Controlled limits of FORC theory: Mean field and Nucleation

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One of the key factors limiting the performance of magnets is that the coercivity and other parameters have a distribution with a finite width, and the reversal starts at the weakest link.

We must first determine the distribution of coercivities to learn how to reduce the width of this distribution.

The FORC technique characterizes a system via the distribution the local coercivities $H_c$ and in addition the local interaction fields $H_b$.

FORC captures these as a joint distribution $\rho(H_c, H_b)$ instead of a product of two distributions, thus capturing underlying correlations.
Hysteretic behavior modeled as a collection of hysterons (Preisach 1935)

Hysterons are two state systems with an interaction field $H_b$ and a coercivity $H_c$, with distribution $\rho(H_c,H_b)$

- Mathematical foundation: Mayergorz 1987
- Phenomenology, modelling: Dellatorre, Vajda, Cardelli
  - Assume coercivity & bias field distribution $\rho(H_c,H_b)$
  - Fit parameters of $\rho(H_c,H_b)$ to reproduce hysteresis loop
1. Measure First Order Reversal Curves (FORCs) on sample

2. Select a model and simulate FORCs

3. Determine $\rho(H, H_R)$ from both

$$\rho(H, H_R) = -\frac{1}{2} \frac{\partial^2 M(H, H_R)}{\partial H \partial H_R}$$

4. Evaluate/develop model based on how well simulated FORCs reproduce experimental FORCs

UCDavis: Pike et al 1998; Katzgraber, GTZ PRL 2002; Winklhofer, GTZ 2006
Iasi: Stancu 2003; Stoleriu, Postolache, Spinu 2003 - ....
Characterizing interactions in fine magnetic particle systems using first order reversal curves

... 7. FORC diagrams for a system of single domain particles in three models: a noninteracting model; b mean interaction field model with k 6, and c mean interaction field model with k 6 sw 120 and sw 0.4. 6662 J. Appl. Phys., Vol. 85, No. 9, 1 May 1999 Pike, Roberts, and Verosub ...
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The evolution of theories for interacting systems:

(1) Mean field theory

(2) Controlled fluctuation expansion around mean field
   Not available for FORC until recently

1. Strong dipolar interaction:
   1.1. Mean Field Theory of FORC
   1.2 Controlled local fluctuations corrections
   1.3 Test/verify theory experimentally on nanoparticle arrays

2. Strong exchange interaction:
   2.1 Analyze FORC diagrams in terms of nucleation
   2.2 Establish phase diagram
the average field

A normal distribution of the critical fields was chosen, with account for that, a small dispersion of coercivities was con-

cylindrical wires with the same length (for state dependent interactions between wires.

FORC diagrams, we propose a very simple model that accounts

can slightly modify the individual switching fields,

teristics of the individual wires, small deviations from paral-

different interwire distances in order to obtain interaction

each wire is opposed to the applied field and it has a demag-

e.g., in the positive saturation state, the interaction field in

III. ISING-TYPE MODEL FOR THE FORC DIAGRAMS

The FORC diagram is the contour plot representation of

The magnetostatic interaction field, in axial direction

nanowires with the length larger than 1

states of a nanowire are just

using

The pillars in this array do not have a perfectly

20 Oe.

Nanowire

arrays, Stancu

JAP (2013)

Nd2Fe14B,

Schrefl, GTZ

JAP (2012)

Perpendicular recording

media, Ross, Pike, GTZ,

PRB (2005)
Dipolar Interaction Strong: Mean Field appropriate

Proposition: Mean field theory can explain wishbone FORC

We simulated a 100x100 dipole array with mean field interactions at T=0, determined FORC diagram

Each dipole has its own anisotropy $H_k^i$

Distribution $D(H_k^i)$: rectangular, Gaussian

Interaction - Mean Field: $H_{\text{int}}^i = \alpha M(H)$

Down-flip: $H + H_{\text{int}}^i < -H_k^i$

Up-flip: $H + H_{\text{int}}^i > H_k^i$

Re-evaluate $M(H)$, keep flipping until all dipoles stable

Rotate from $(H, HR)$ to $(H_b, H_c)$ axes: $H_B = (H_{up} + H_{dn})/2 \quad H_C = (H_{up} - H_{dn})/2$
FORC $\rho$ is not 0 when slope of neighboring FORCs is different

$P_i(H_k^i)$ down-flips at $H_{dn}^i = -H_k^i$ and up-flips at $H_{up}^i = H_k^i$

1. For rectangular $D(H_k)$ neighboring FORCs match for most $H$:
   
   $$-d(dM/dH)/dH_R = \rho = 0$$

2. $H_R = -H_k^i$, $P_i$ is last to down-flip, last to up-flip:

   $dM/dH$ of last upflip unmatched by nearest $H_R$: $-d(dM/dH)/dH_R > 0$:

   Number of unmatched last upflips = number of hysterons with $H_k^i$:

   FORC is a ridge along $H_c$: $\rho(H_b=0,H_c)=D(H_k)$, the coercivity distribution
Demagnetizing Arrays - Ridge \parallel Hc axis

\[ H_{tot} = H + \alpha M(H) \quad \alpha < 0 \]

1. \( P(H_k^{\text{min}}) \) unmatched (min)
2. \( H_{dn}^{\text{min}} = -H_k^{\text{min}} - \alpha M_S \)
3. \( H_{up}^{\text{min}} = H_k^{\text{min}} - \alpha M_S \)
4. \( H_B = (H_{up} + H_{dn})/2 \quad H_C = (H_{up} - H_{dn})/2 \)

Low \( H_C \) end shifted by

\[ \Delta H_B = -\alpha M_S \quad \Delta H_C = 0 \]

1. \( P(H_k^{\text{max}}) \) unmatched (max)
2. \( H_{dn}^{\text{max}} = -H_k^{\text{max}} + \alpha M_S \)
3. \( H_{up}^{\text{max}} = H_k^{\text{max}} - \alpha M_S \)

High \( H_C \) end shifted by

\[ \Delta H_B = 0 \quad \Delta H_C = -\alpha M_S \]
Demagnetizing Arrays – Ridge

Last upflip fields shifted by interaction

\[ H_{tot} = H + \alpha M(H) \quad \alpha < 0 \]

Ridge modified by mean field:

1. Low \( H_C \) end shifted by

\[ \Delta H_B = -\alpha M_S \] off \( H_C \) axis

2. High \( H_C \) end not shifted off \( H_C \) axis

3. Ridge length increases

Dobrota, Stancu (2013)
Demagnetizing Arrays – Edge || Hb axis

On every FORC $P(H_k^{\text{min}})$ first to upflip

- No interaction: 1st upflips at $H=H_k^{\text{min}}$
  Same matched field every FORC, $\rho=0$
- Interaction: 1st upflip fields shifted, thus unmatched
  Top FORC
  $H=H_k^{\text{min}}+\alpha M_S$, $H_R=-H_k^{\text{min}}-\alpha M_S$
  Bottom FORC:
  $H=H_k^{\text{min}}+\alpha M_S$, $H_R=-H_k^{\text{max}}+\alpha M_S$

First upflip fields are not matched
- A $\rho>0$ edge forms by interaction,
- Edge is tilted
Many FORCs exhibit negative regions.

Change rectangular $D(H_K)$ to Gaussian.

For decreasing half of Gaussian $D(H_K)$, the number of dipoles along FORCs is decreasing in the high $H_K$ region - Previously matching $dM/dH$s now decrease.

Negative FORC $\rho$ (only) in high $H_K$ region.
Demagnetizing Arrays – Mean Field Theory

Ridge: unmatched last upflip,
Represents Hk coercivity distribution

Edge: unmatched first upflip
Represents interaction parameters

Effects of Mean Field Interactions:
1. Min \( H_K \) end shifts to \( H_B > 0 \)
2. Max \( H_K \) end stays at \( H_B = 0 \)
3. Ridge length increases by \( -\alpha M_S \)
4. Edge develops to negative HR
5. Negative region: from peaked \( D(H_K) \)

Fig. 3, Gilbert et. al.
Beyond Mean Field: Nearest Neighbor Interaction

Controlled expansion around Mean Field: local fluctuations
Include nearest neighbor non-mean field terms

(dn1) positive saturation $\rightarrow$ checkerboard ($\uparrow \uparrow \uparrow \rightarrow \downarrow \downarrow \uparrow : H_{int}=2H_{n.n.}$)
(dn2) checkerboard $\rightarrow$ negative saturation ($\downarrow \uparrow \downarrow \rightarrow \downarrow \downarrow \downarrow : H_{int}=-2H_{n.n.}$)
(dn3) frust. checkerboard $\rightarrow$ frust. checkerboard ($\downarrow \uparrow \uparrow \rightarrow \downarrow \downarrow \uparrow : H_{int}=0$)
Experimental Test of Mean Field Theory

(K. Liu, R. Dumas)
Polycrystalline Co ellipses
E-beam lithography
Liftoff technique
Major/minor axis: 220/110nm
Created 50x50 micron array of ellipses
Measure middle of the array to avoid edge effects
- magnetizing arrays
- demagnetizing arrays
Varied coupling strength by varying separation: 150/200/250 nm
Experiment vs. Mean Field Theory

MF+NN close to experiment
SUMMARY, Part 1: Mean Field FORC

1. Developed Mean Field Theory of FORC technique
2. Explained paradigmatic wishbone structure, present in many classes of magnets
   - Ridge: Represents $H_k$ coercivity distribution
   - Edge: Represents interaction parameters
3. Developed controlled fluctuation expansion around Mean Field
4. Verified on controlled arrays of nanoparticles
Another two-branched FORC is observed on certain FeNdB permanent magnets where exchange is much stronger.

We termed these FORCs “boomerangs”
Boomerangs can be unmasked in large number of FORCs with deshearing

Schrefl, Zimanyi et al, JAP (2012)
**Wishbone vs. Boomerangs**

**Wishbone**
- Ridge tracks Hc axis
- Edge tracks Hb axis
- High Hc end on Hc axis
- Ridge-edge angle < 90

**Boomerang**
- Ridge 45 from Hc axis, tracks H axis
- Edge 45 from Hb axis, tracks HR axis
- High Hc end away from Hc axis
- Ridge-edge angle = 90
Simulation Details

$5 \times 5 \times 1 \mu m$ sample with 50nmx100x100nm and 50nmx200x200nm grains $\sim 10,000 - 50,000$ grains

Use OOMMF code

Individual grains modeled without internal structure: no multi-domain grain

Parameters need to be scaled:

- $M_s$ naturally scaled with the grain volume
- $K$ naturally scaled by grain volume, or logarithmically corrected
- $A(scaled)$ inter-grain exchange is known poorly:
  
  (1) Estimate range of $A(scaled)$ – Skomsky theory
  
  (2) Explore estimated range of $A(scaled)$
Scaling A

Grain-Boundary Micromagnetism
R. Skomski, H. Zeng, and D. J. Sellmyer

\[ J_{\text{eff}} = \frac{L^2 \sqrt{AK}}{1 + \frac{t \sqrt{AK}}{2A'}} \]

For 50-200 nm grain size and t~1-2nm, A(scaled) can sweep [0.1-10]x10^{-9} J/m for A'(microscopic)/A(microscopic) ~0.05-0.5
Sweeping with Exchange A

\[ A = 1 \times 10^{-9} \text{ J/m} \]
\[ A = 5 \times 10^{-10} \text{ J/m} \]
\[ A = 1 \times 10^{-10} \text{ J/m} \]

\[ \mu_0 M_s = 1.64 \text{ T} \]
\[ K_U = 4.3 \text{ MJ/m}^3 \]
\[ 5 \mu m \times 5 \mu m \times 1 \mu m \]
Sweeping with Exchange A – Zooming to Transition

$A = 5 \times 10^{-9}$ J/m

$A = 2 \times 10^{-9}$ J/m

$A = 3 \times 10^{-9}$ J/m

$A = 4 \times 10^{-9}$ J/m

$A = 6 \times 10^{-9}$ J/m

$A = 7 \times 10^{-9}$ J/m

$M_S = 1.64$ T

$K_U = 4.3$ MJ/m$^3$

$5\mu m \times 5\mu m \times 1\mu m$
- Left facing boomerang FORCs observed for $A_{\text{scaled}} > 2 \times 10^{-9} \text{ J/m}$

- Proposition: Boomerangs indicate reversal by exchange-driven domain wall growth

- Relatively strong inter-grain exchange, $A'/A > 0.1$ needed to explain boomerang FORCs
Diagnostics of Energy Terms

For $A > 3 \times 10^{-9} \text{ J/m}$

$E(\text{exchange}) \sim E(\text{dipolar})$

Evidence for importance of exchange, but not decisive
Energy FORCs

Magnetic FORC

Exchange FORC

Demag FORC

Anisotropy FORC

Zeeman FORC

A=2.5E-9 J/m
The FORC of Anisotropy closely tracks the FORC of the Exchange

Anisotropy and Exchange may be viewed as acting together
Diagnostics of Energy Terms

For $A > 3 \times 10^{-9} \text{ J/m}$

$E(\text{exchange}) + E(\text{anisotropy}) > E(\text{dipolar})$

Evidence that exchange is important driver of reversal
The Physics of Boomerangs: Ridge $\parallel H$ axis

First/smallest $H_R$:
1. Large down-flip avalanche at specific $H_R$: the first nucleated down-flip domain rapidly propagates, as the exchange coupling from the already down-flipped spins drives the rapid domain wall propagation.
2. The up-flip along first FORC is not by avalanche, as the up-flipping domains see a multi-domain background. Up-flip events occur through a range of $H$ fields.

Ridge forms in FORC, at smallest $H_R$, parallel to $H$

Down-flip: single large avalanche at specific $HR$
Up-flip: sequential flips over a range of $H$

$\leftarrow$ Wishbone: ridge parallel to $H_c$
The Physics of Boomerangs: Edge $\parallel H_R$ axis

Larger $H_R$s: Mirror image of ridge

1) Major loop up-flips by large avalanche at specific $H$, as the exchange coupling from the already up-flipped spins drives the domain wall propagation.

2) FORCs close to down-saturation: their down-flip was not accelerated by exchange, so it took place over a range of $H_R$s.

When these few down-flipped domains up-flip, they propagate across a largely down-flipped region, so the exchange drives the domain wall propagation in an avalanche largely similar to the avalanche of the major loop: occurs at single $H$.

Edge forms in FORC, larger $H_R$s, parallel to $H_R$

Down-flip: occurred at a range of $H_R$
Up-flip: single large avalanche at specific $H$
The Physics of Boomerangs: Negative Region

FORC slope increases: positive
FORC amplitude: ridge

FORC slope decreases because of magnetizing right shift of up-flip field: negative
FORC amplitude

FORC slope increases: positive
FORC amplitude: edge
Inter-grain Exchange A’ in TMC Samples

Inter-grain exchange $A'/A \sim 0.6$ observed in some NdFeB samples

High-coercivity ultrafine-grained anisotropic Nd–Fe–B magnets processed by hot deformation and the Nd–Cu grain boundary diffusion process

H. Sepehri-Amin $^{a,b}$, T. Ohkubo $^{a,b}$, S. Nagashima $^c$, M. Yano $^c$, T. Shoji $^c$, A. Kato, T. Schrefl $^d$, K. Hono $^{a,b,*}$

nets has been published. Here, we assumed that the exchange stiffness of the intergranular phase, with a slight segregation of Nd, is $8 \, \text{pJ m}^{-1}$, which is slightly less than that of the Nd$_2$Fe$_{14}$B phase ($12 \, \text{pJ m}^{-1}$). Fig. 12(b) shows
1. Left facing boomerang FORCs observed for $A > 2 \times 10^{-9}$ J/m

2. Boomerang transitions to right facing boomerang for $A < 2 \times 10^{-9}$ J/m

Decreasing $A$: Importance of dipolar fields grow
They generate interaction fields $H_b$
Edge moves SW, Ridge moves NE
Demagnetizing Arrays - Ridge

Wishbone/Dipolar vs. Boomerang/Exchange

Dipolar interaction dominates:

Wishbone FORC
- Ridge tracks Hc axis
- Edge tracks Hb axis
- High Hc end on Hc axis
- Ridge-edge angle < 90
- Domain growth is less important for reversal

Exchange dominates:

Boomerang FORC
- Ridge 45 from Hc axis, tracks H axis
- Edge 45 from Hb axis, tracks HR axis
- High Hc end away from Hc axis
- Ridge-edge angle = 90
- Domain growth is important for reversal
Wishbone/Dipolar to Boomerang/Exchange Transition
Wishbone/Dipolar to Boomerang/Exchange Transition

Demagnetizing planar

Magnetizing wall

Planar

Wall

A=1E-10

A=1E-9

A=6E-9

A=4E-9

A=1.8E-8

A=1E-8
Wishbone/Dipolar to Boomerang/Exchange Transition

\[ \Delta K_U \left( \times 10^6 \text{ J/m}^3 \right) \]

- Reversal as Isolated Elements
- Wishbone FORC Feature
- Calculated Domain Propagation Boundary
- Wishbone
- Left-Boomerang
- Right-Boomerang

\[ A' \ (\text{J/m}) \]
1. **Strong dipolar interactions**: Developed Mean Field + Local Fluctuations
   - Explained wishbone FORC; verified with controlled experiments
2. **Strong exchange**: Developed reversal by domain wall propagation picture
   - Explained boomerang FORC
   - Left facing boomerang FORCs observed for $A > 2 \times 10^{-9}$ J/m
   - Transition to right facing boomerang for smaller $A$
SUMMARY

1. Strong dipolar interactions: Developed Mean Field + Local Fluctuations
   - Explained wishbone FORC; verified with controlled experiments
2. Strong exchange: Developed reversal by domain wall propagation picture
   - Explained boomerang FORC
   - Left facing boomerang FORCs observed for $A>2 \times 10^{-9}$ J/m
   - Transition to right facing boomerang for smaller $A$