Controlled limits of FORC theory: Mean field and Nucleation

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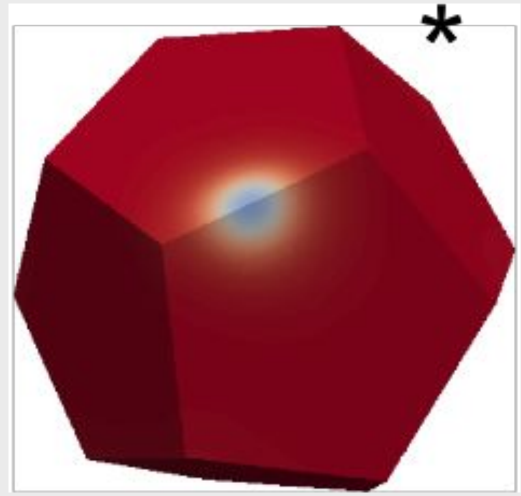
Scientific Reports 4, 4204 (2014)

Thanks to Alexandru Stancu and the Iasi group

The FORC framework

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.



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

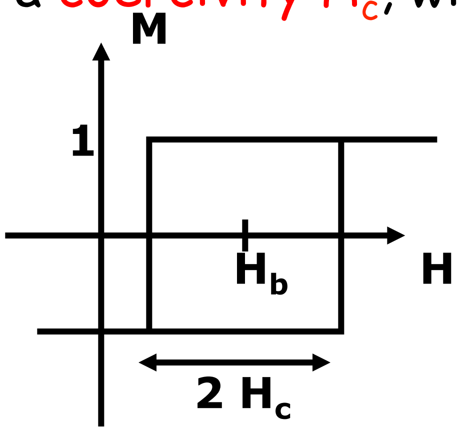
The FORC technique characterizes a system via the distribution the local coercivities *Hc* <u>and</u> in addition the local interaction fields *Hb*

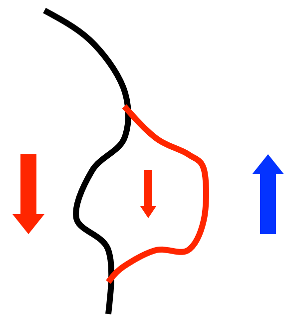
FORC captures these as a joint distribution *ρ(Hc,Hb)* instead of a product of two distributions, thus capturing underlying correlations

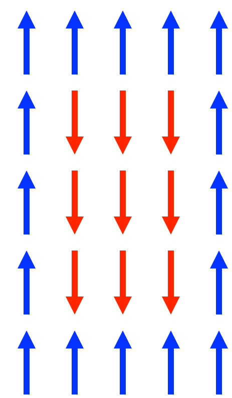
FORC basics – Building on Preisach modeling

- 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 $\rho(Hc,Hb)$



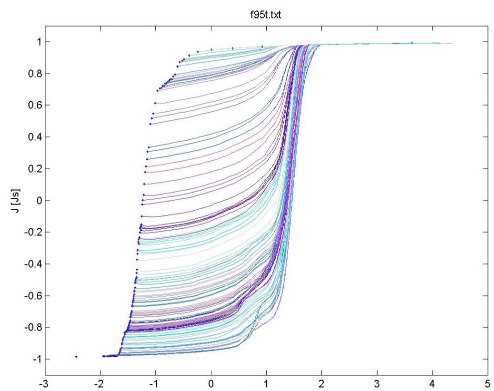


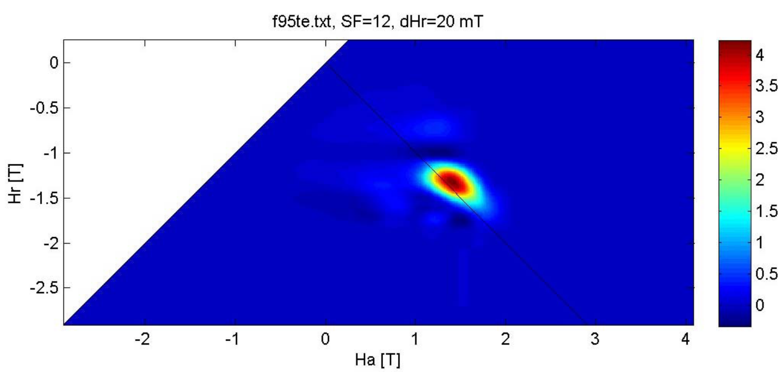


- Mathematical foundation: Mayergorz 1987
- Phenomenology, modelling: Dellatorre, Vajda, Cardelli
    - Assume coercivity & bias field distribution $\rho(Hc,Hb)$
    - Fit parameters of $\rho(Hc,Hb)$ to reproduce hysteresis loop

FORC basics

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 - ….

FORC in the literature

Characterizing interactions in fine magnetic particle systems using first order reversal curves
CR **Pike**, AP Roberts, KL Verosub - Journal of Applied Physics, 1999 - scitation.aip.org
... 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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Outline

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

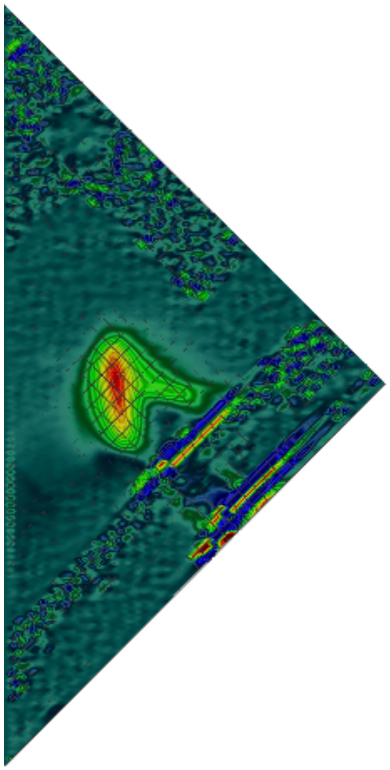
1.3 Test/verify theory experimentally on nanoparticle arrays

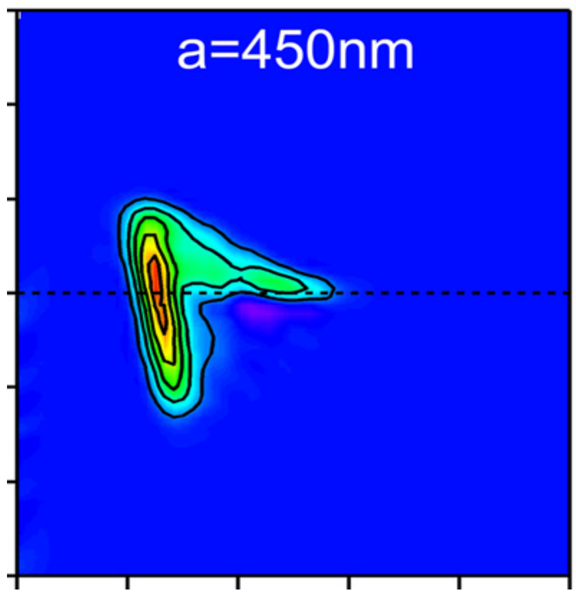
2. Strong exchange interaction:

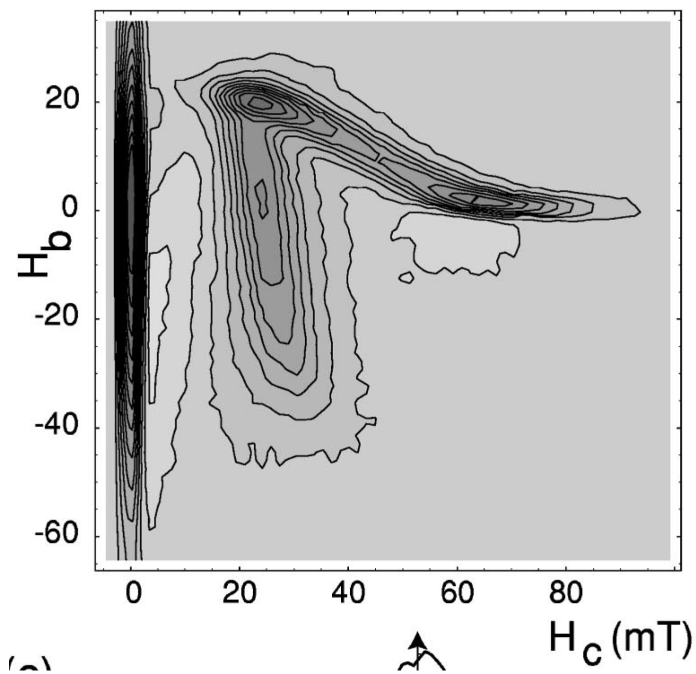
2.1 Analyze FORC diagrams in terms of nucleation

2.2 Establish phase diagram

Wishbone FORCs: Paradigm in Many Classes of Magnets







Nd2Fe14B, Schrefl, GTZ JAP (2012)

Nanowire arrays, Stancu JAP (2013)

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: $H_{int}^i = \alpha M(H)$

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

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

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

Rotate from (H,HR) to (Hb,Hc) axes: $H_B=(H_{up}+H_{dn})/2$ $H_C=(H_{up}-H_{dn})/2$

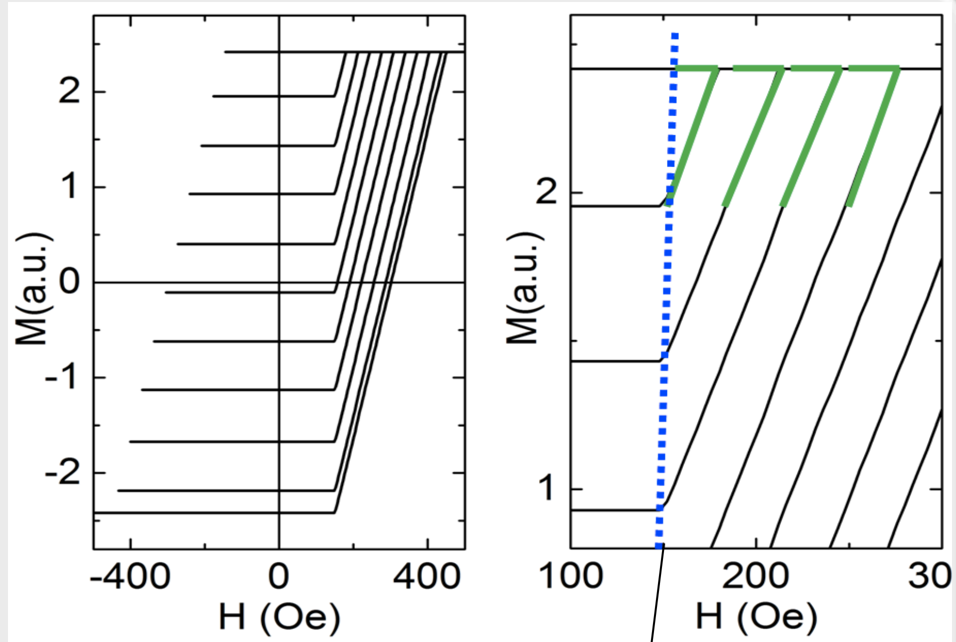
Non-Interacting Arrays – Ridge || Hc axis

FORC ρ 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:

   $-d(dM/dH)/dH_R = \rho = 0$



H_k(min)

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 Hc**: $\rho(Hb=0,Hc)=D(H_K)$, the coercivity distribution

Demagnetizing Arrays – Ridge || Hc axis

$H_{tot}=H+\alpha M(H)$ $\alpha<0$

$P(H_k^{min})$ unmatched (min)

$H_{dn}^{min}=-H_K^{min}-\alpha M_S$

$H_{up}^{min}=H_K^{min}-\alpha M_S$

$H_B=(H_{up}+H_{dn})/2$ $H_C=(H_{up}-H_{dn})/2$

Low H_C end shifted by

$\Delta H_B=-\alpha M_S$ $\Delta H_C=0$

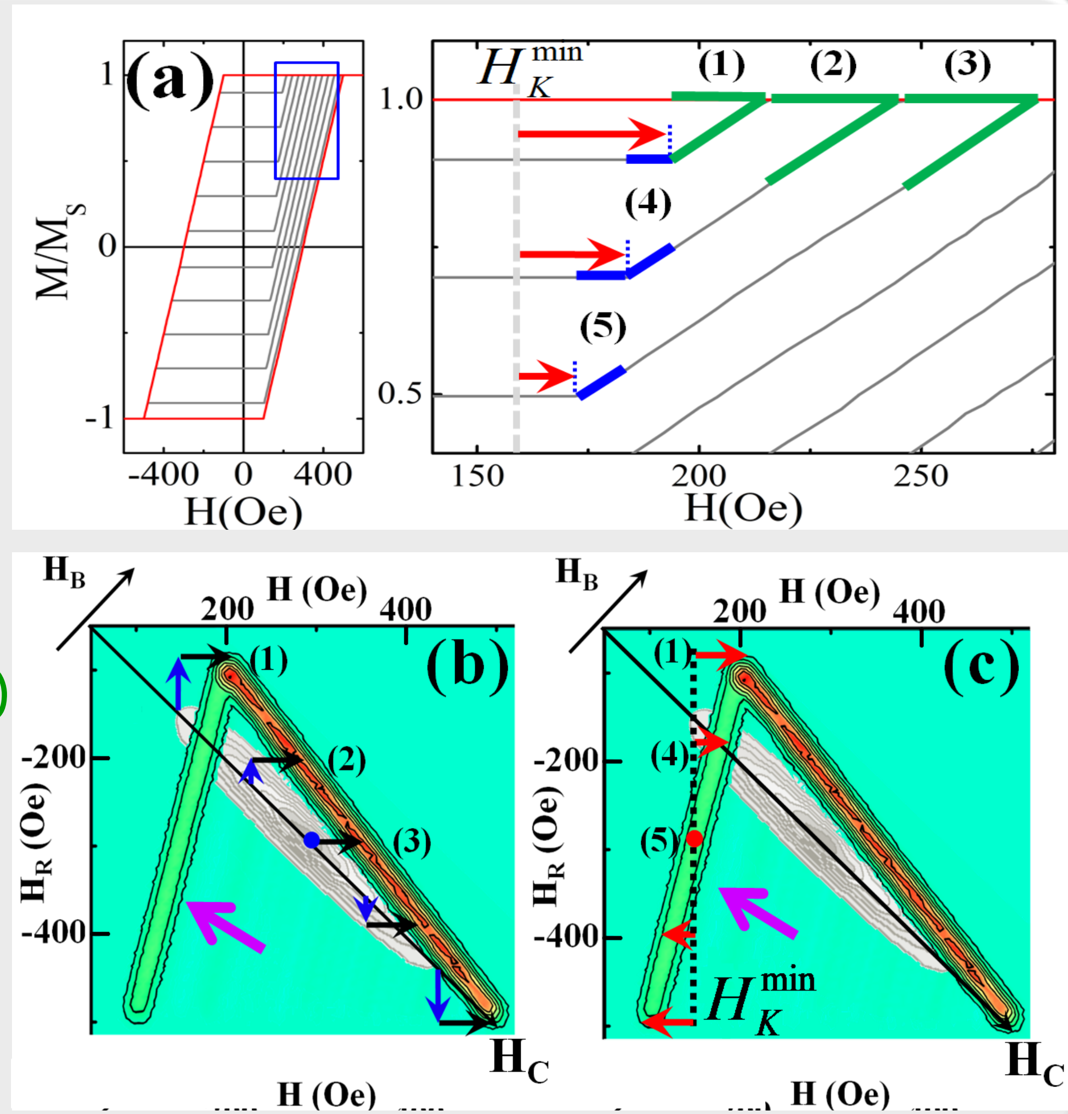
$P(H_k^{max})$ unmatched (max)

$H_{dn}^{max}=-H_k^{max}+\alpha M_S$

$H_{up}^{max}=H_K^{max}-\alpha M_S$

High H_C end shifted by

$\Delta H_B=0$ $\Delta H_C=-\alpha M_S$



Demagnetizing Arrays – Ridge

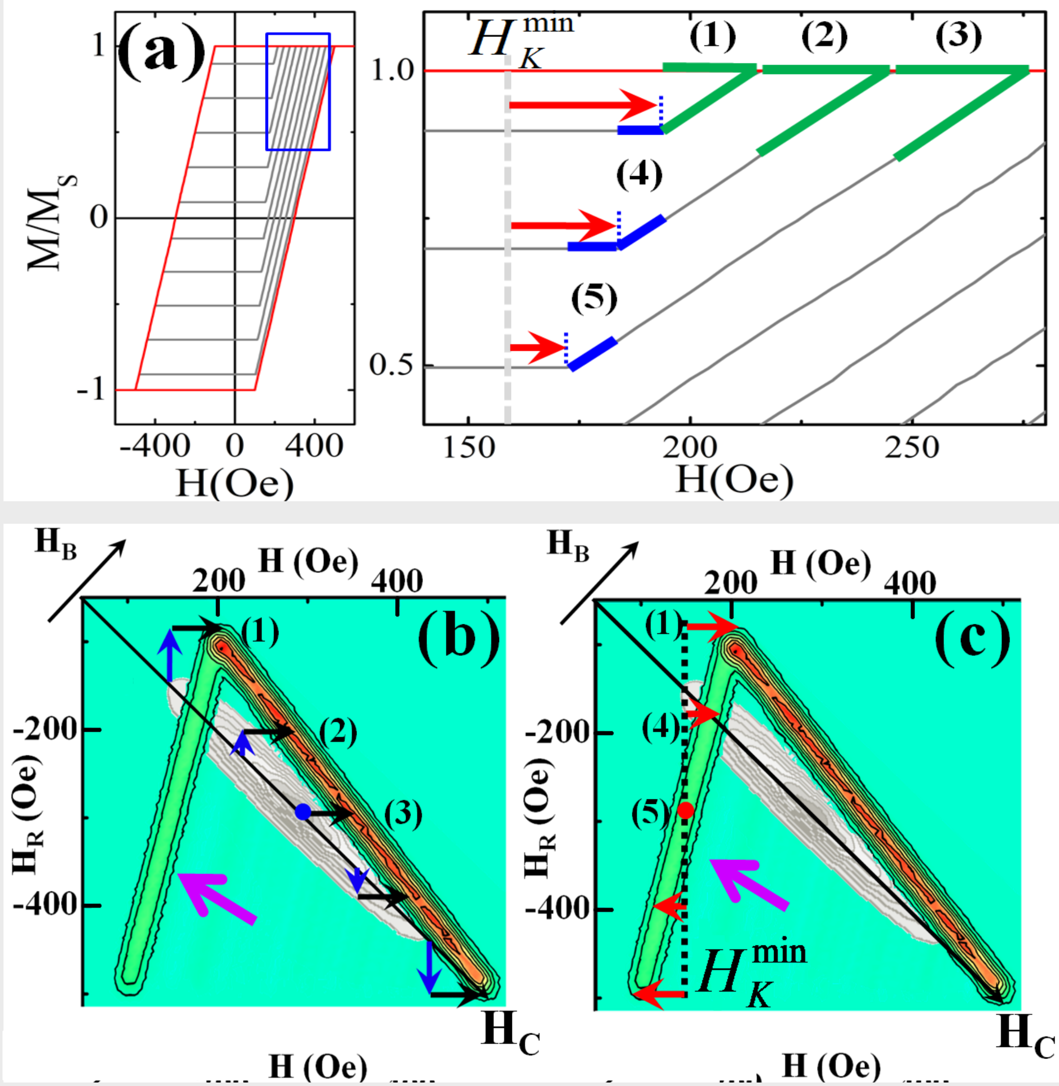
Last upflip fields shifted by interaction

$H_{tot}=H+\alpha M(H)$ $\alpha<0$

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 $P(H_k^{min})$ first to upflip

-No interaction: 1st upflips at $H=H_k^{min}$

Same matched field every FORC, $\rho=0$

-Interaction: 1st upflip fields shifted, thus unmatched

Top FORC

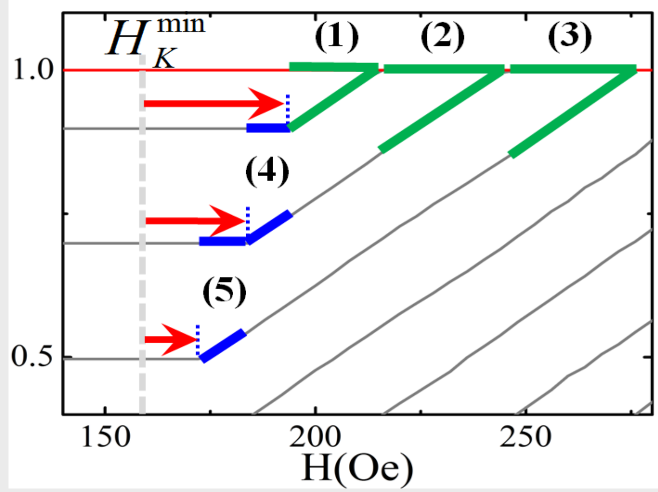
$H=H_K^{min}-\alpha M_S$, $H_R=-H_K^{min}-\alpha M_S$

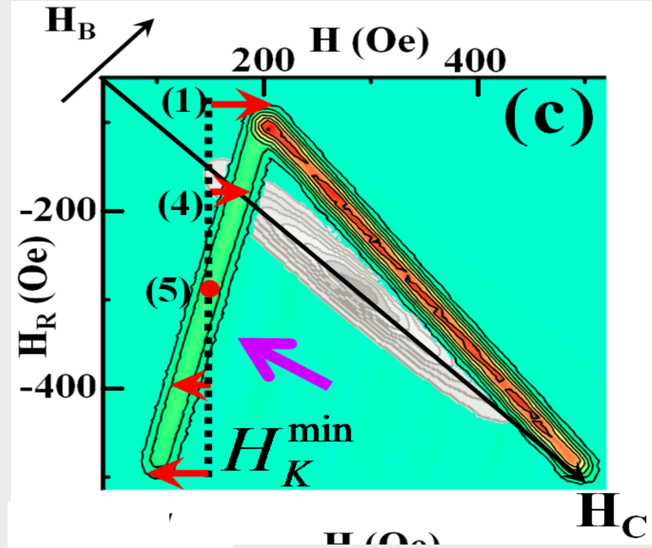
Bottom FORC:

$H=H_K^{min}+\alpha M_S$, $H_R=-H_K^{max}+\alpha M_S$

First upflip fields are not matched

- A $\rho>0$ edge forms by interaction,
- Edge is tilted





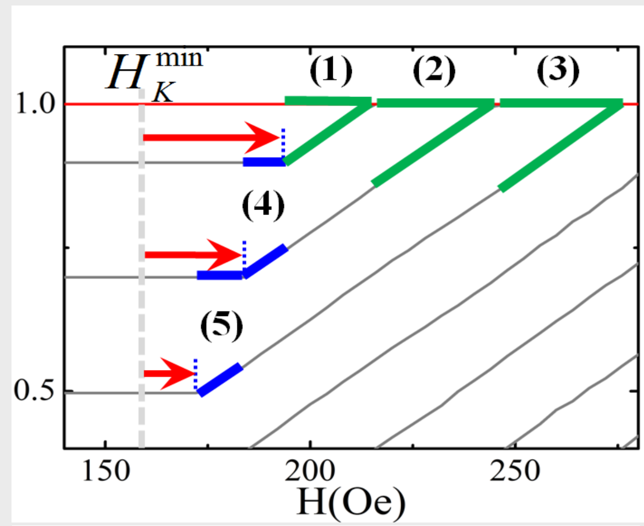
Demagnetizing Arrays–Negative FORC Region

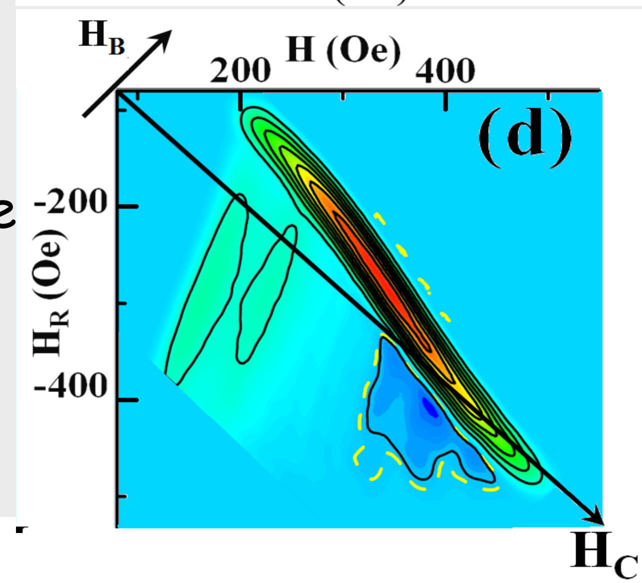
Many FORCs exhibit negative regions

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/dHs now decrease

Negative FORC ρ (only) in high H_K region





Demagnetizing Arrays – Mean Field Theory

Ridge: **unmatched last upflip,**

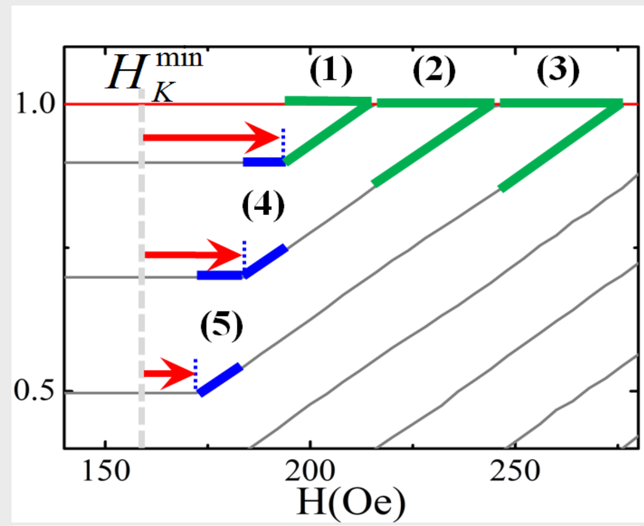
Represents Hk coercivity distribution

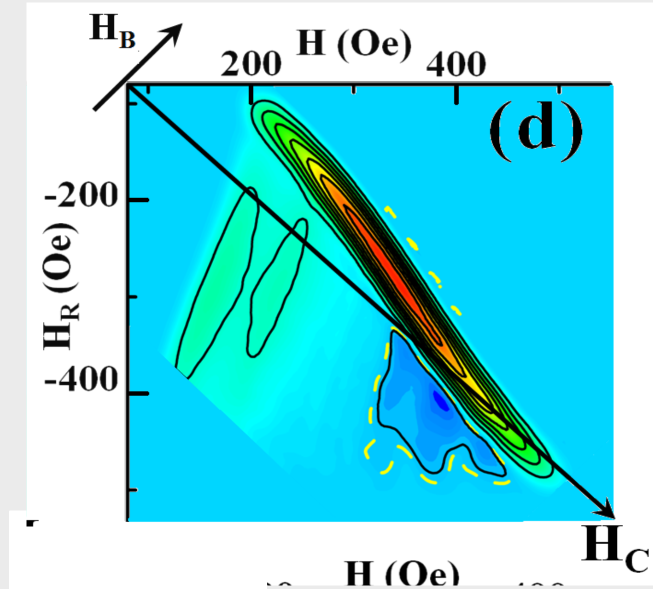
Edge: **unmatched first upflip**

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 $D(H_K)$

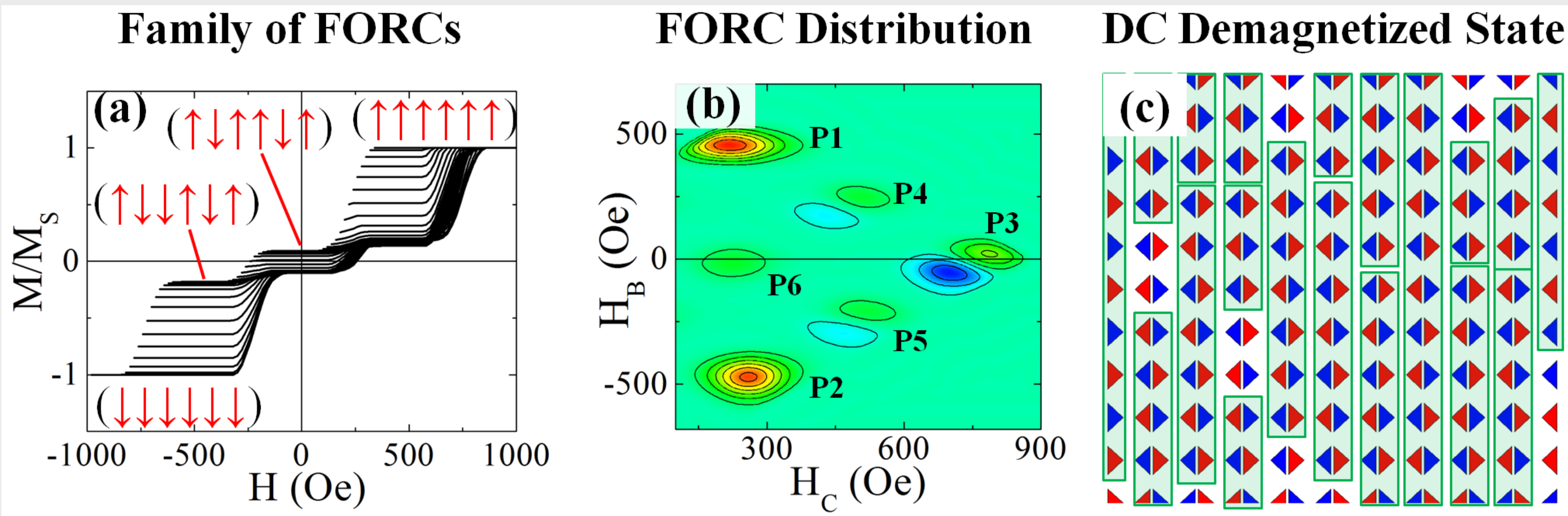




Beyond Mean Field: Nearest Neighbor Interaction

Controlled expansion around Mean Field: local fluctuations

Include nearest neighbor non-mean field terms



(dn1) positive saturation → checkerboard (↑ ↑ ↑ → ↑ ↓ ↑ : H_{int}=2$H_{n.n.}$)

(dn2) checkerboard → negative saturation (↓ ↑ ↓ → ↓ ↓ ↓ : H_{int}= -2$H_{n.n.}$)

(dn3) frust. checkerboard → frust. checkerboard (↓ ↑ ↑ → ↓ ↓ ↑ : H_{int}=0)

Experimental Test of Mean Field Theory

(K. Liu, R. Dumas)

Polycrystalline Co ellipses

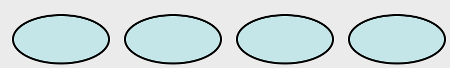
E-beam lithography

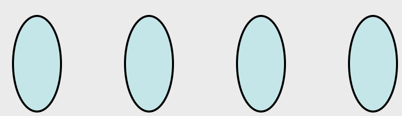
Liftoff technique

Major/minor axis: 220/110nm

Created 50x50 micron array of ellipses

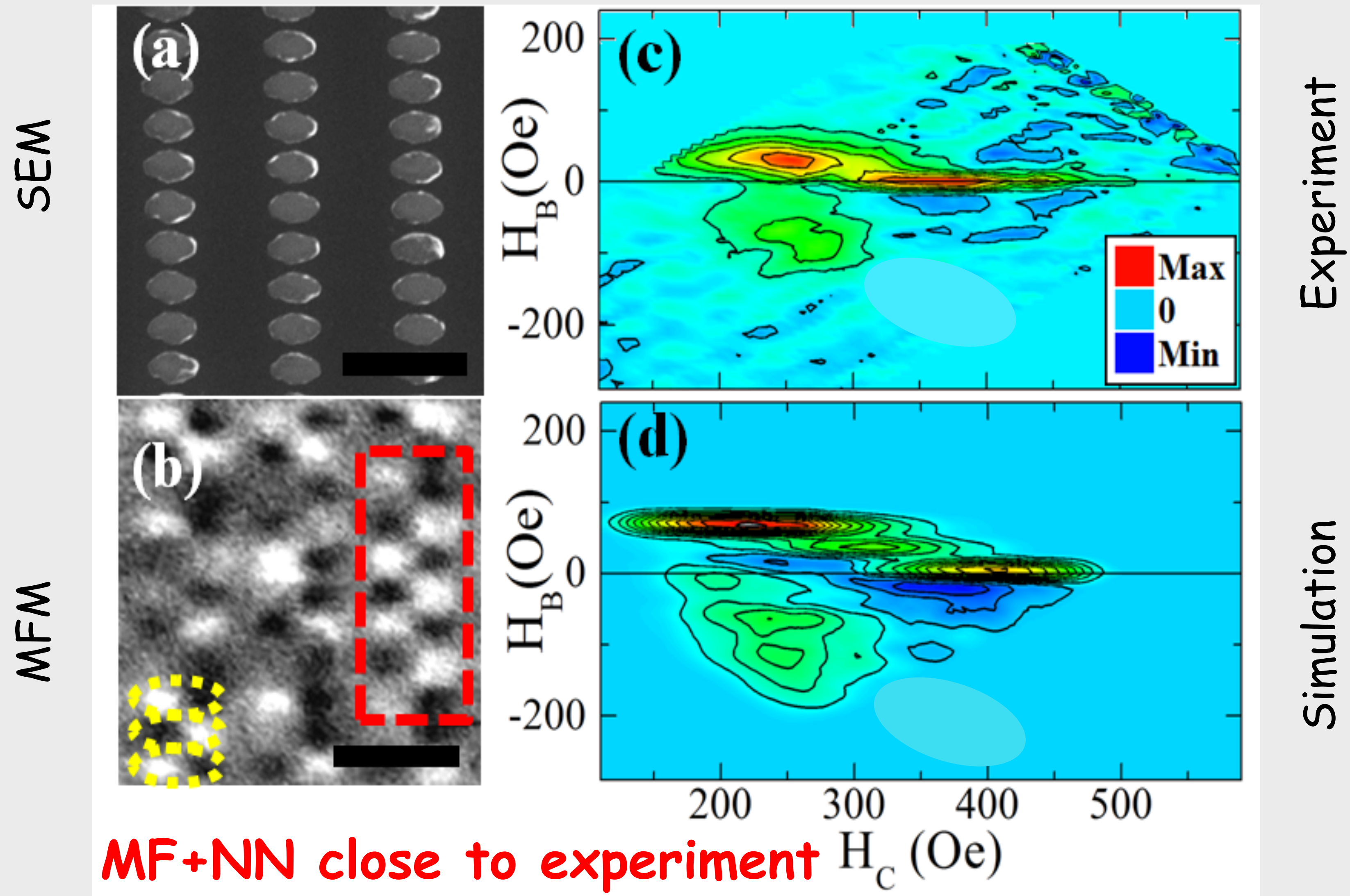
Measure middle of the array to avoid edge effects

- 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 Hk coercivity distribution

- Edge: Represents interaction parameters

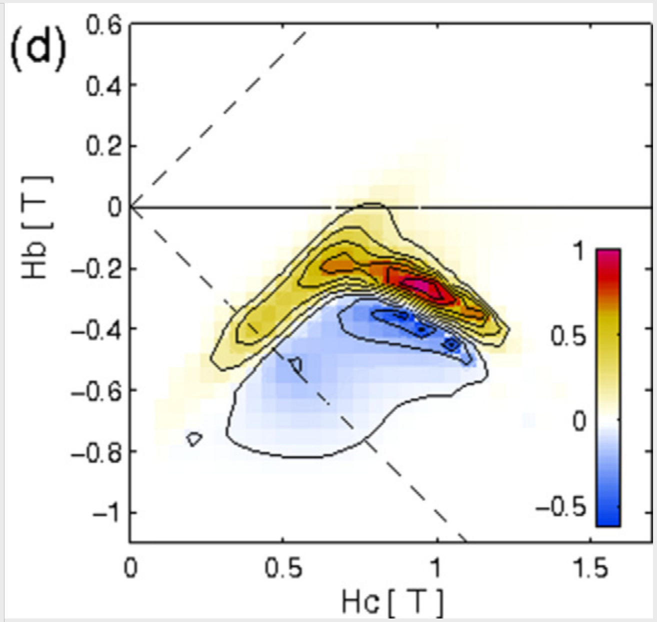
3. Developed controlled fluctuation expansion around Mean Field

4. Verified on controlled arrays of nanoparticles

2. Strong Exchange: Mean Field not appropriate: Boomerang FORCs

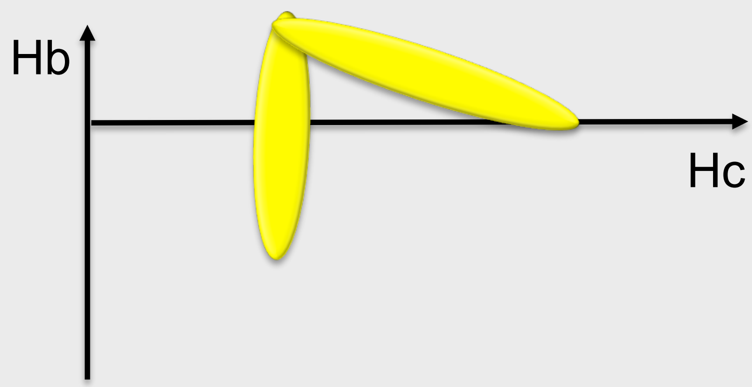
Another two-branched FORC is observed on certain FeNdB permanent magnets where exchange is much stronger.

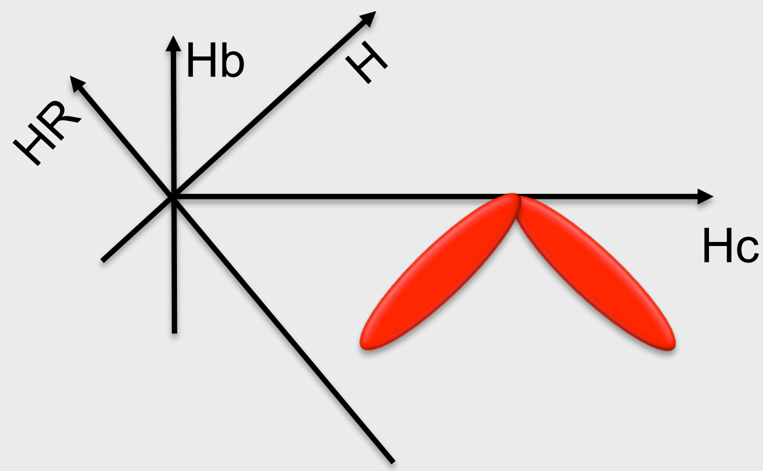
We termed these FORCs “boomerangs”
Boomerangs can be unmasked in large number of FORCs with deshearing



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

5μx5μx1μ sample with 50nmx100x100nm and 50nmx200x200nm grains ~ 10,000 - 50,000 grains

Use OOMMF code

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

Parameters need to be scaled:

- Ms 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)

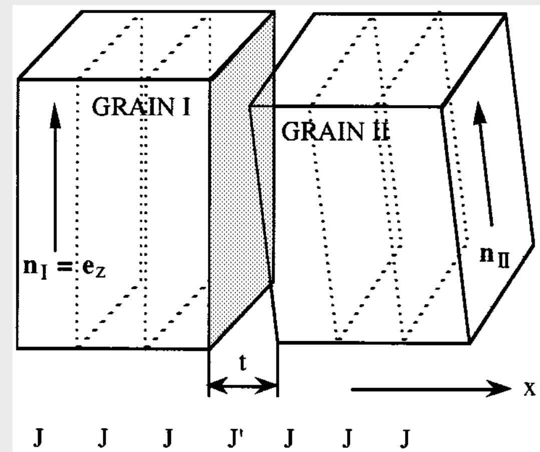
Scaling A

IEEE TRANSACTIONS ON MAGNETICS, VOL. 37, NO. 4, JULY 2001

Grain-Boundary Micromagnetism

R. Skomski, H. Zeng, and D. J. Sellmyer

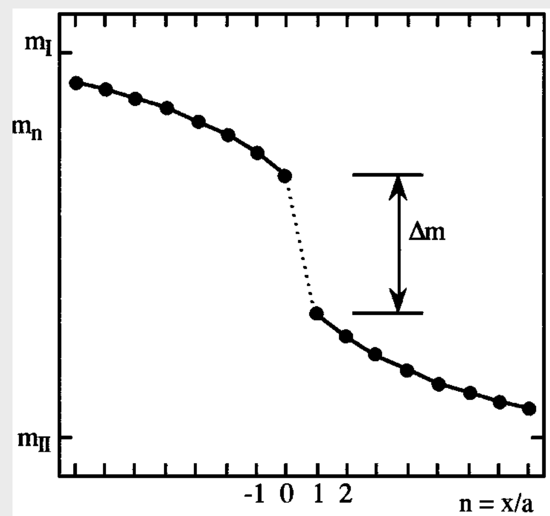
$$J_{\text{eff}} = L^2\sqrt{AK}/\left(1+\frac{t\sqrt{AK}}{2A'}\right)$$



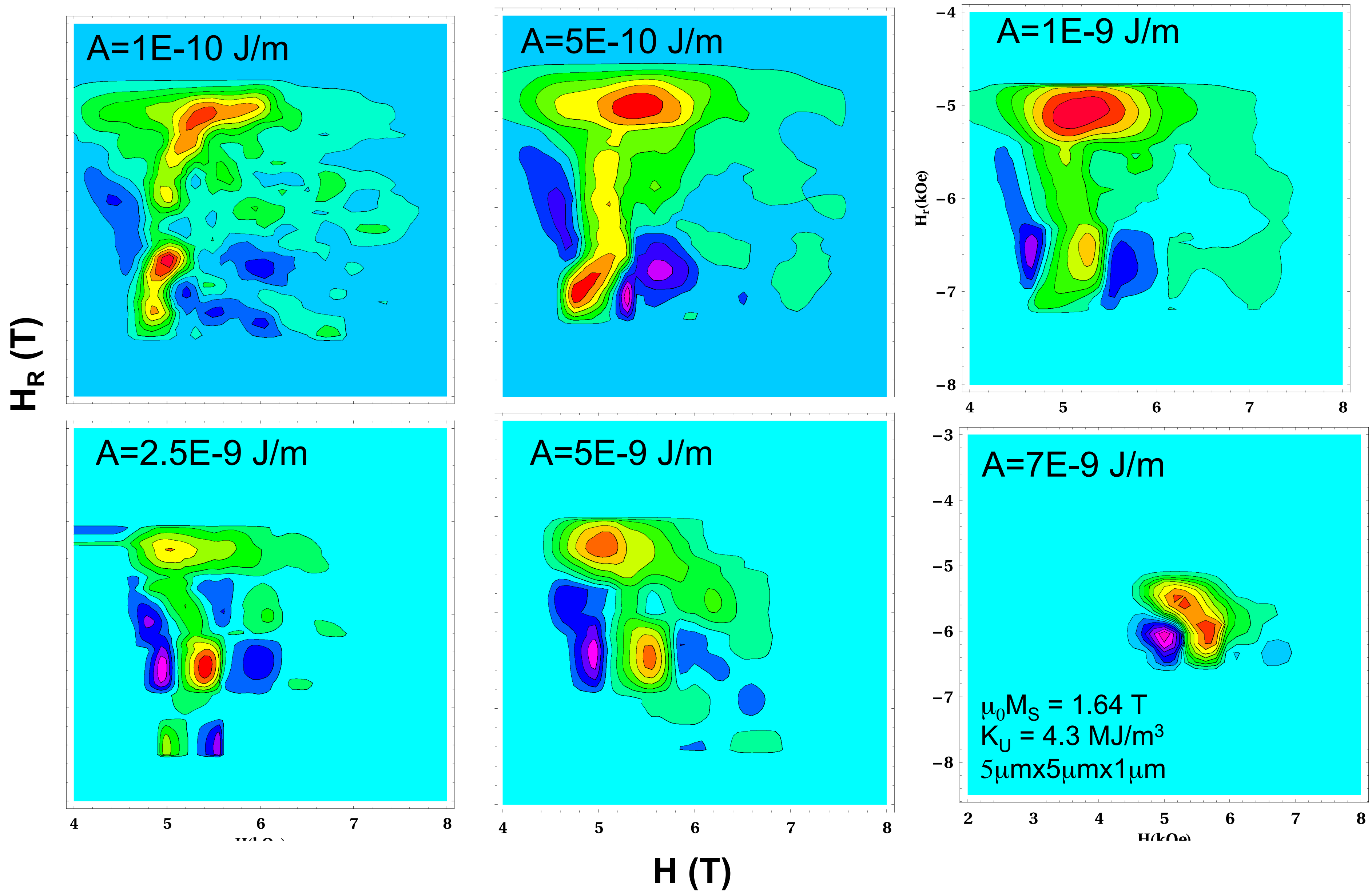
For 50-200 nm grain size and t~1-2nm,

A(scaled) can sweep **[0.1-10]x10^{-9} J/m**

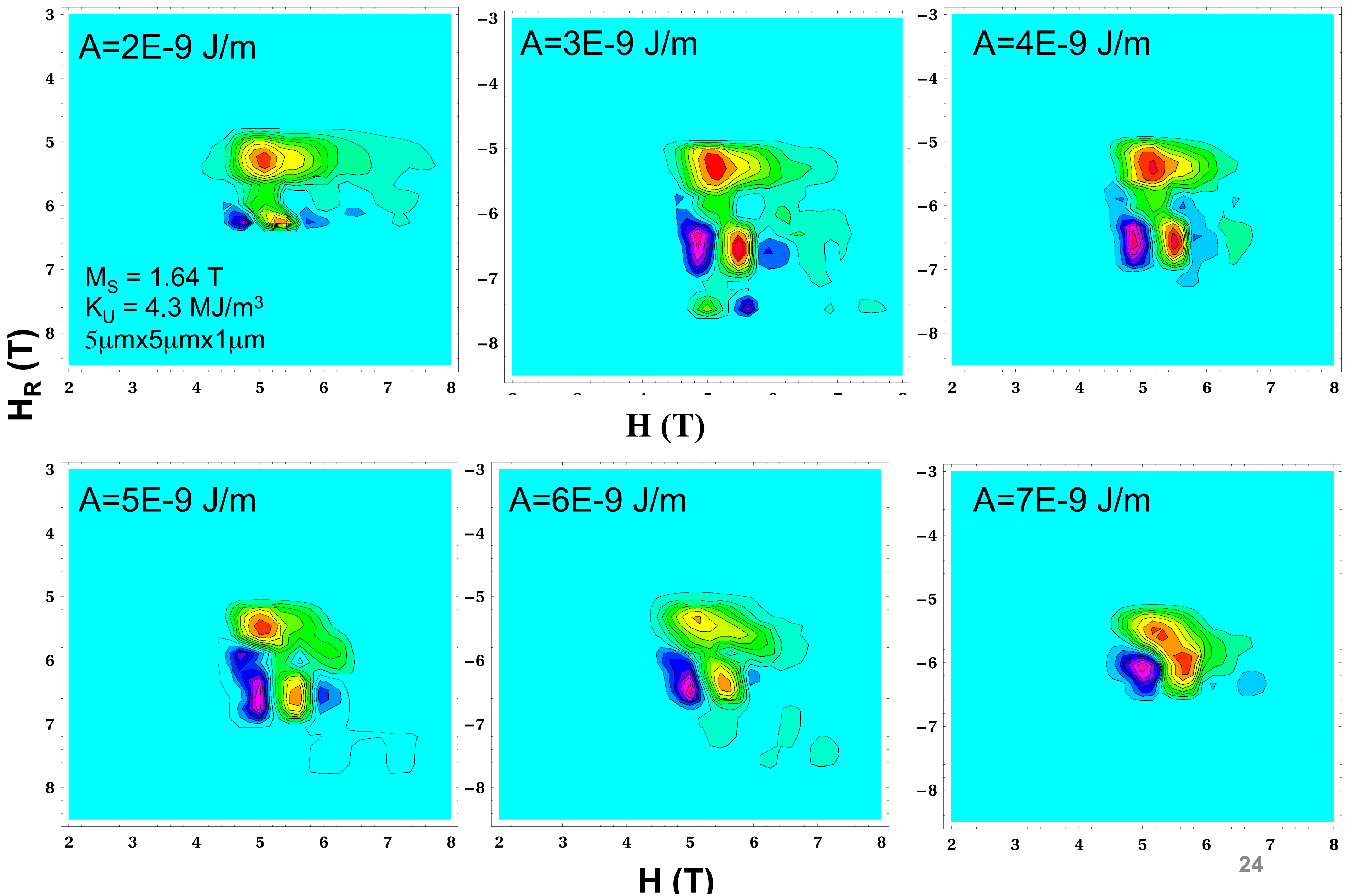
for A'(microscopic)/A(microscopic) ~0.05-0.5



Sweeping with Exchange A



Sweeping with Exchange A – Zooming to Transition

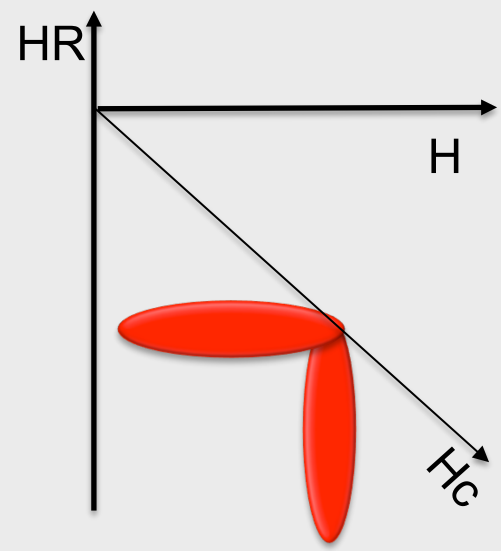


Strong Exchange creates Reversal by Domain growth, Explains boomerangs

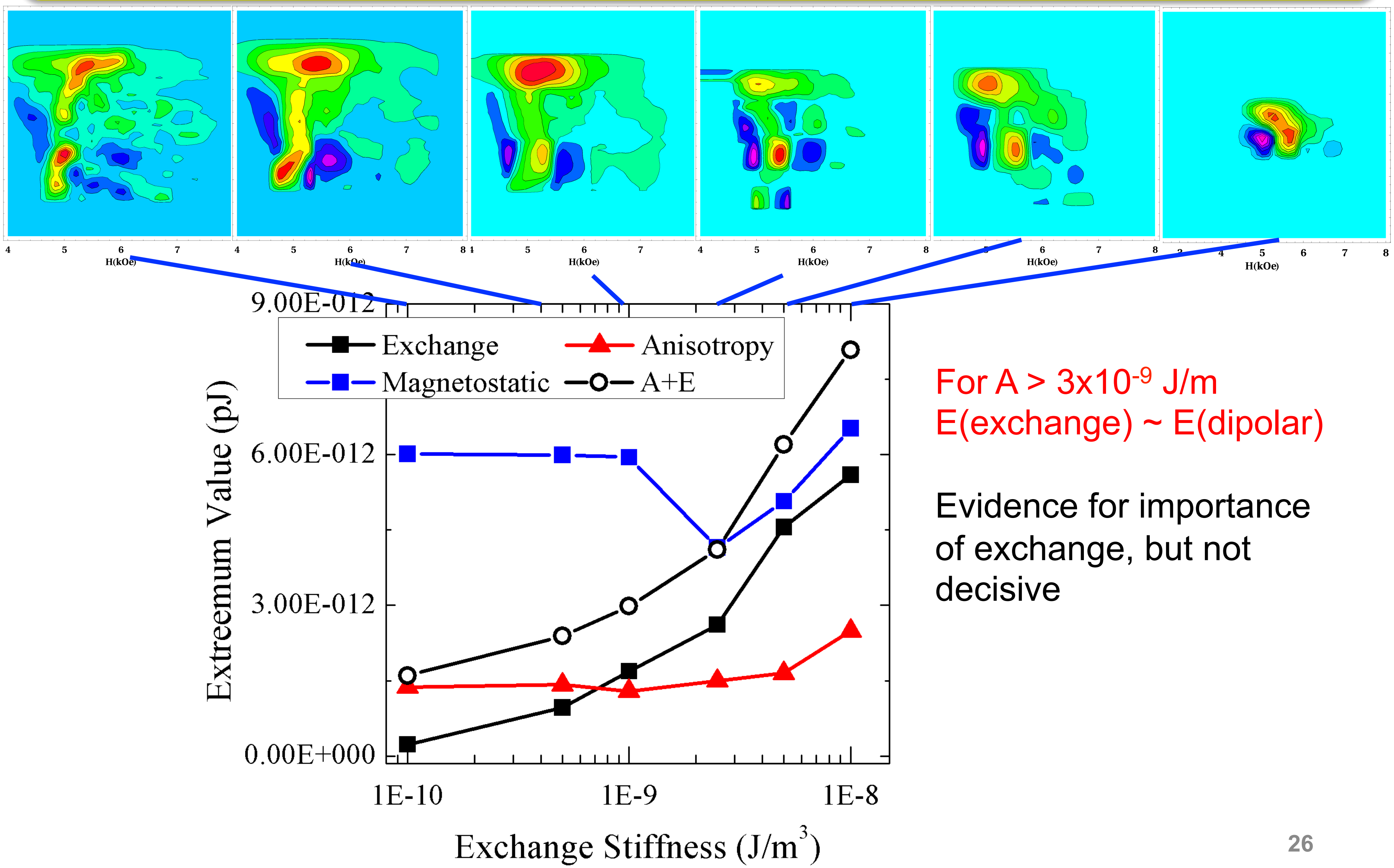
- Left facing boomerang FORCs observed for A(scaled) >$2x10^{-9}$ J/m

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

- Relatively strong inter-grain exchange, A'/A > 0.1
  needed to explain boomerang FORCs



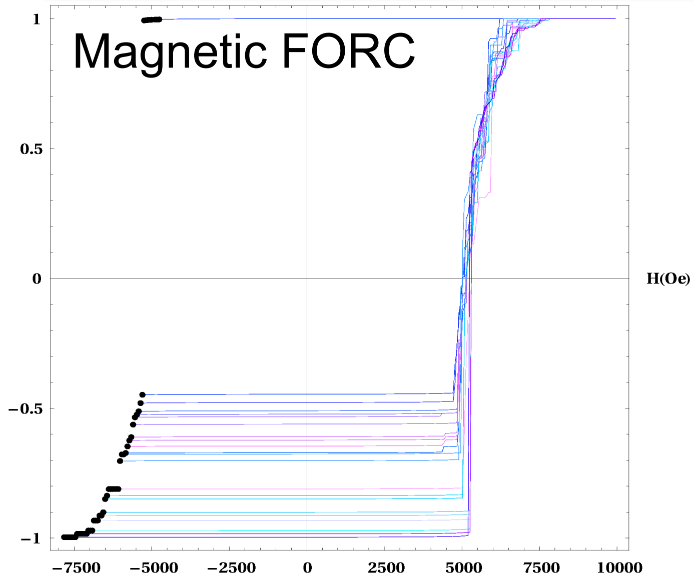
Diagnostics of Energy Terms

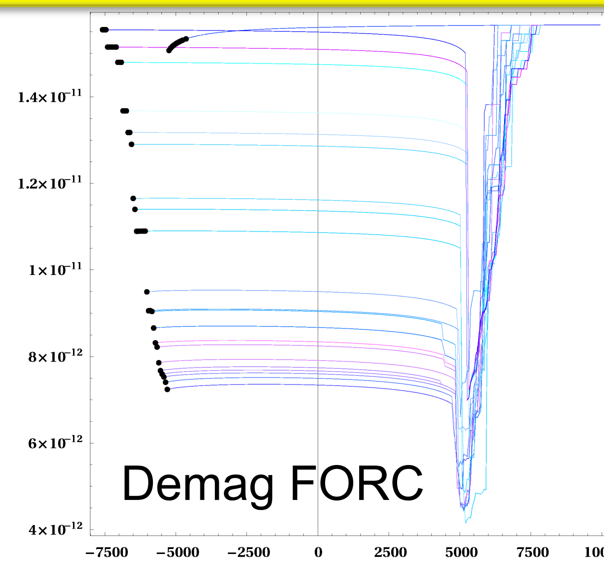


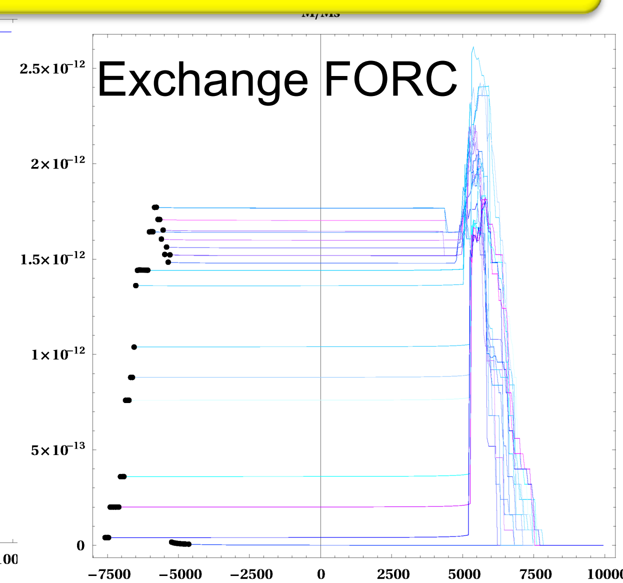
For A > 3x10^{-9} J/m
E(exchange) ~ E(dipolar)

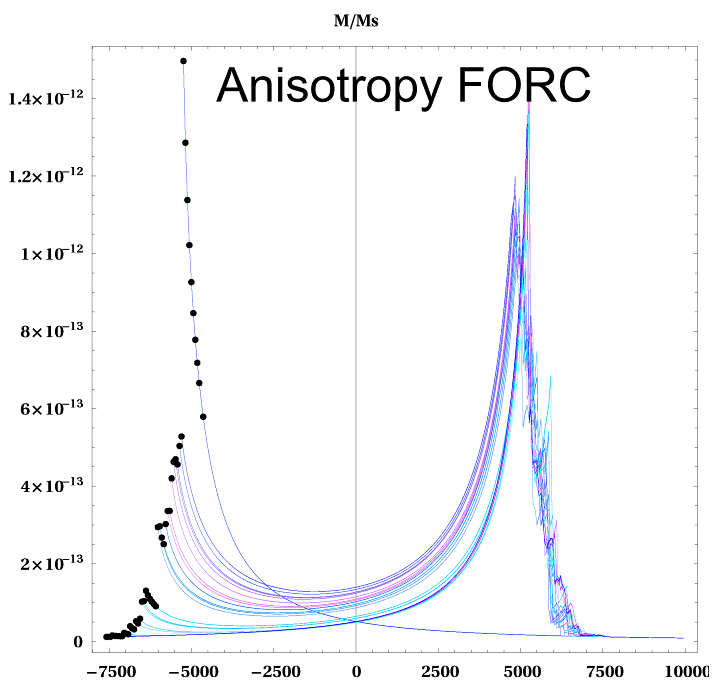
Evidence for importance of exchange, but not decisive

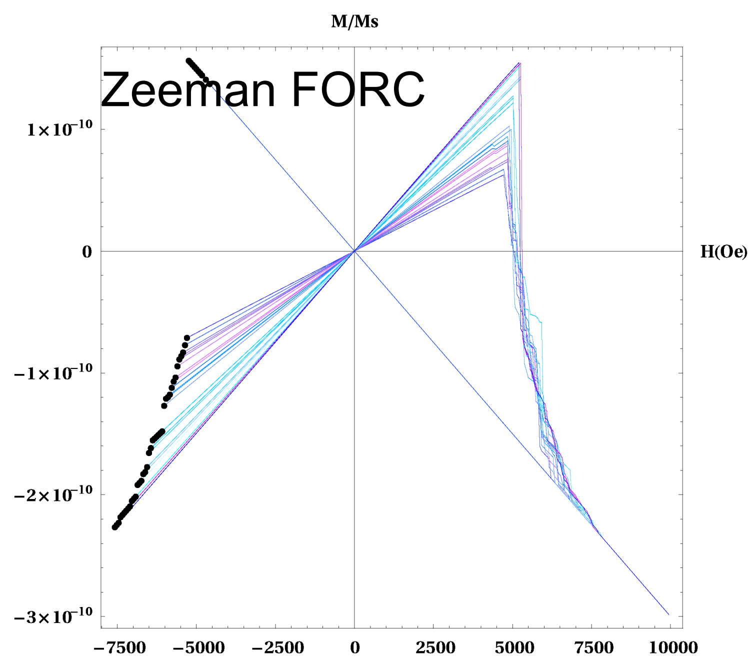
Energy FORCs





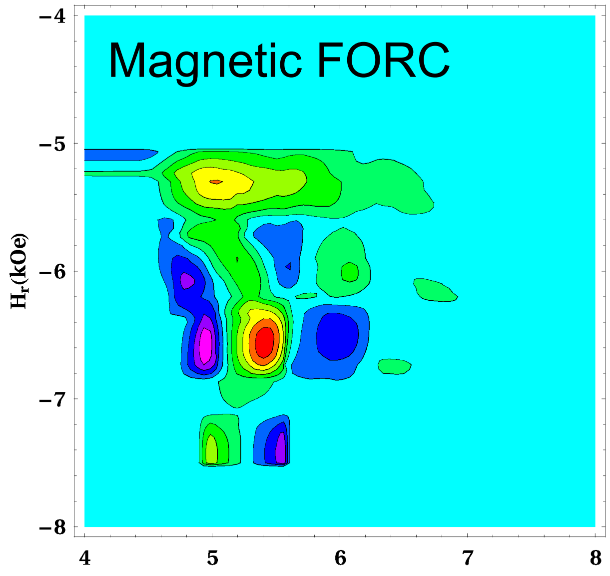


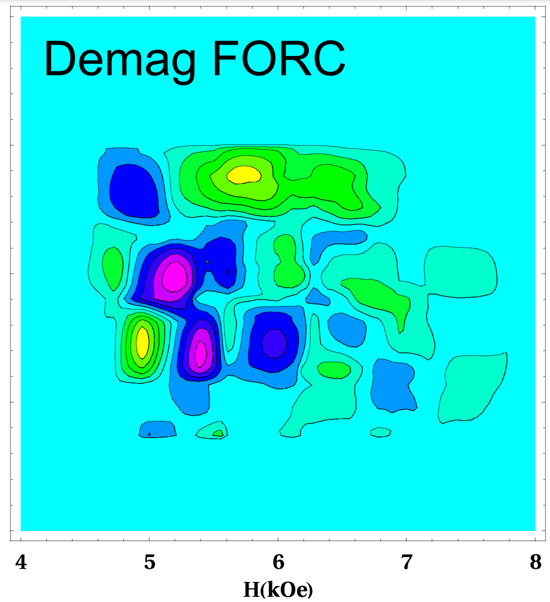


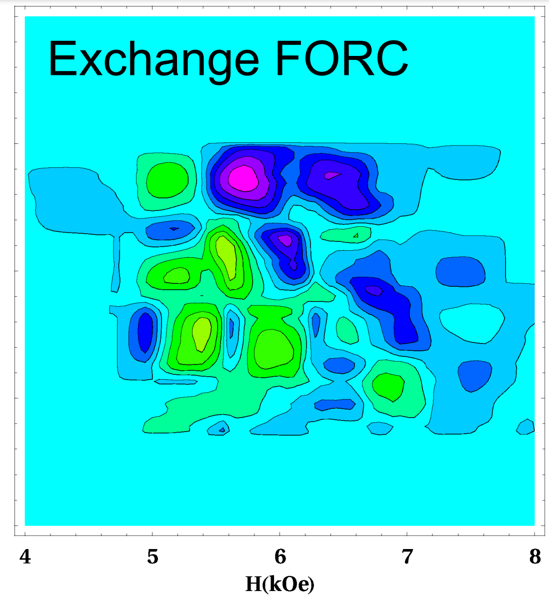


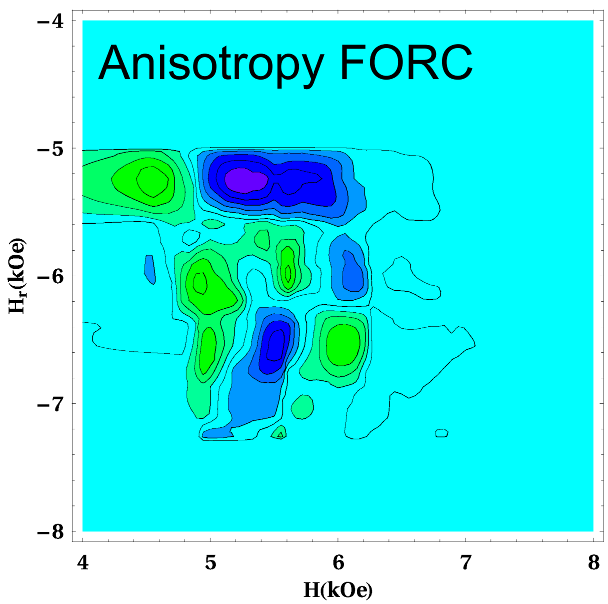
A=2.5E-9 J/m

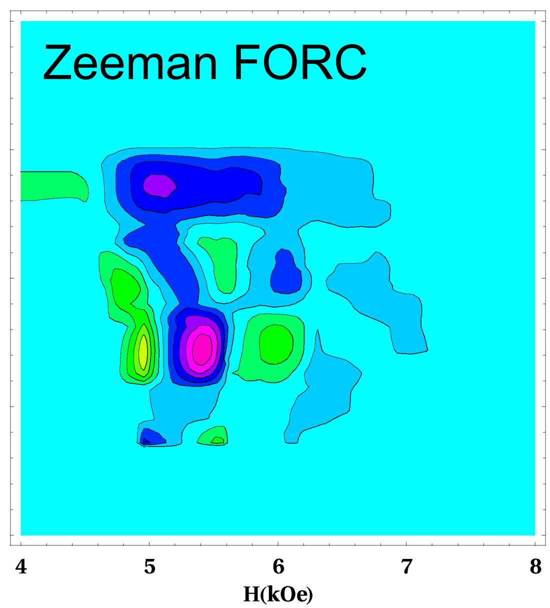
Energy FORC Diagram







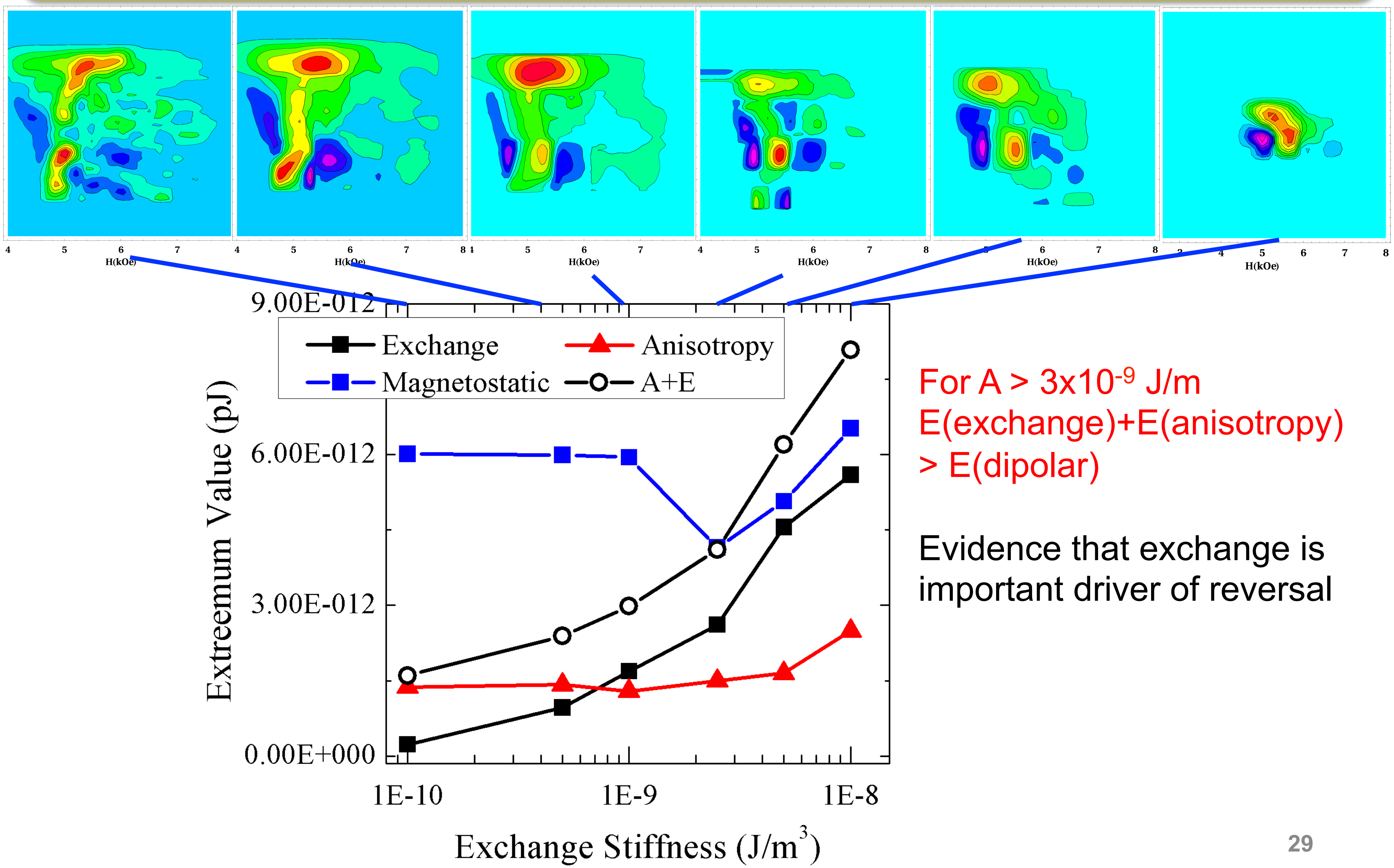




The FORC of Anisotropy closely tracks the FORC of the Exchange

Anisotropy and Exchange may be viewed as acting together

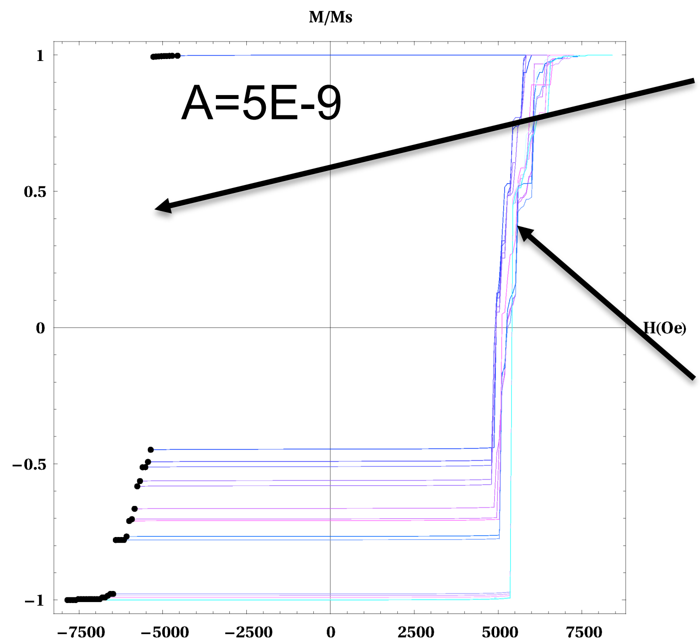
Diagnostics of Energy Terms



For A > 3×10^{-9} J/m
E(exchange)+E(anisotropy) > E(dipolar)

Evidence that exchange is important driver of reversal

The Physics of Boomerangs: Ridge || 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

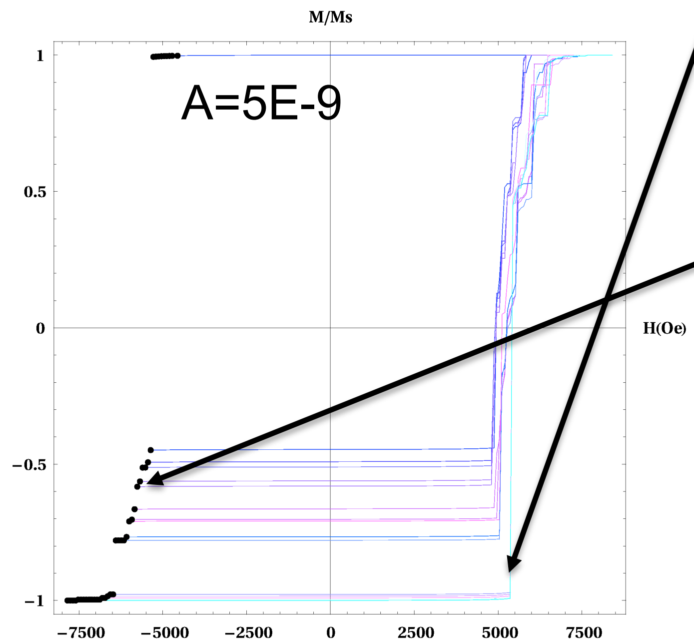
(2) The up-flip along first FORC is not by avalanche, as the up-flipping domains see a multi-domain background. Up-flip events occur through a range of H fields

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

⟷ Wishbone: ridge parallel to Hc

The Physics of Boomerangs: Edge || H_R axis



Larger H_Rs: 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

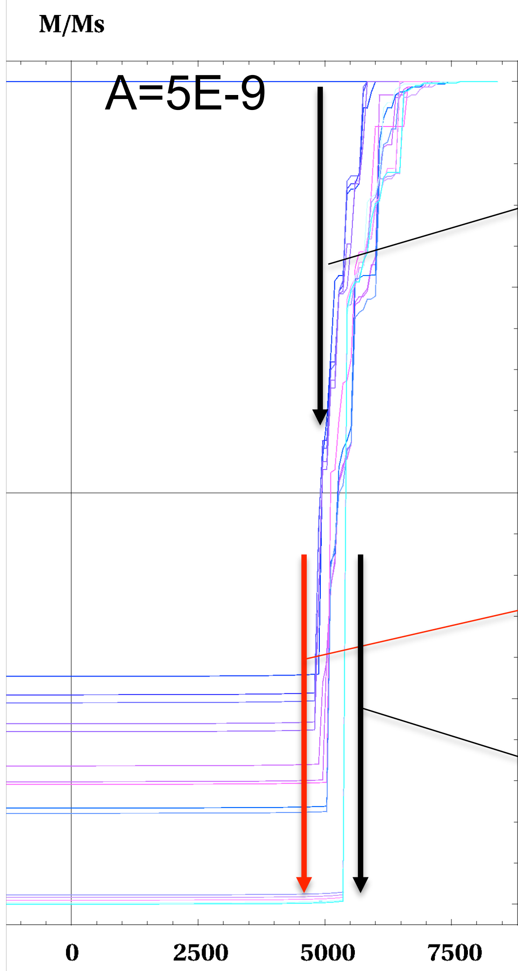
(2) FORCs close to down-saturation: their down-flip was not accelerated by exchange, so it took place over a range of H_Rs.

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_Rs, parallel to H_R

Down-flip: occurred at a range of HR
Up-flip: single large avalanche at specific H

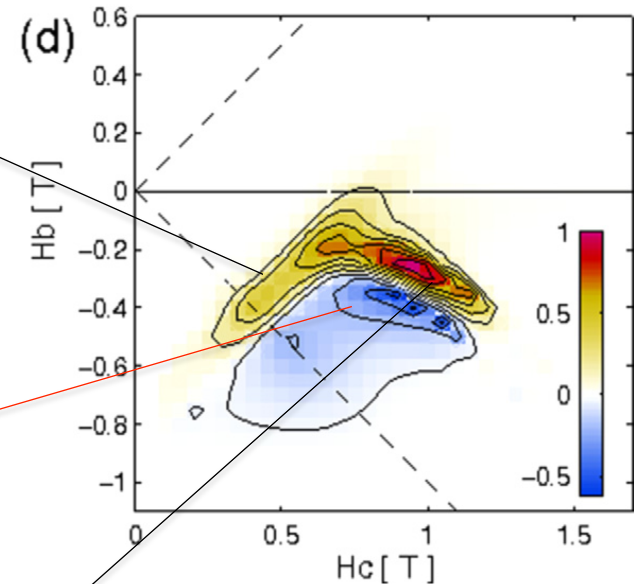
The Physics of Boomerangs: Negative Region



FORC slope increases: positive FORC amplitude: ridge

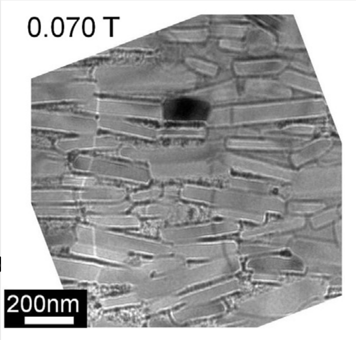
FORC slope decreases because of magnetizing right shift of up-flip field: negative FORC amplitude

FORC slope increases: positive FORC amplitude: edge



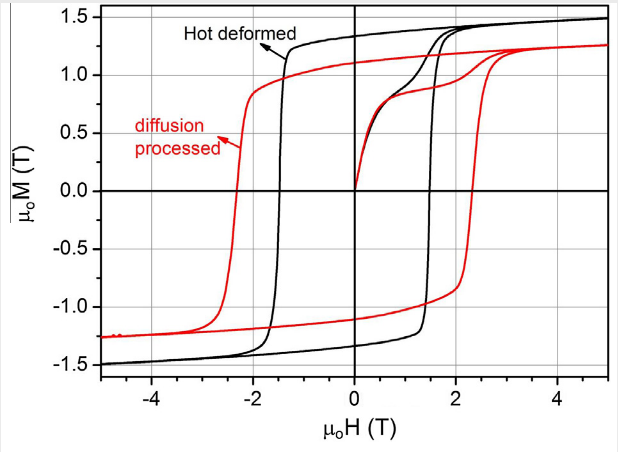
Inter-grain Exchange A’ in TMC Samples

Inter-grain exchange A’/A ~ 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,*]

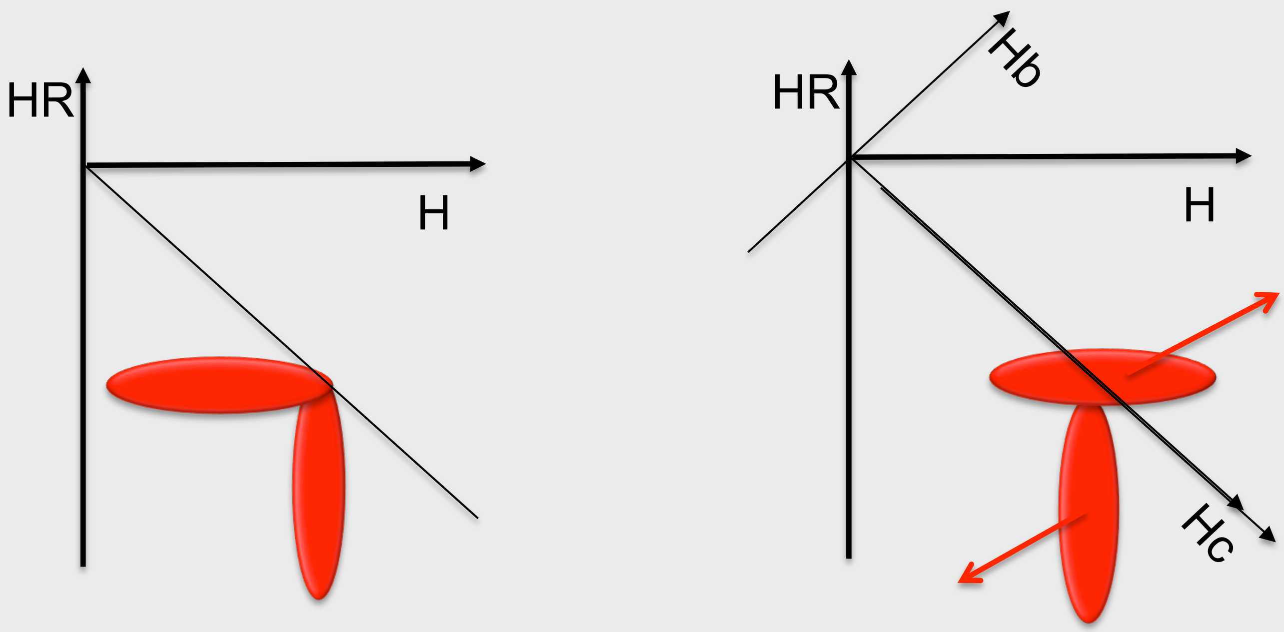


nets has been published. Here, we assumed that the exchange stiffness of the intergranular phase, with a slight segregation of Nd, is 8 pJ m^{-1}, which is slightly less than that of the $Nd_2Fe_{14}B$ phase (12 pJ m^{-1}). Fig. 12(b) shows

Boomerang Transition

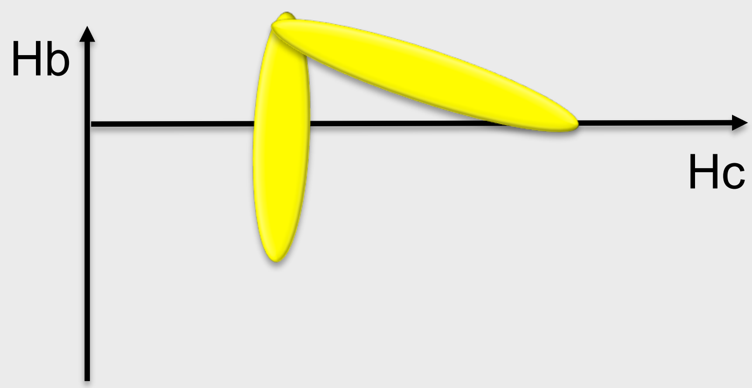
1. Left facing boomerang FORCs observed for $A>2x10^{-9}$ J/m

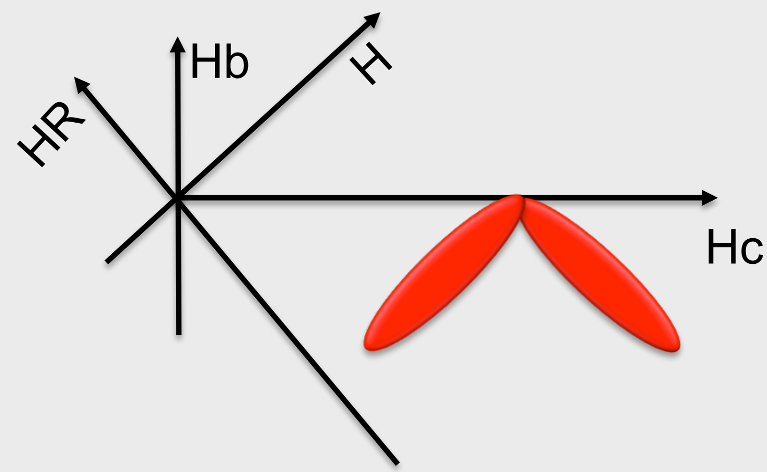
2. Boomerang transitions to right facing boomerang for $A<2x10^{-9}$ J/m



Decreasing A: Importance of dipolar fields grow
They generate interaction fields Hb
Edge moves SW, Ridge moves NE

Wishbone/Dipolar vs. Boomerang/Exchange





<u>**Dipolar interaction dominates:**</u>

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

<u>**Exchange dominates:**</u>

Boomerang FORC

Ridge 45 from Hc axis, tracks H axis

Edge 45 from Hb axis, tracks HR axis

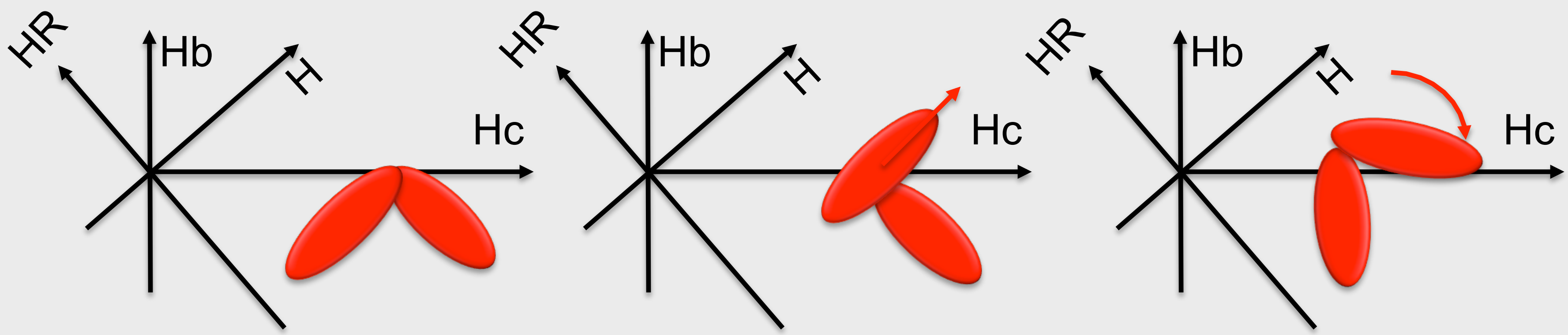
High Hc end away from Hc axis

Ridge-edge angle = 90

Domain growth is important for reversal

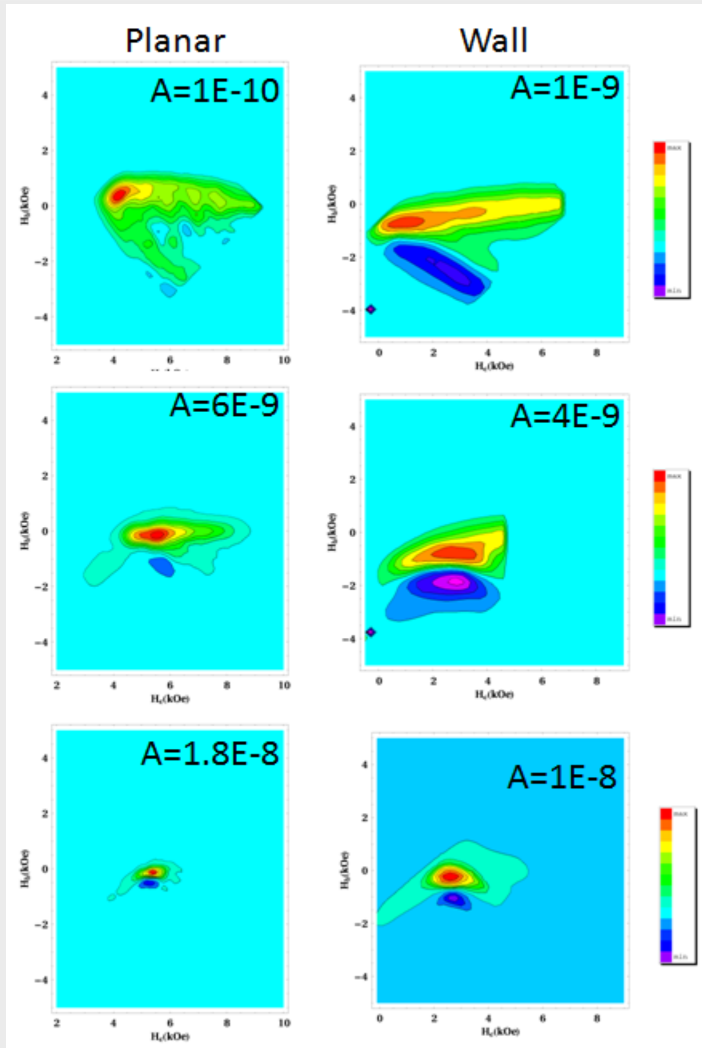
Wishbone/Dipolar to Boomerang/Exchange Transition

Exchange dominates Dipolar dominates

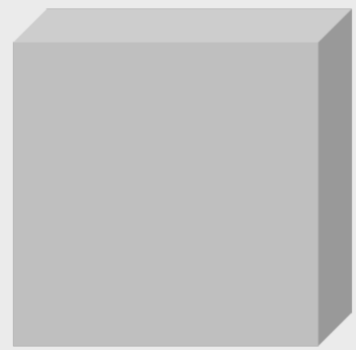


Wishbone/Dipolar to Boomerang/Exchange Transition

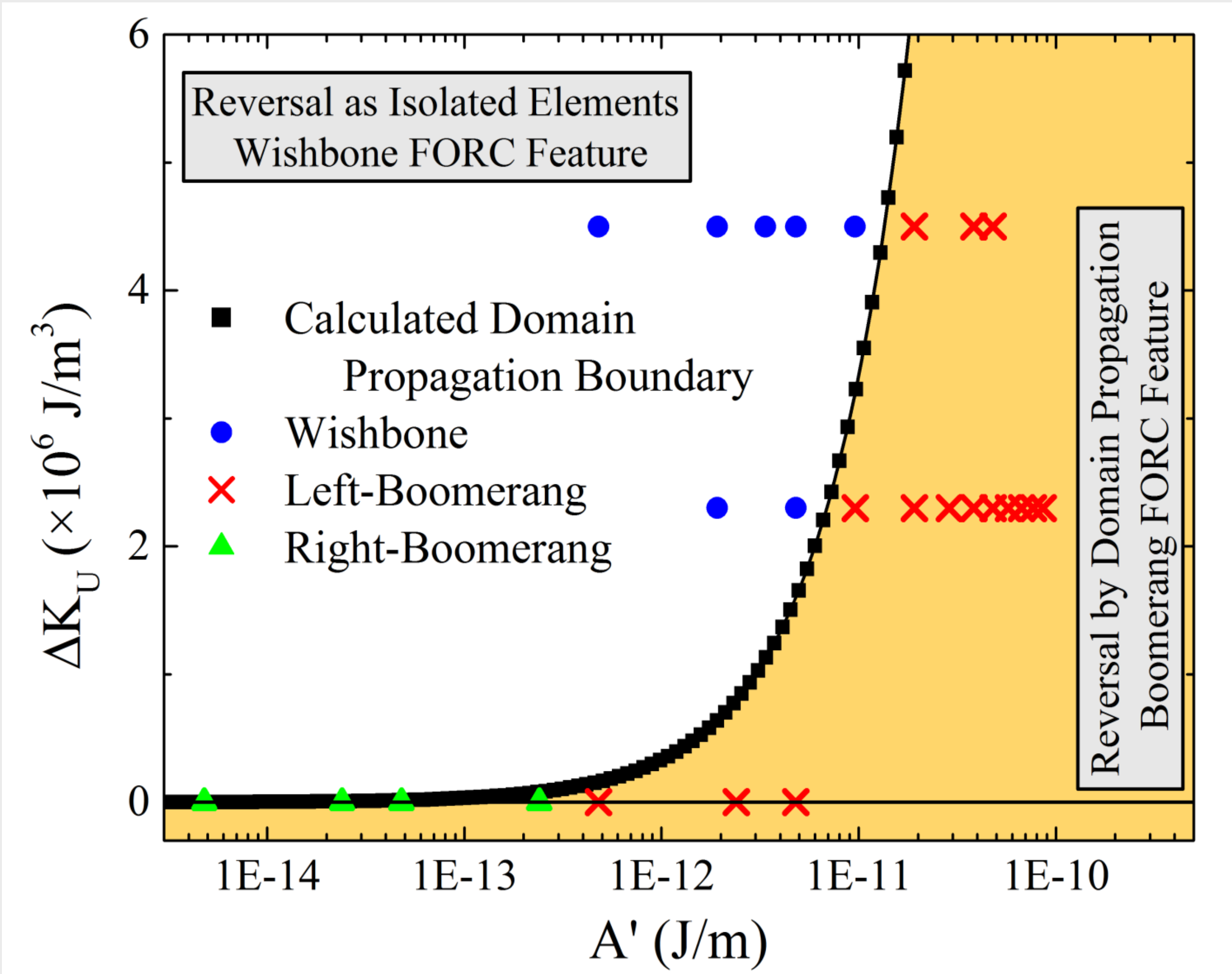
Demagnetizing planar



Magnetizing wall



Wishbone/Dipolar to Boomerang/Exchange Transition



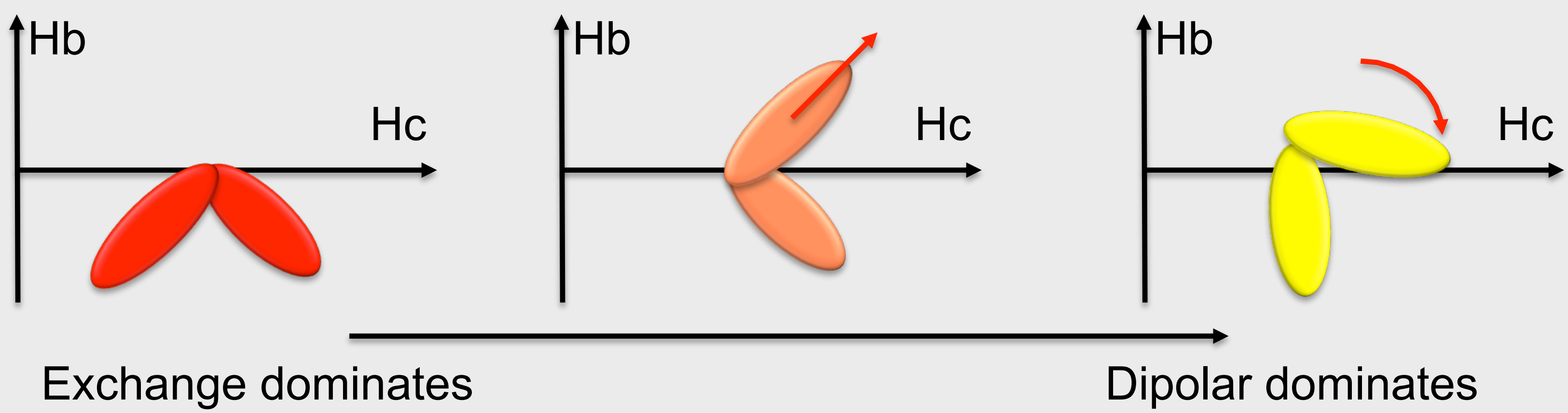
SUMMARY

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 $A>2x10^{-9}$ J/m
- Transition to right facing boomerang for smaller A



SUMMARY

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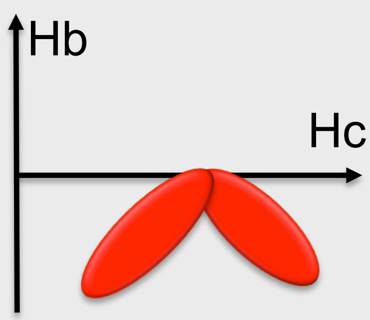
Exchange>Dipolar



Exchange<<Dipolar