{aligned} m(t) &= 2\exp\left[-\Phi(t)\right] - 1 \\ &= 2\exp\left[-\int_0^t I\Omega_d (v_0 t')^d dt'\right] - 1 \\ &= 2\exp\left[-\frac{\Omega_d v_0^d I}{d+1} t^{d+1}\right] - 1 \,, \end{aligned}$$

KJMA

$$\langle \tau \rangle = \left[\frac{\Omega_d v_0^d I}{\ln 2 (d+1)}\right]^{-\frac{1}{d+1}}$$

$$R = \frac{(2\pi/\omega)}{\langle \tau(H_0) \rangle}$$

Lifshitz-Allen-Kahn

$$v(t) \approx \nu\left|H(t)\right|$$

$$\ln 2 = \frac{B(T)\Omega_2 \nu^2}{4H_0^3\omega^3}\left\{H_s^4 \Xi_0^4(T)\Gamma\left[-4,-\frac{\Xi_0(T)}{H_s}\right] - 2H_s^2\Xi_0^6(T)\Gamma\left[-6,-\frac{\Xi_0(T)}{H_s}\right] + \Xi_0^8(T)\Gamma\left[-8,-\frac{\Xi_0(T)}{H_s}\right]\right\},$$

$$\int_0^x u^n e^{-a/u} du = a^{n+1}\Gamma\left[-(1+n),\frac{a}{x}\right]$$

$$\Gamma(a,x) \sim x^{a-1}e^{-x}\left[1 + \frac{a-1}{x} + \frac{(a-1)(a-2)}{x^2} + \ldots\right]$$

$$\left(\frac{H_s}{\Xi_0(T)}\right)^{1} \exp\left[-\frac{H_s}{\Xi_0(T)}\right] \approx (DH_0\omega)^3$$

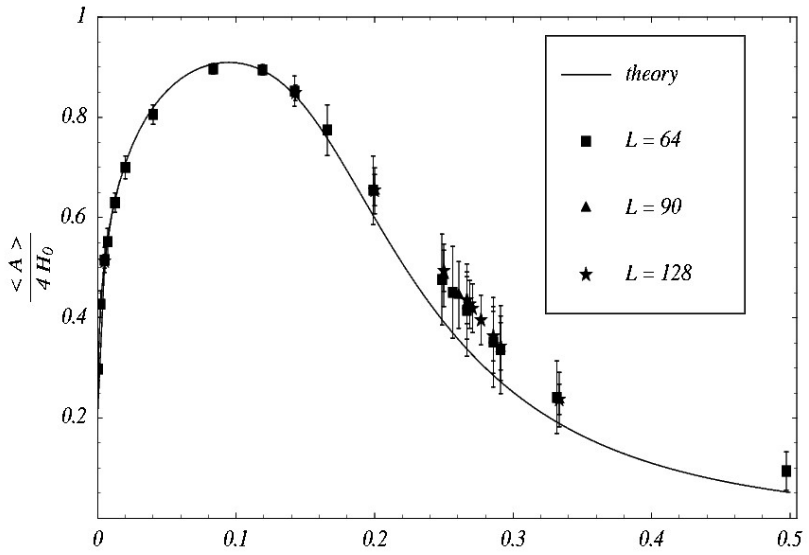
$$D = \left(\frac{\ln 2}{2B(T)\Omega_2\nu^2\Xi_0^8(T)}\right)^{\frac{1}{3}}$$

$$\langle A \rangle_{\mathrm{LF}} \approx \frac{4}{3}\Xi_0(T)\left[-\ln(DH_0\omega)\right]^{-1}$$

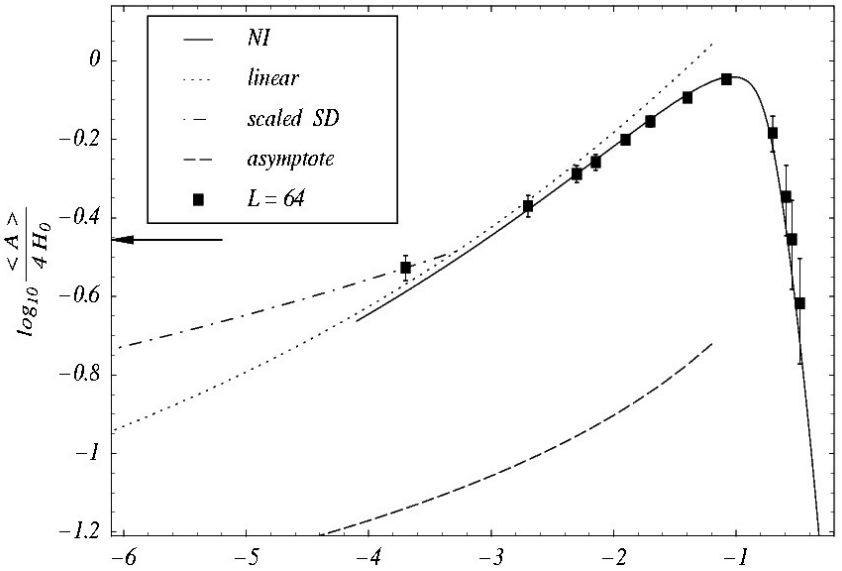
Average loop area, <*A*> *d*=2 Ising

$$T = 0.8T_c\ ,\ H_0 = 0.3J$$

vs *Frequency*Lifetime* vs $\log_{10}$[*Frequency*Lifetime*]



*Frequency*Lifetime*

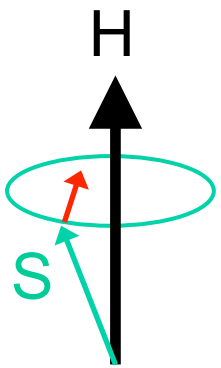


$\log_{10}$[*Frequency*Lifetime*]

Slow crossover to $A = [-\log(const.\ H_0\ \omega)]^{-1/(d-1)}$!!

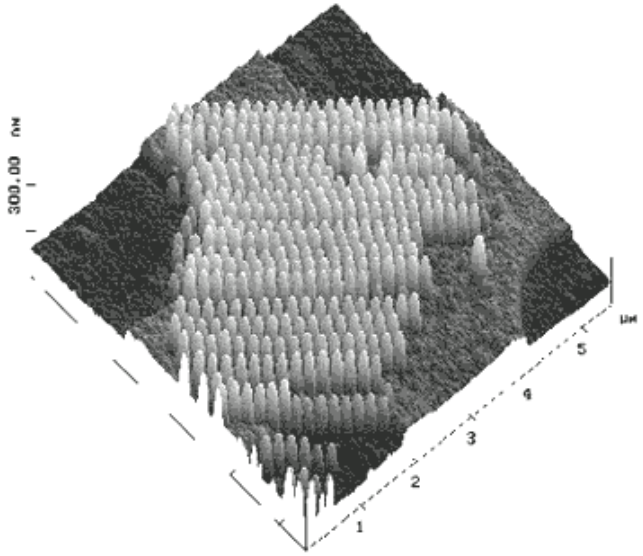
$$R = \frac{(2\pi/\omega)}{\langle \tau(H_0) \rangle}$$

Finite-temperature Micromagnetics



$$\frac{dS}{dt} = -\frac{\gamma_0}{1+\alpha^2} S \times \left\{ H - \alpha \left(S \times H \right) \right\}$$

$$H_i = H_{ex,i} + H_{dd,i} + H_{zm,i} + H_{th,i}$$



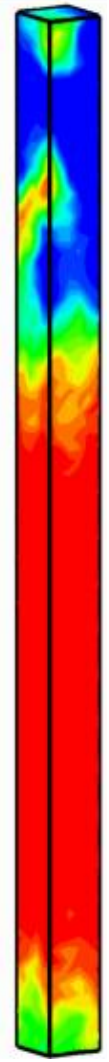
Parameters match those of bulk iron

M_s = 1700 emu/cm^3

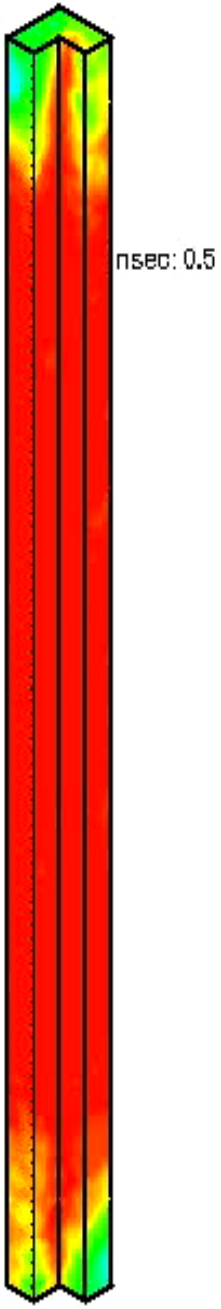
l_{ex} = 3.6 nm

dt = 0.85 fs

α=0.1



Simulated Nanomagnets



M_z

9 nm x 9 nm x 150 nm

Fe particle

H_0=800 Oe, T=20 K

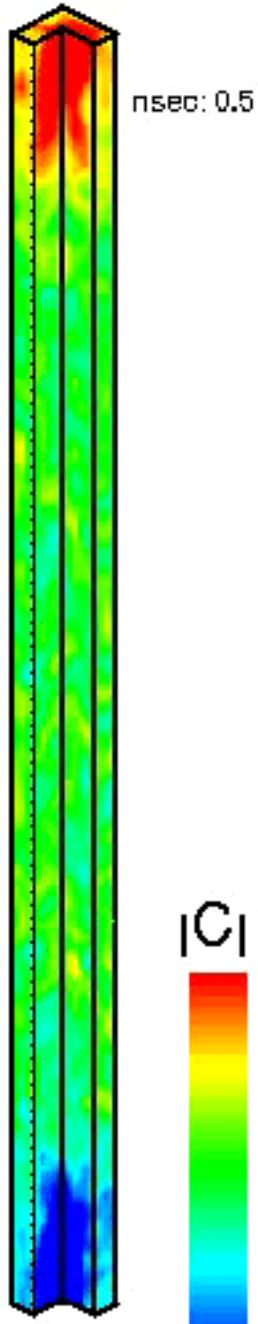
Landau-Lifshitz-Gilbert

Langevin simulation

4949 lattice points

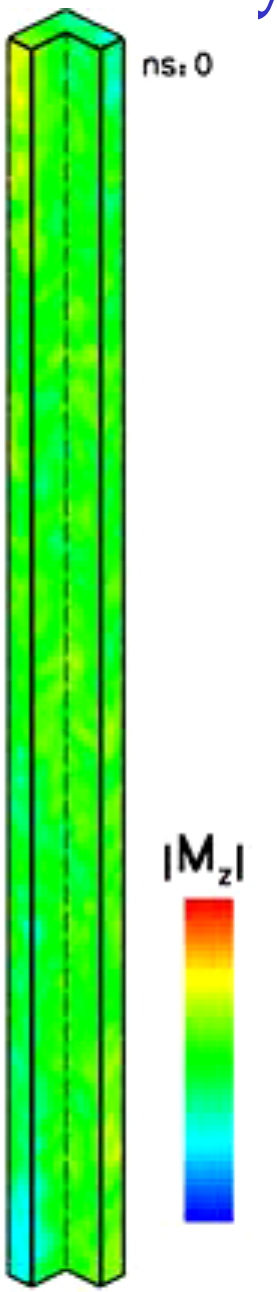
Time: 1.2 ns

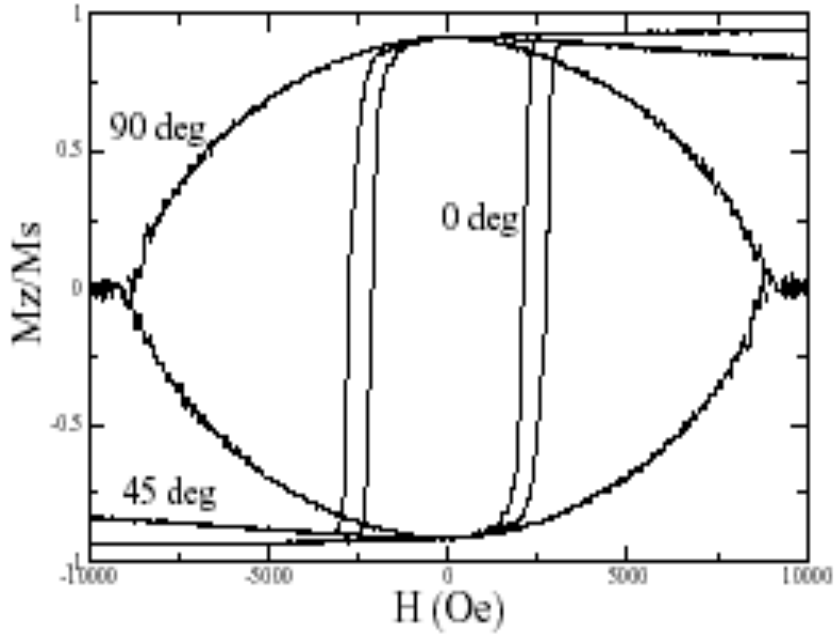
Curl



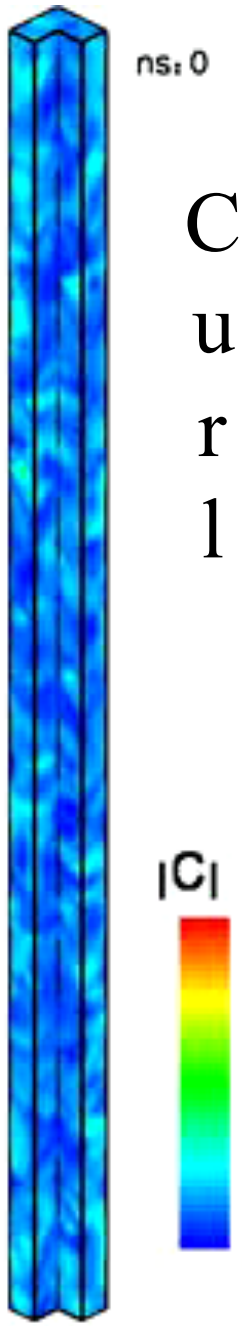
Hysteresis in transverse field

M_z





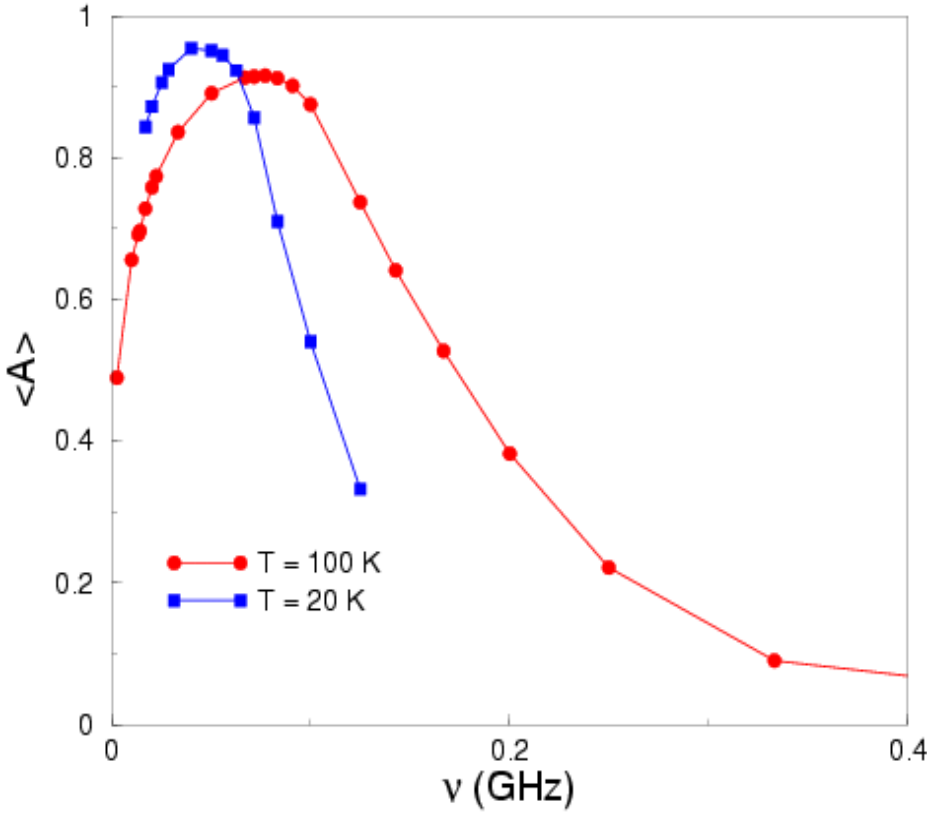
10 nm x 10 nm x 150 nm Iron Pillar
6 x 6 x 90 lattice
$T = 20$ K
$H_0 = 10{,}000$ Oe
Period = 15 ns



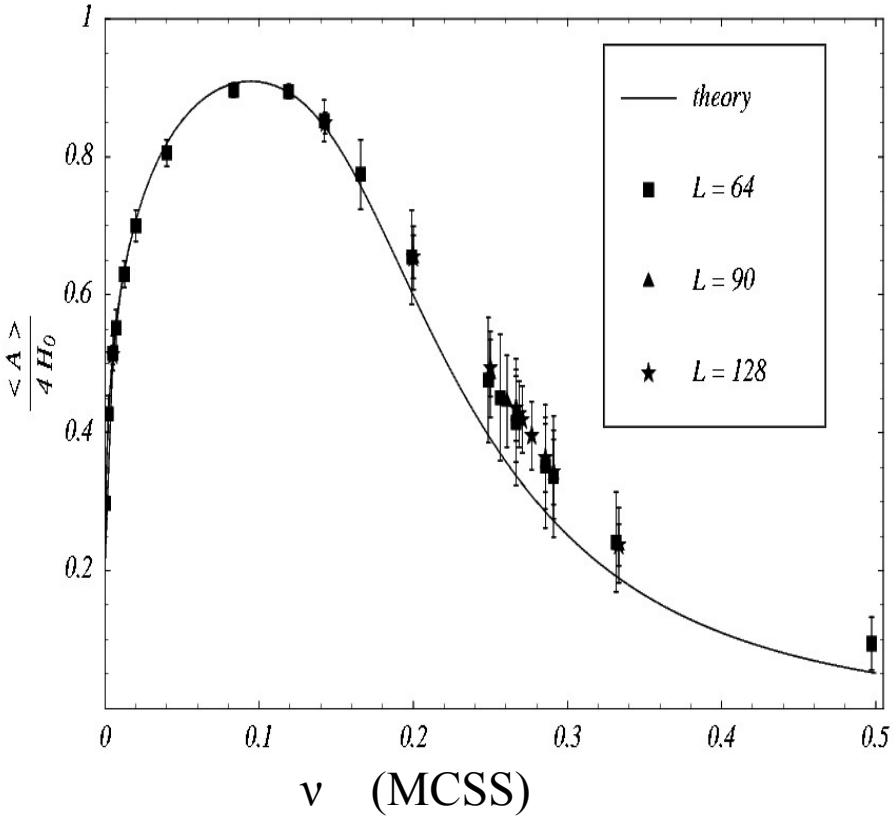
Curl

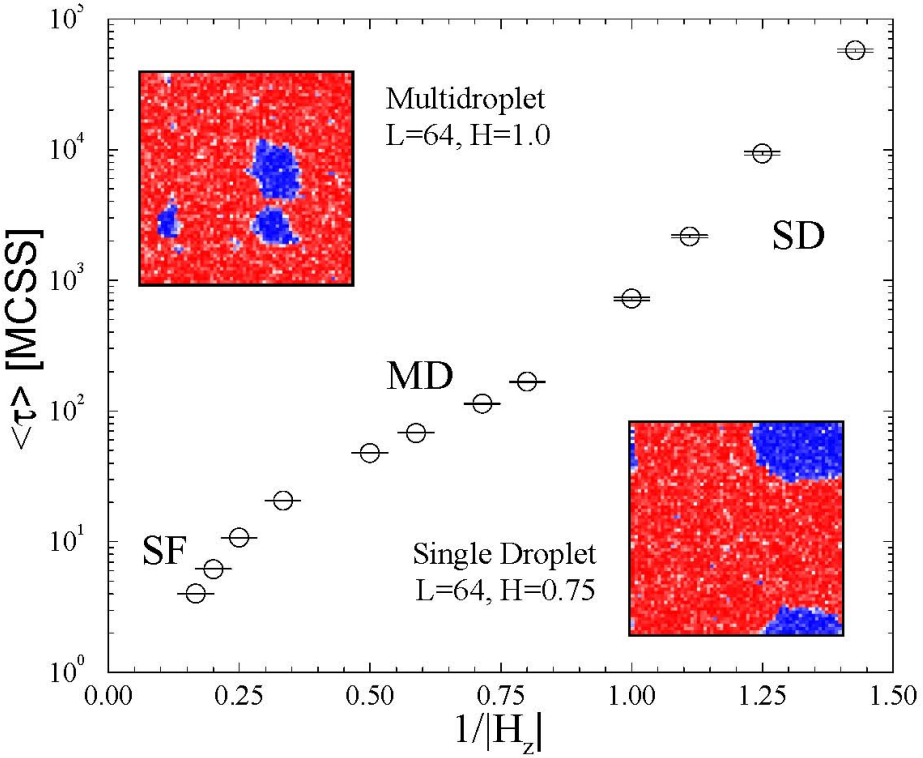
Average loop area vs frequency

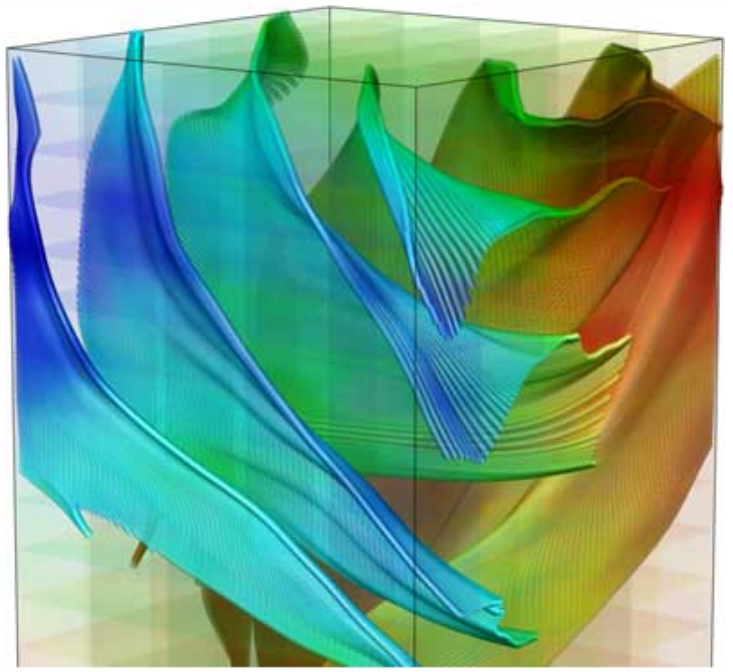
Micromagnetics



Kinetic Ising Model







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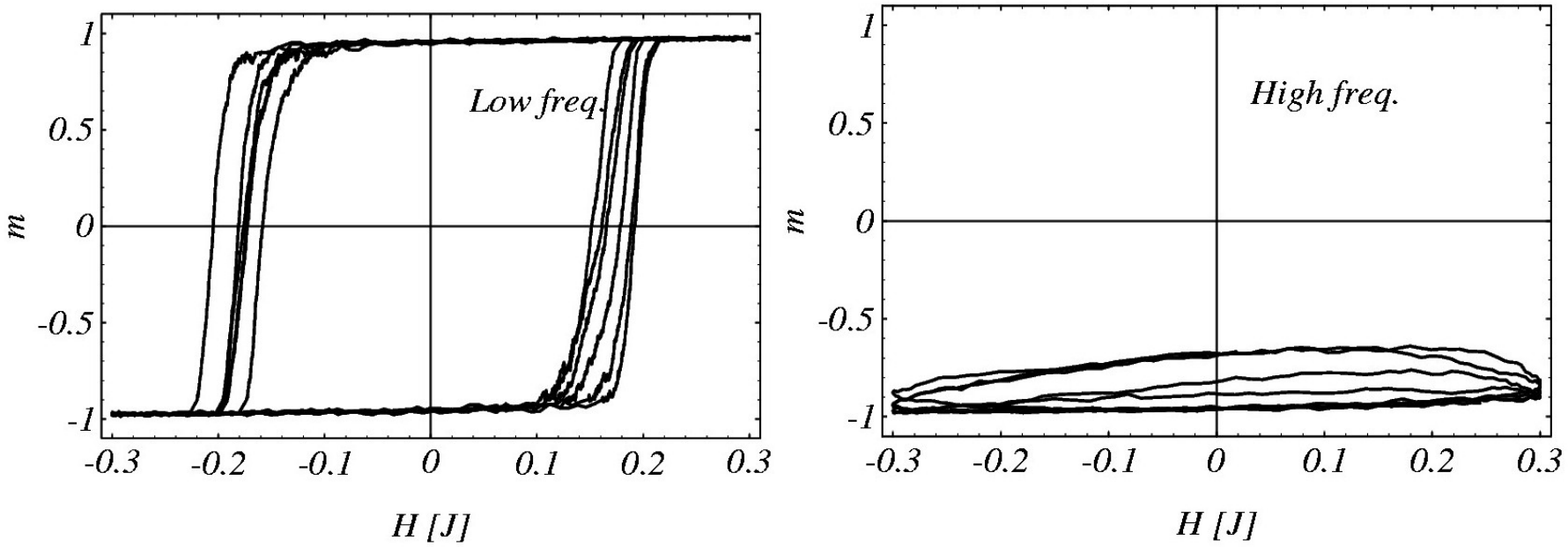
Symmetry breaking in hysteresis loops

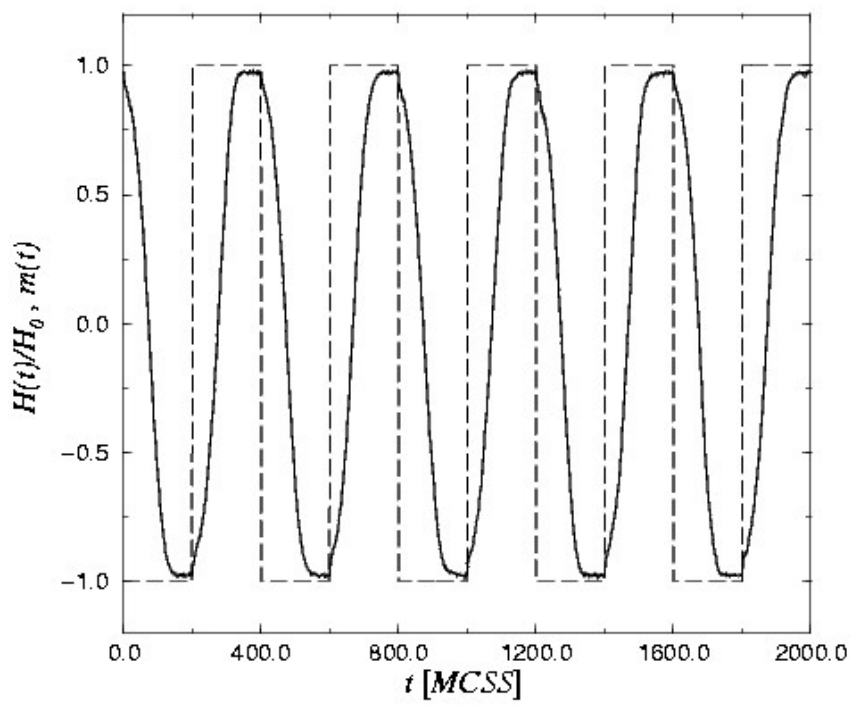
Ising model in sinusoidal field at $0.8T_{\mathrm{c}}$



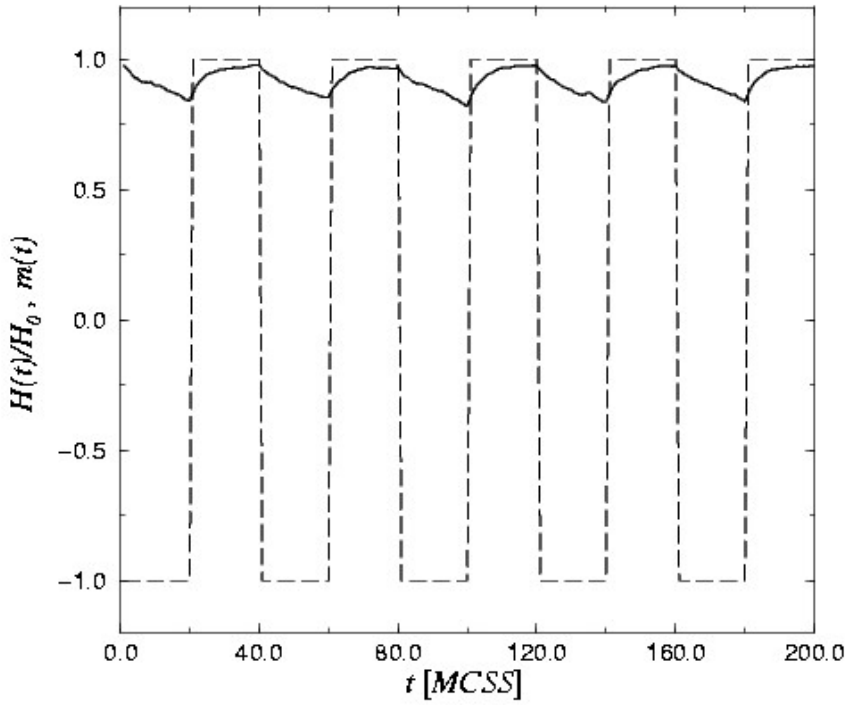
Dynamic phase transition

$$T = 0.8T_c\ ,\ H_0 = 0.3J$$

Low frequency



High frequency



Symmetry breaking!

Hysteresis

Apply oscillating field,

$$\text{Commonly: } H(t) = H_0 \sin(\pi t/t_{1/2})$$

$$\text{Or square wave: } H(t) = H_0(-1)^{\text{int}(t/t_{1/2})}$$

Time-dependent nucleation rate in adiabatic limit:

$$I(T, H(t)) = B(T)|H(t)|^K \exp\left[-\frac{\beta\Xi(T)}{|H(t)|^{d-1}}\right]$$

and interface velocity

$$v(H(t)) \propto |H(t)|$$

Scaled field period:

$$\Theta = \frac{\text{field half} - \text{period}}{\text{metastable lifetime}} = \frac{t_{1/2}}{\langle\tau(H_0, T)\rangle}$$

Square-wave Field: Simulation Details

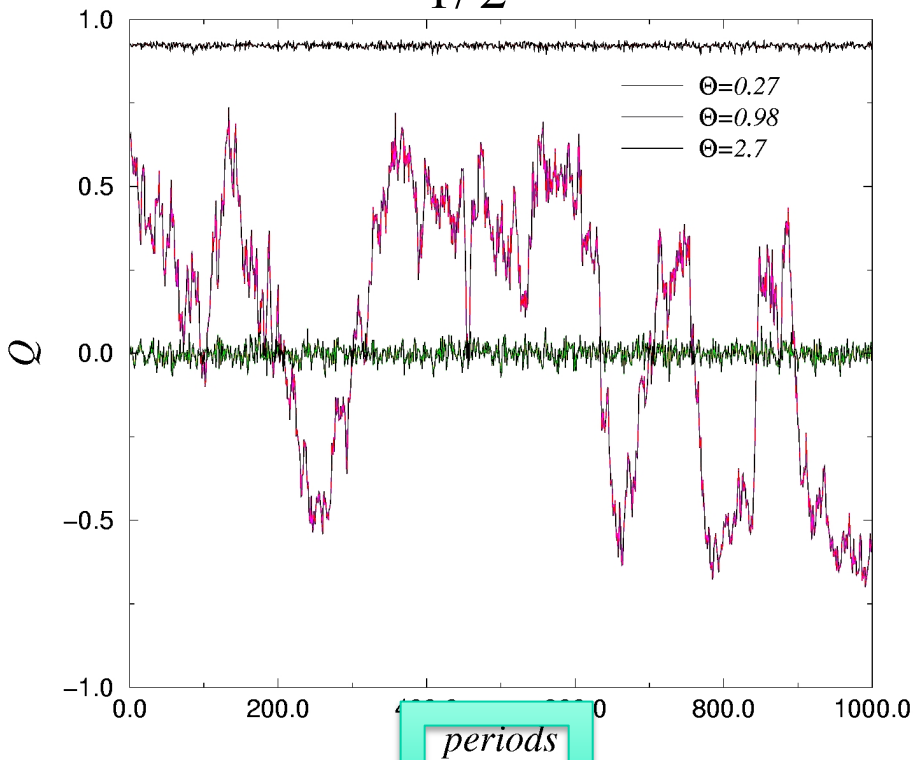
1. **Parameters**
   - Temperature: T=0.8T_c
   - Square lattice, L=64, 90, 128, 256, 512
   - Applied square-wave field:
     $H(t) = H_0(-1)^{\mathrm{int}(t/t_{1/2})}$, $H_0 = 0.3J$.
   - Lifetime: $\langle \tau(H = H_0, T) \rangle = 75$
   - Droplet separation: $R_0 \approx 10$
   - Dimensionless field period: $\Theta = \frac{t_{1/2}}{\langle \tau(H_0,T) \rangle}$
   - Run lengths: $0.3 - 1.5 \times 10^7$ MCSS

2. **Analysis**
   - Period-averaged magnetization: $Q = \frac{1}{2t_{1/2}} \oint m(t) \mathrm{d}t$

     is the dynamic order parameter

Analyze the period-averaged order parameter

$$Q = \frac{1}{2t_{1/2}} \oint m(t)\mathrm{d}t$$

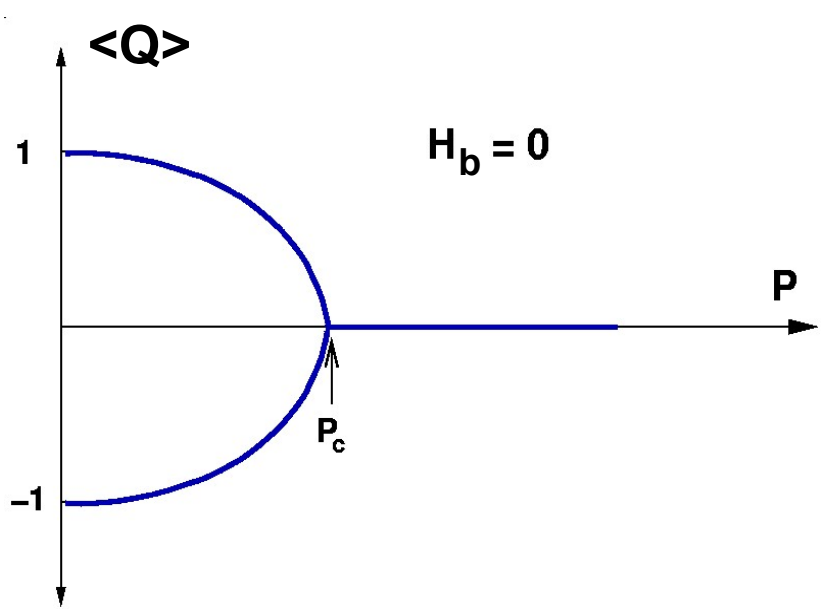


Dimensionless period: Θ = *Period/Lifetime*

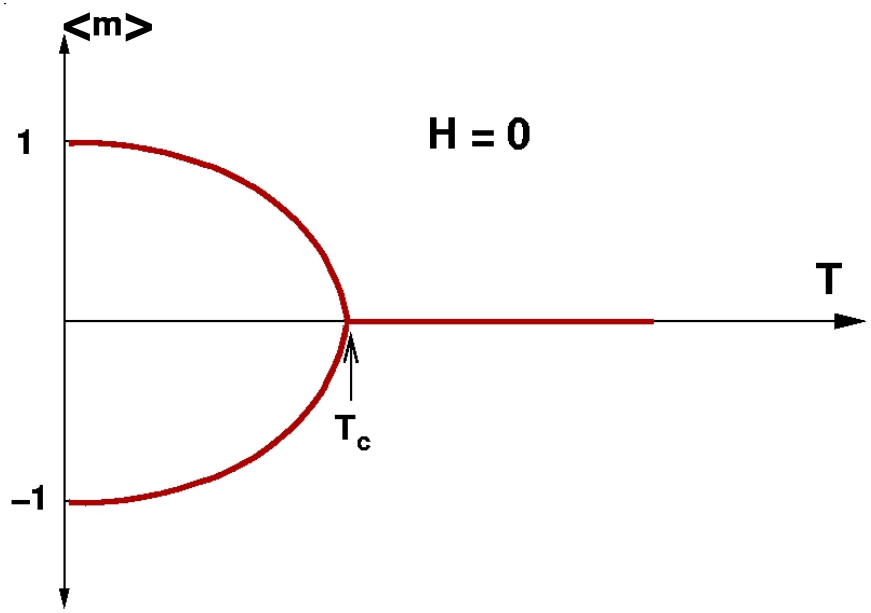
$$T = 0.8T_c,\ H_0 = 0.3J$$

Dynamic Phase Transition (III)

- Non-equilibrium phase diagram is analagous to equilibrium diagram



Non-equilibrium (DPT)



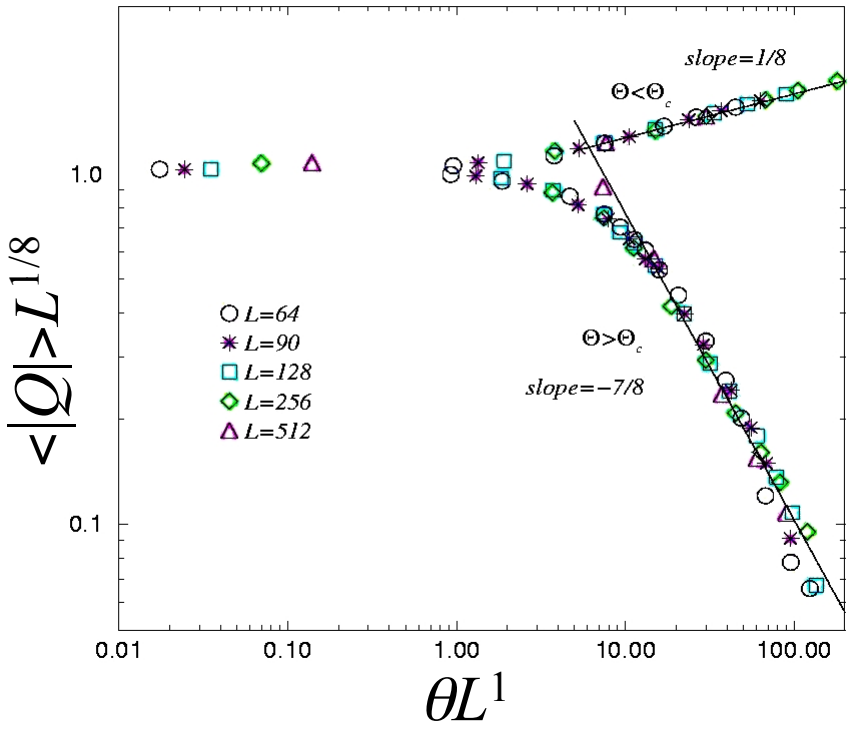
Equilibrium

- Time-averaged 'bias field' $H_b = \frac{1}{P}\int_{t=0}^{t=P} H(t)dt$ conjectured to be field conjugate to dynamic order parameter Q_i

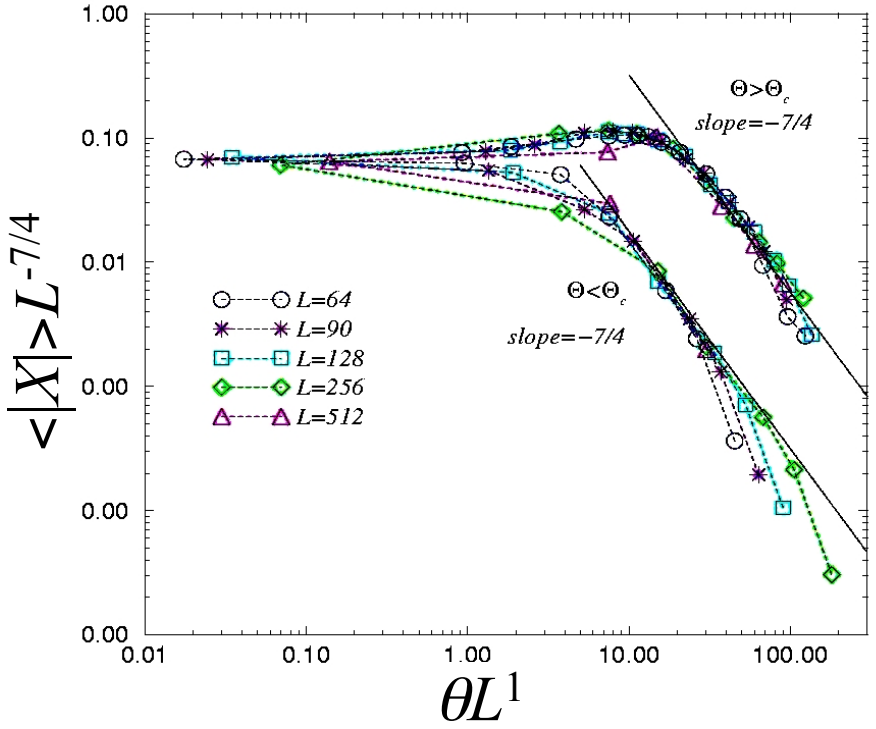
Data collapse with Ising exponents

$$\beta = 1/8\ ,\ \gamma = 7/4\ ,\ \nu = 1$$

Order parameter Q $(\theta = |\Theta - \Theta_c|/\Theta_c)$:



Order-parameter fluctuations:



Ising universality class predicted (cellular automata with +-symmetry):
G. Grinstein, C. Jayaprakash, and Y. He, Phys. Rev. Lett. 1985

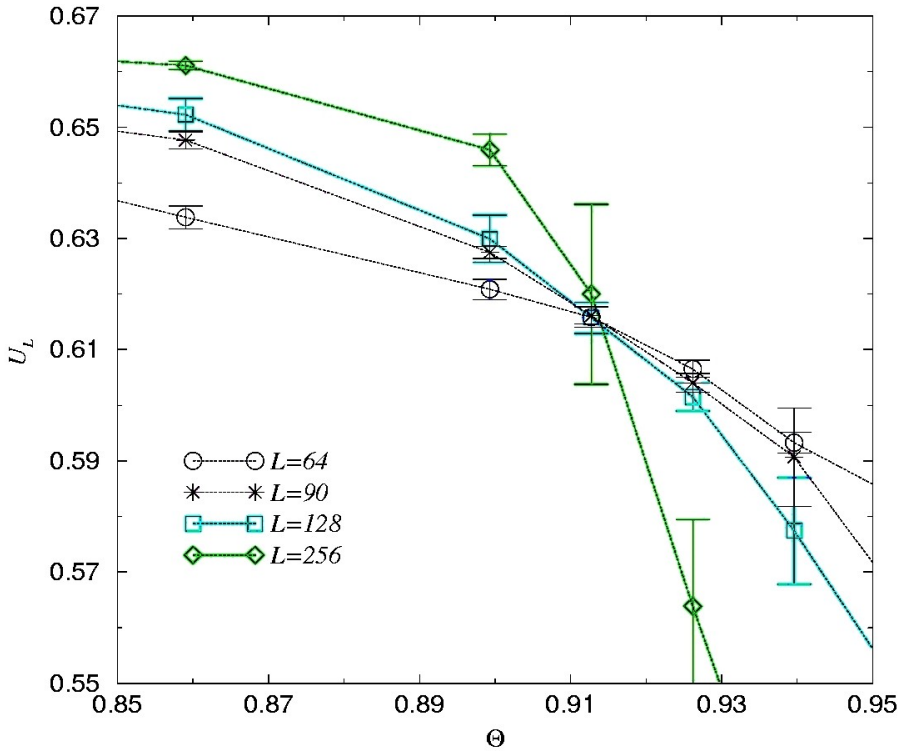
Finite-size scaling

Fourth-order cumulant ratio

$$U_L = 1 - \frac{\langle |Q|^4 \rangle_L}{3\langle |Q|^2 \rangle_L^2}$$

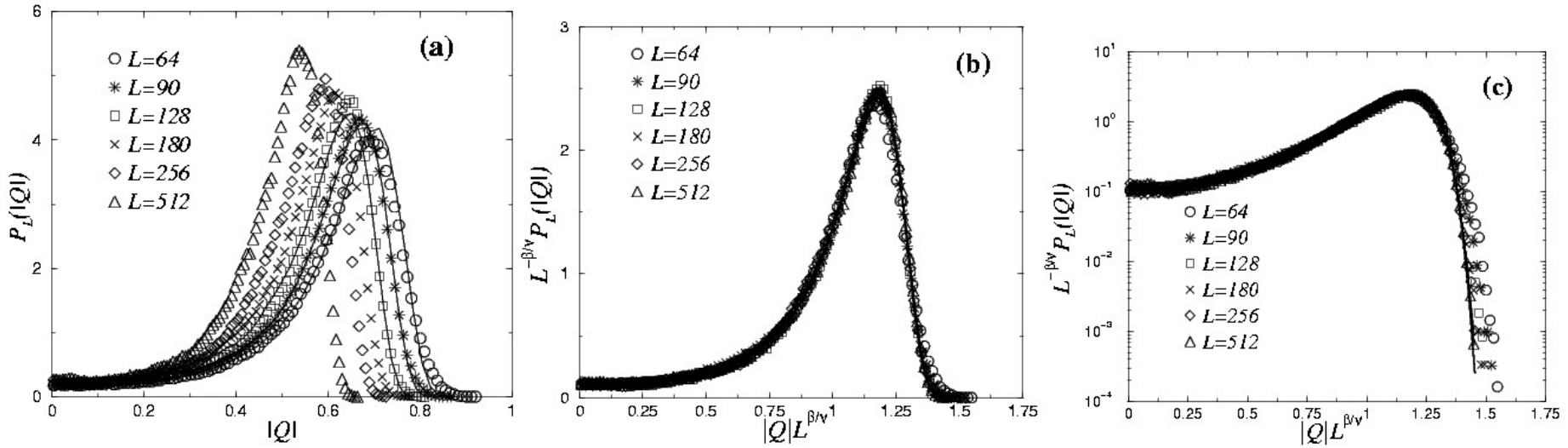
Describes shape of order-parameter distribution. Fixed point

$U^* = 0.611(3)$, $\Theta_c = 0.918(5)$



Scaling of order-parameter distribution, $P_L(|Q|)$

Scaling with Ising exponents, $\beta/\nu = 1/8$

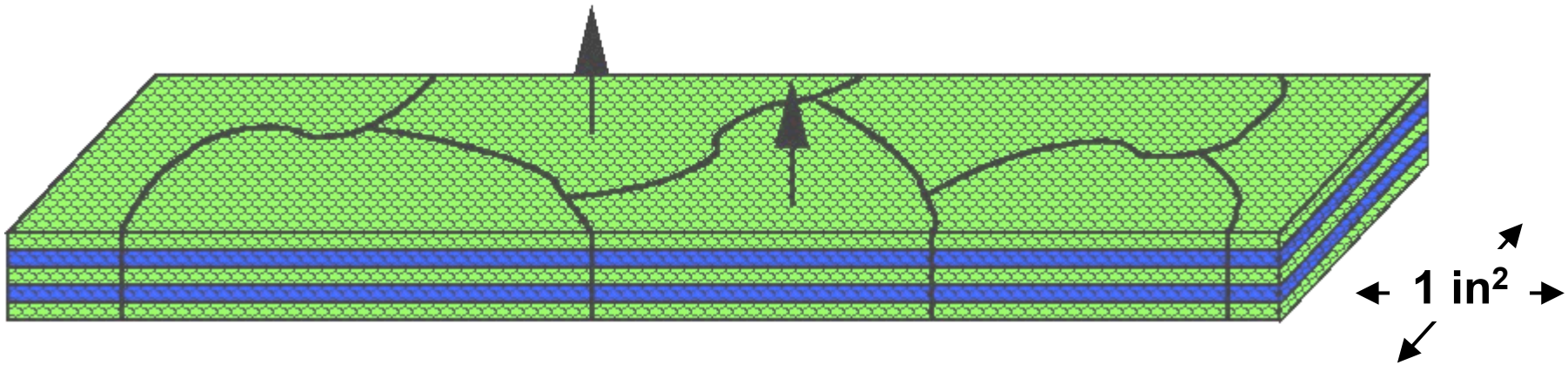


Unscaled Scaled Lin/Log

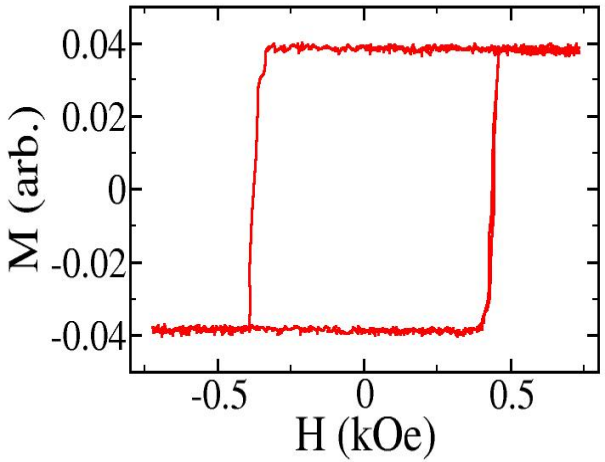
Conclusion: This nonequilibrium phase transition is in the equilibrium Ising universality class!!

(Confirmed analytically, Fujisaka, Tutu, Rikvold)

Experimental $[Co/Pt]_3$ multilayer system

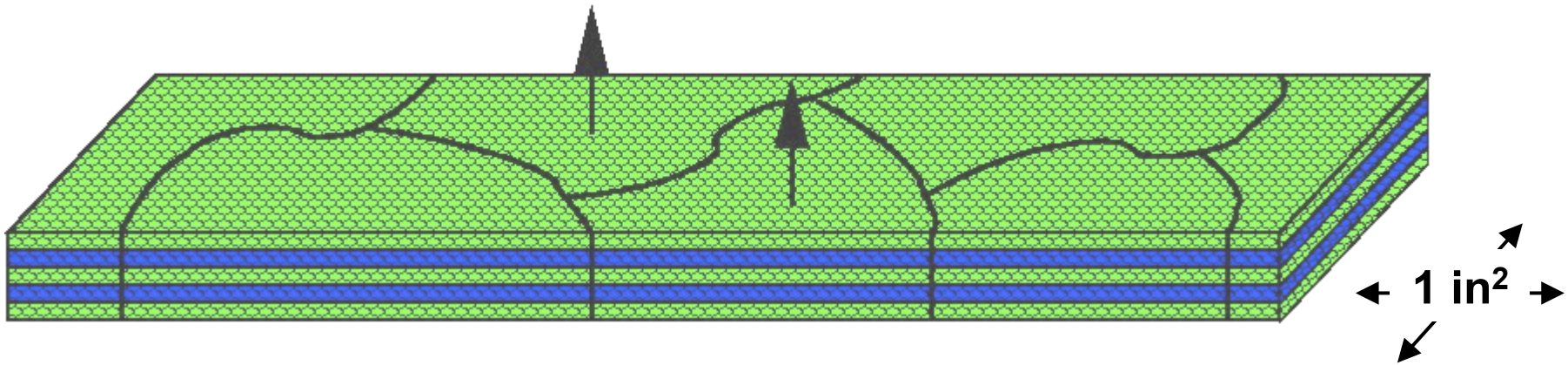


- Strong perpendicular anisotropy in ultra-thin Co(0.4nm) layers
- Pt interlayers, at thickness 0.7nm, couple three Co layers into effectively a single film
- Dipolar interactions are weak → single-domain state at saturation/remanence
- Theoretical and experimental evidence that equilibrium behavior is in universality class of 2-d Ising model



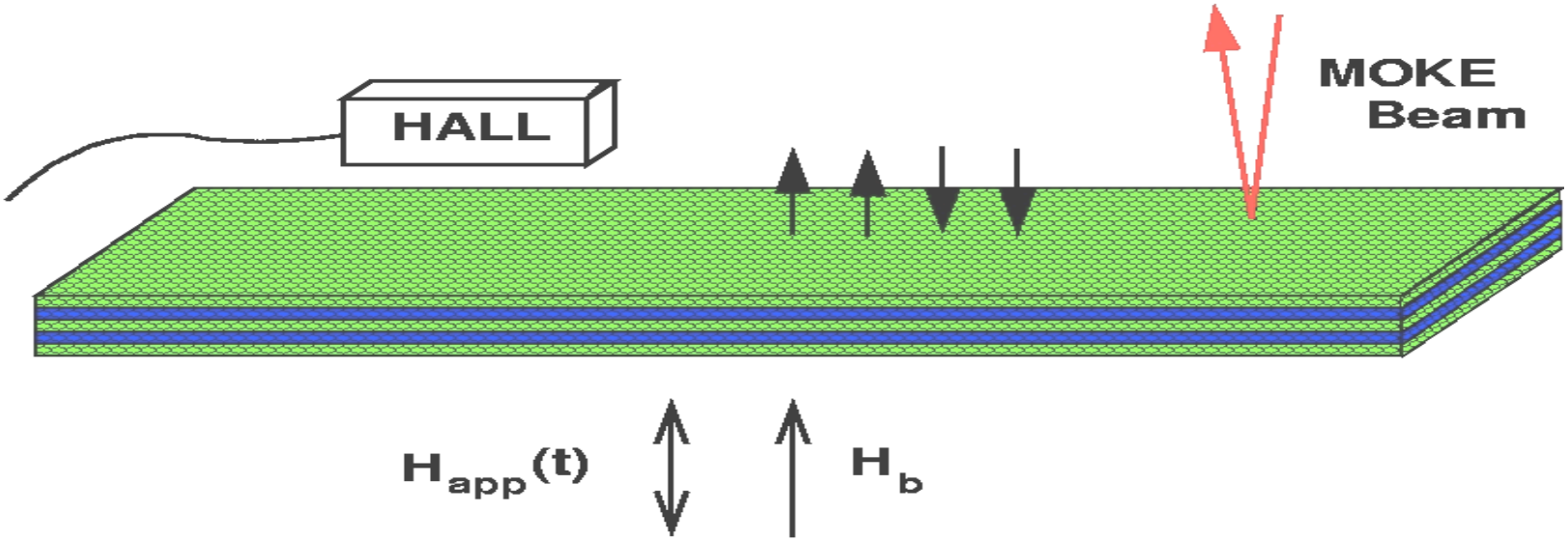
Back et al., Nature 378: 597 (1995)

Experimental $[Co/Pt]_3$ multilayer system (II)



- Grain size of 300-3000 nm inherited from silicon substrate
- Variation of in-plane crystallographic axes at grain boundaries creates variations in strength of anisotropy
- Multilayer surface is atomically smooth, with single-step boundaries between terraces
- Both effects create pinning centers for droplet wall motion

Experimental procedure



- Apply out-of-plane, sawtooth magnetic field for 50 cyles, with periods in range P = 7.6 - 26.4 s, using an electromagnet

- Sensitivity of DPT to non-zero average field → measure response of system in small additional constant bias fields H_b of varying strength

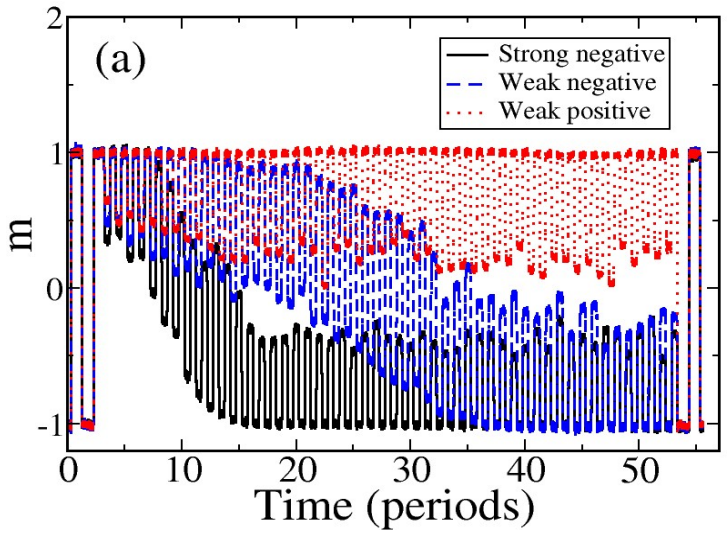
- Record net magnetic field above sample using Hall probe

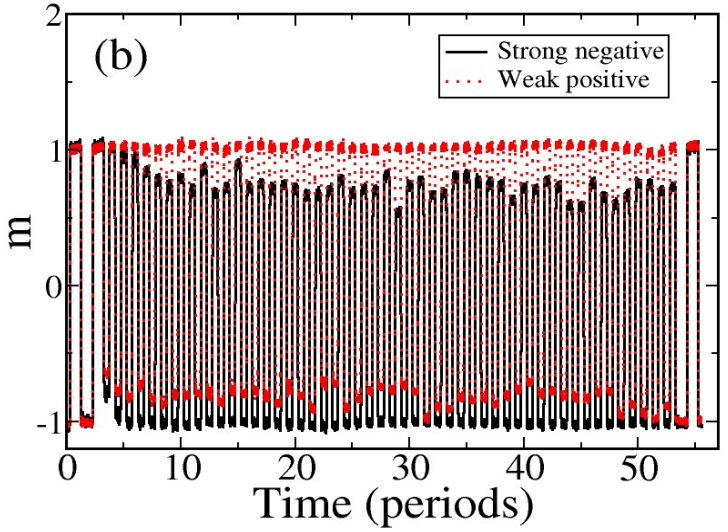
- Measure magnetization using polar MOKE beam (spot size ≈ 1 mm^2)

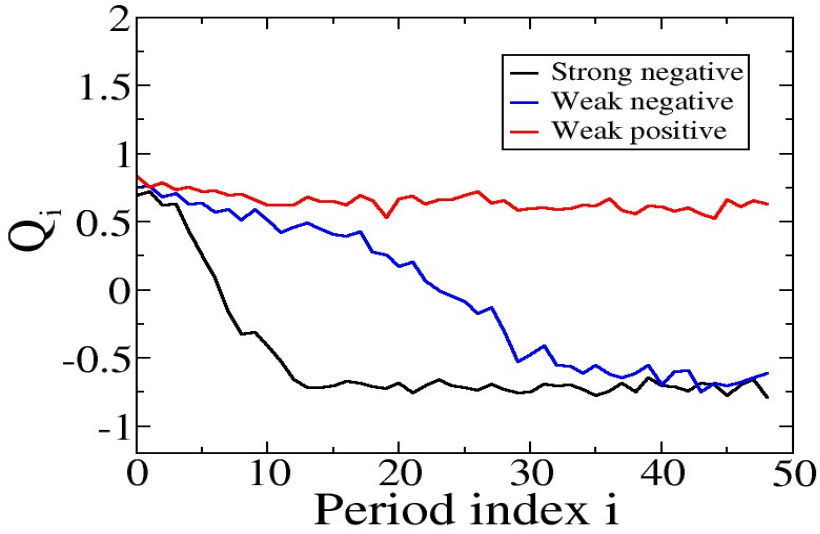
Experimental results: time series of magnetization

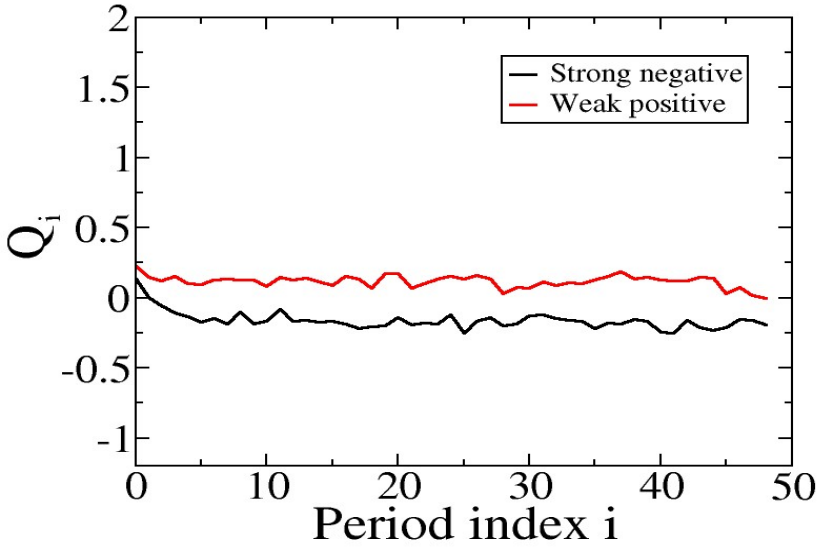
P = 16.2 s

P = 38.1 s





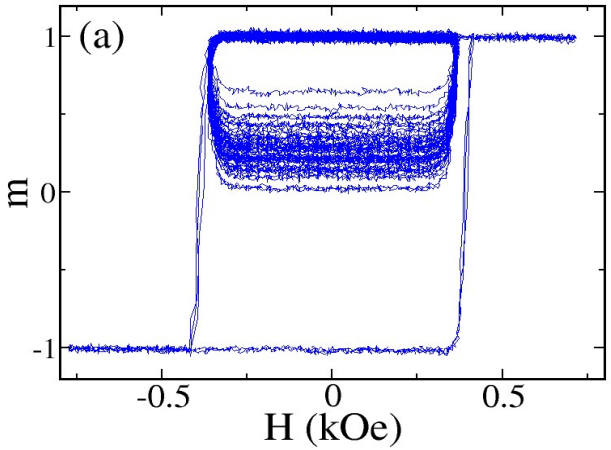


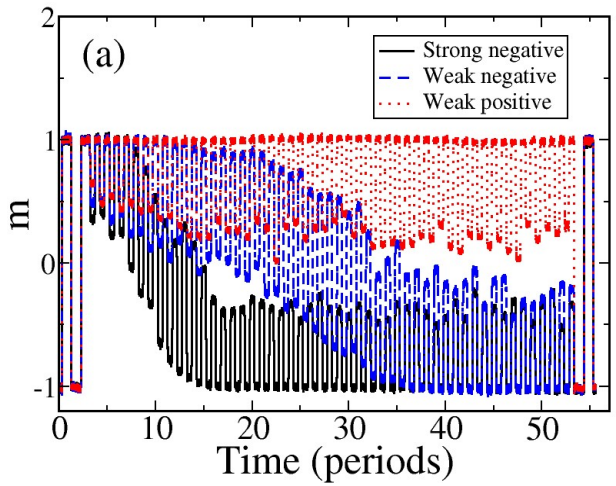


Comparison: experiment with kinetic Ising model (I)

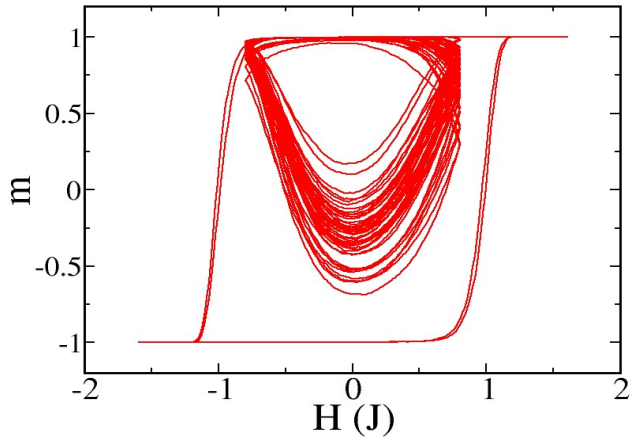
Robb et al, PRE 2008

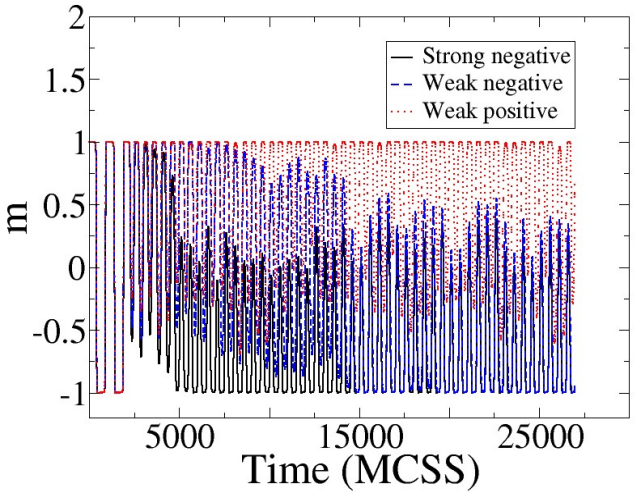
Multilayer data : P = 16.2 s





Ising simulation : P = 500 MCSS

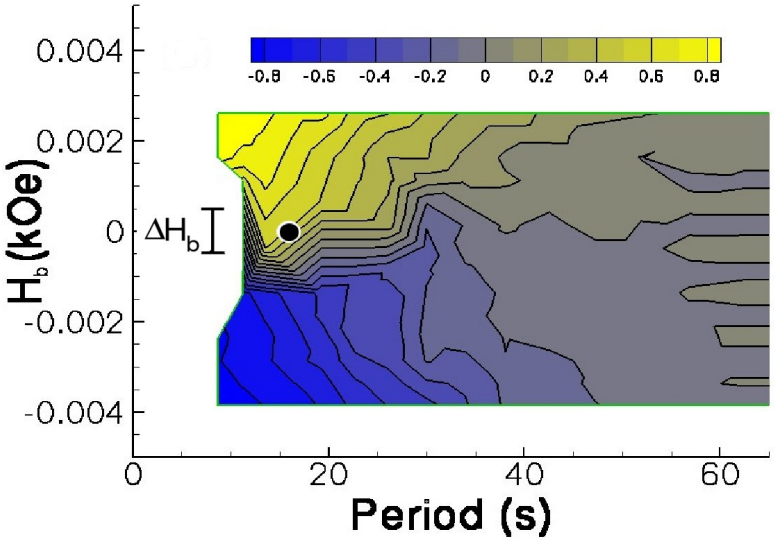




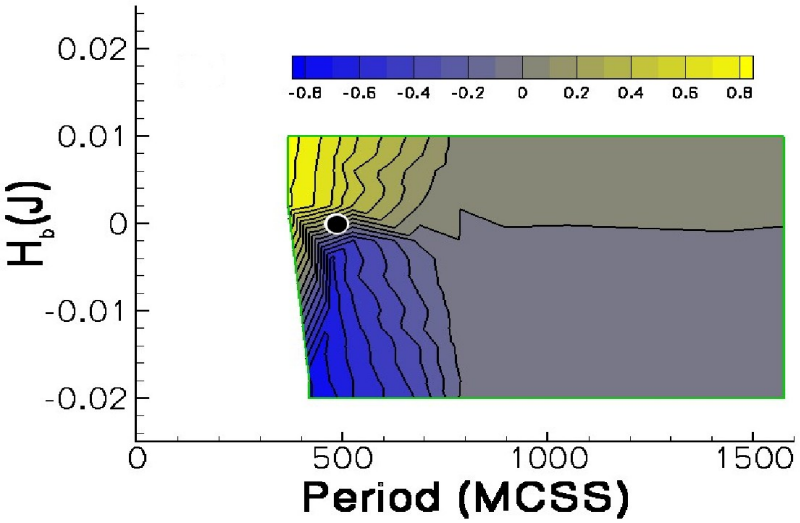
Comparison: experiment with kinetic Ising model (II)

Robb et al, PRE 2008

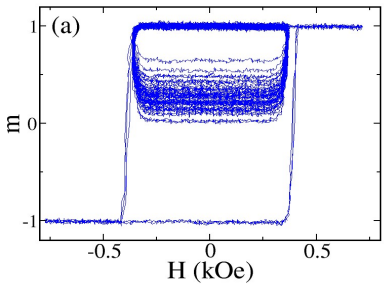
Multilayer data

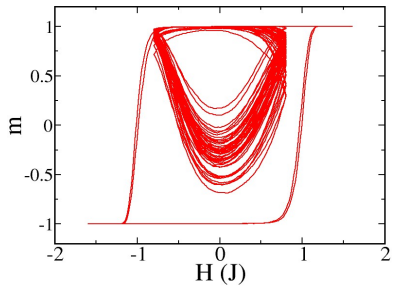


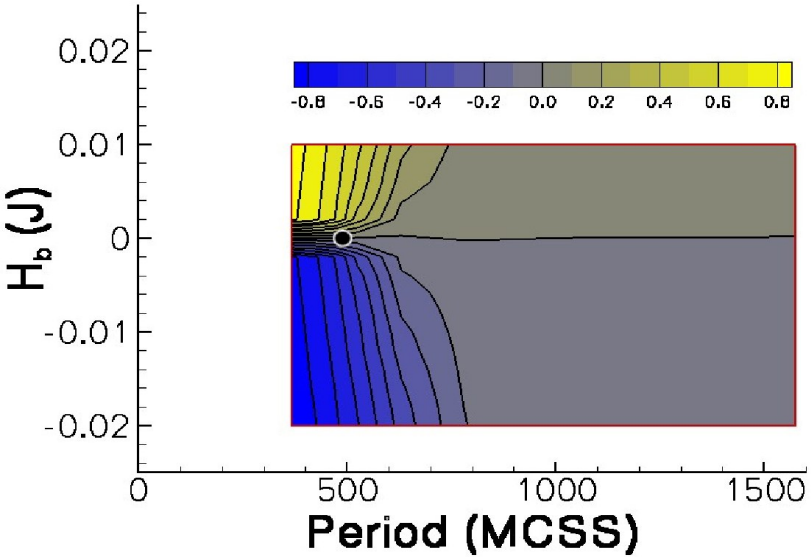
Ising simulation



$\langle Q \rangle$ **vs.** P **and** H_b



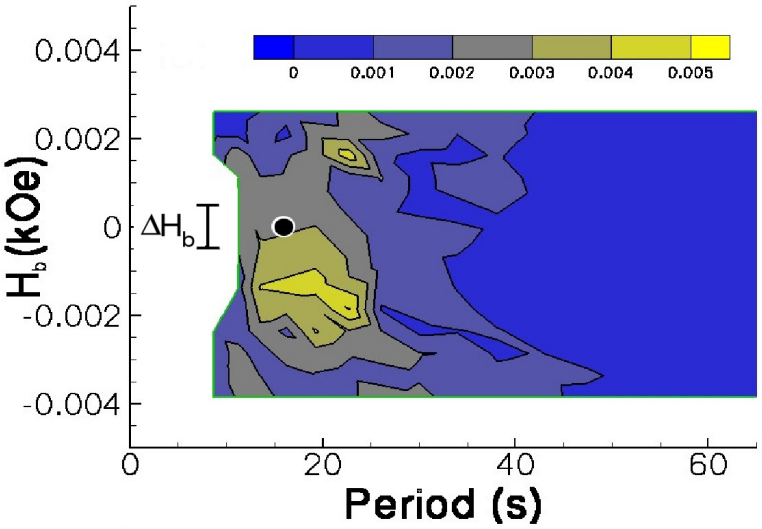




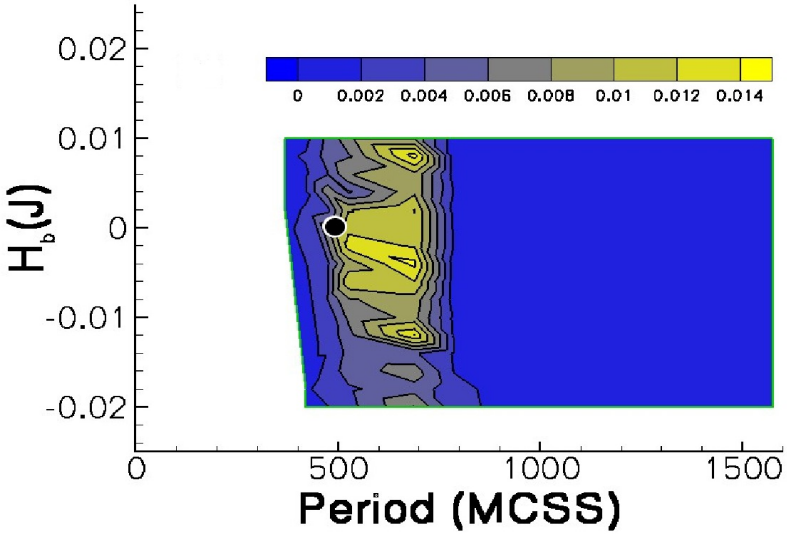
Comparison to kinetic Ising model (III)

Robb et al, PRE 2008

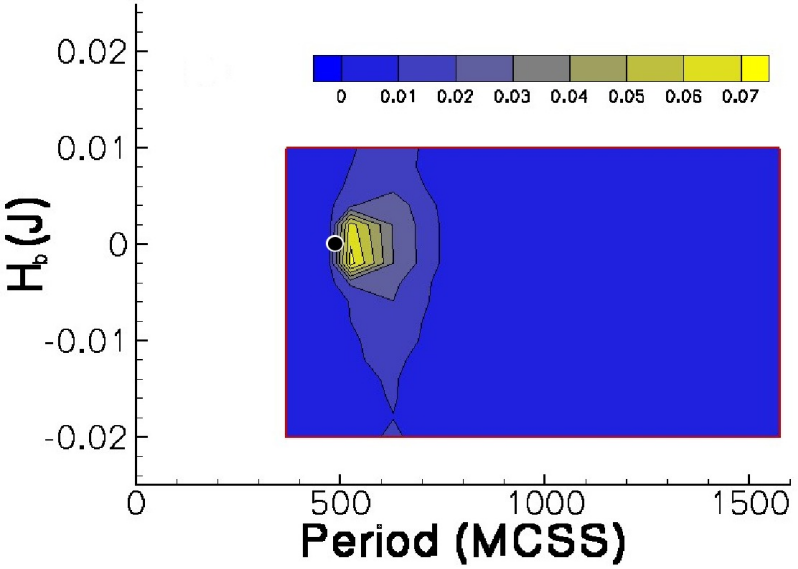
Multilayer data



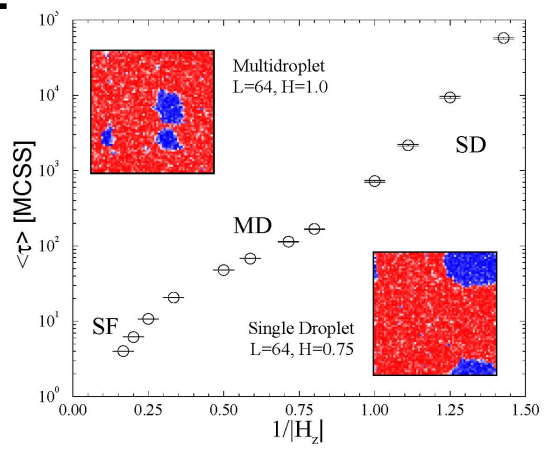
Ising simulation

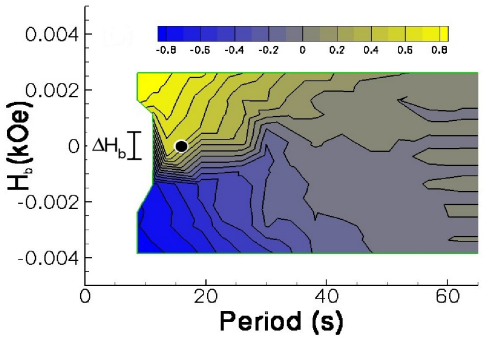


$\sigma^2(Q_i)$ **vs.** P **and** H_b



CONCLUSIONS
HOMOGENEOUS
nucleation and growth
and metastability

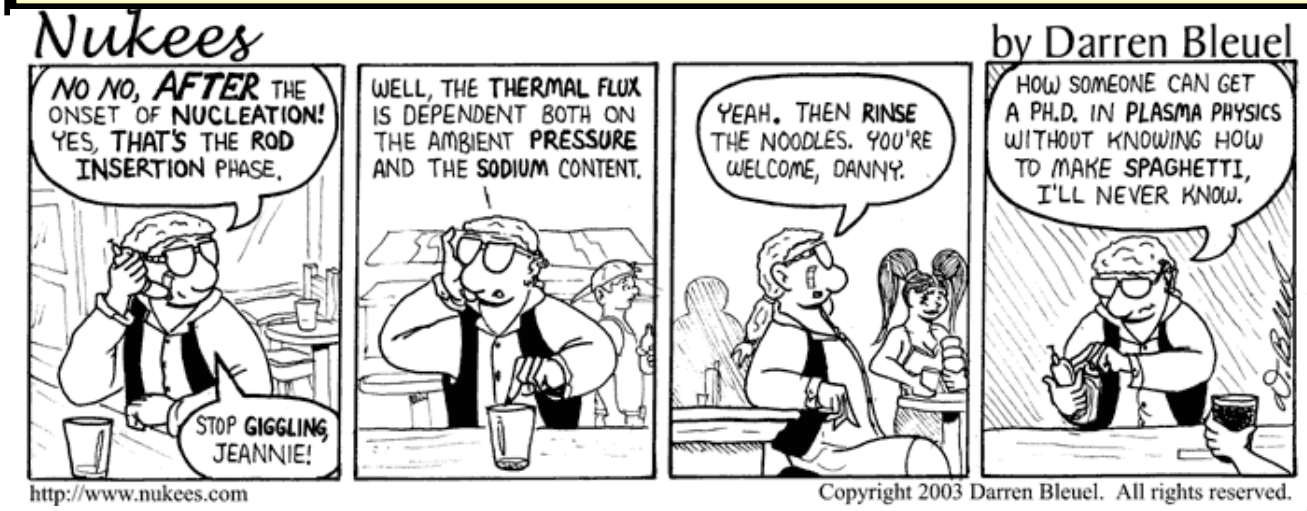




HOMOGENEOUS NUCLEATION / GROWTH / METASTABILITY

- Homogeneous nucleation and growth is complicated!
  - At least 4 length scales
- Algorithms can be devised to efficiently study nucleation & growth
  - Monte Carlo with Absorbing Markov Chains (MCAMC)
  - Projective Dynamics (PD)
  - Non-trivial parallelization
- How does hysteresis loop area depend on f for oscillating field?
  - What frequencies are you investigating?
- Can changing dynamic change nucleation rate exponential?
  - Yes, at low temperature for discrete state-space models
- There is a dynamic phase transition, with an experimental realization.
  - Universality class of normal Ising model
  - Multilayered thin films of Co/Pt

Nucleation and Growth ---



Spasiba!

Thank you!