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# Neutron scattering studies of exchange bias in $\text{Fe}_3\text{O}_4/\text{CoO}$ epitaxial thin films

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# Outline

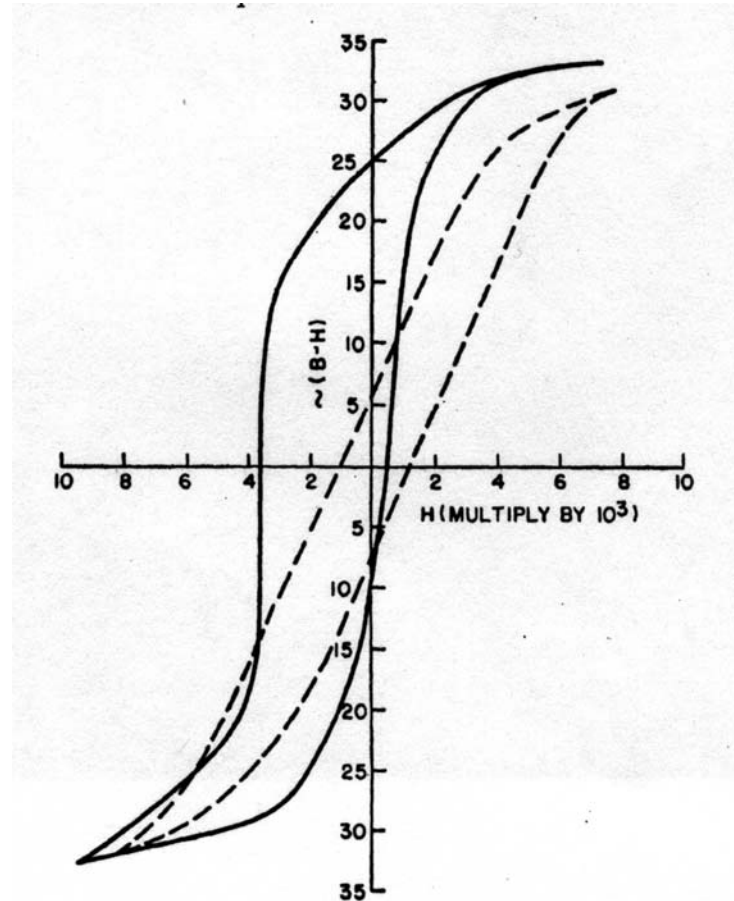
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- Background on exchange biasing
  - Definitions and models
  - Approach
- Experimental details
  - System choice
  - Measurement geometry for neutron scattering experiments
- Experimental results
  - Perpendicular coupling of AF and F spins
  - Inequivalence of  $T_N$  vs  $T_B$
  - Connecting perpendicular coupling to blocking temperature and biasing behavior
- Theoretical interpretation
  - Dzaloshinski/Moriya exchange

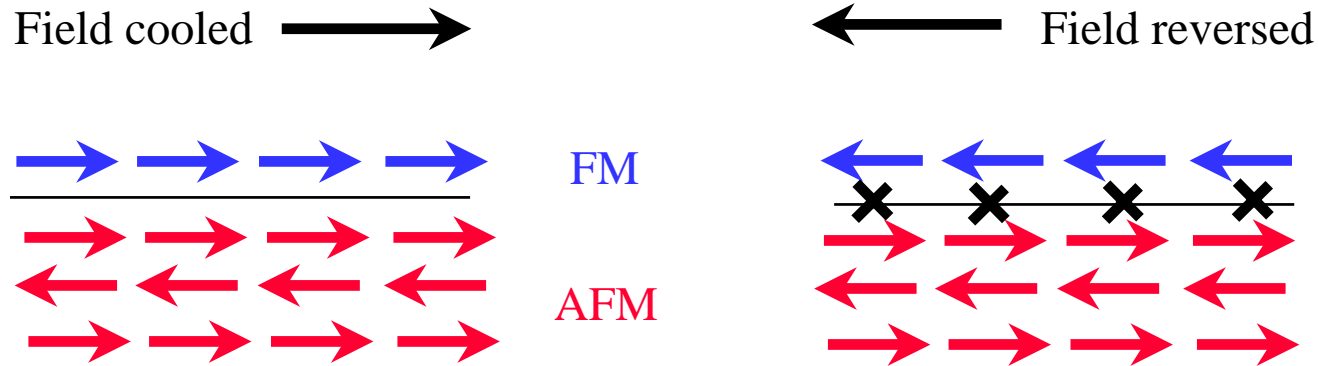
# Background-Exchange Bias or Anisotropy

- Main features
  - AFM in direct contact with FM
  - Cool system in magnetic field, through AFM Néel temperature
  - Observe unidirectional shift along field axis
- Issues
  - Increase in coercivity?
  - Asymmetries in loop shape?
  - Onset of shifted hysteresis loops vs  $T_n$ ?
- Uses in spin valves, read head sensors



Meiklejohn and Bean, *Phys. Rev.* **102**, 1413 (1956).

# Earliest model: Meiklejohn, Bean

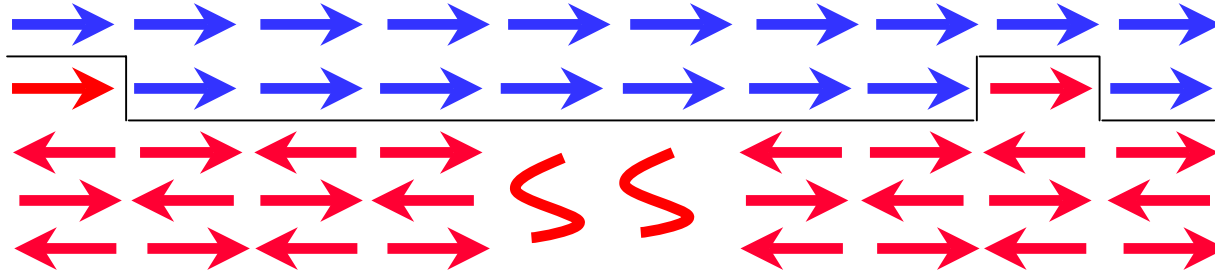


- Works well for a very well characterized system: Fe/Cr multilayers
  - Jiang, Felcher, Inomata, Goyette, Nelson, and Bader, *PRB* **61**, 9653 (2000)
- In general:
  - difficulties with size/temp. dependence/direction of shift
  - issues on thickness/growth/roughness/interface dependence
  - **questions about the nature of exchange**

# Models based on random field approach

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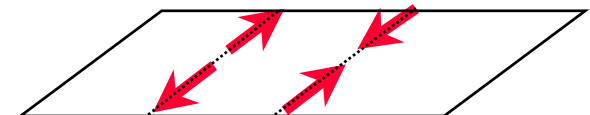
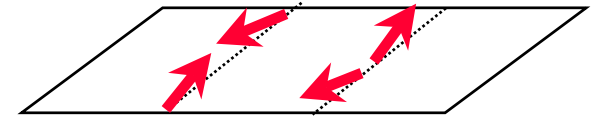
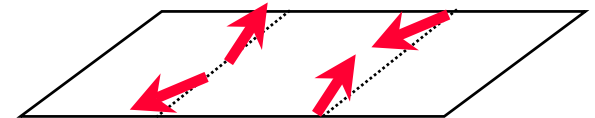
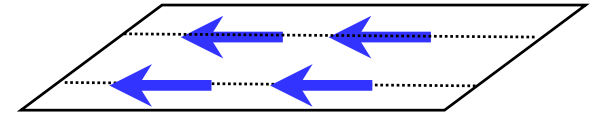


- Essential features
  - AFM at interface mostly compensated, broken into domains with a net uncompensated moment
  - Cooling field aligns all uncompensated moments
  - Imry and Ma, *PRL* **35**, 1399 (1975); Malozemoff, *JAP* **63**, 3874 (1988).
- Experimental verification: moments, domains
  - Kappenberger, Martin, Pellmont, Hug, Kortright, Hellwig, Fullerton, *PRL* **91**, 267202 (2003)
  - Miltenyi, Gierlings, Keller, Beschoten, Gunterrodt, Nowak, and Usadel, *PRL* **84**, 4224 (2000)

# Models based on spin flop coupling

- Essential features

- FM spins align perpendicular to AFM easy axis, analogous to AFM in high magnetic field
- Domain wall parallel to the interface
- Hinchey and Mills, *PRB* **34**, 1689 (1986) and Koon, *PRL* **78**, 4865 (1997).



- Problems

- Works for x-y spins, not Heisenberg, leading to coercivity not bias
- Compatibility with random field model?
- Schulthess and Butler, *PRL* **81**, 4516 (1998); Stiles and McMichael, *PRB* **59**, 3722 (1999)

# Experimental approach

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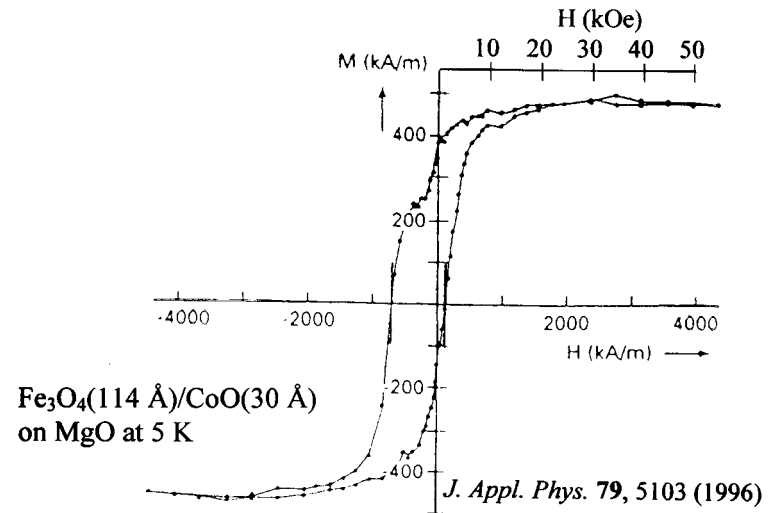
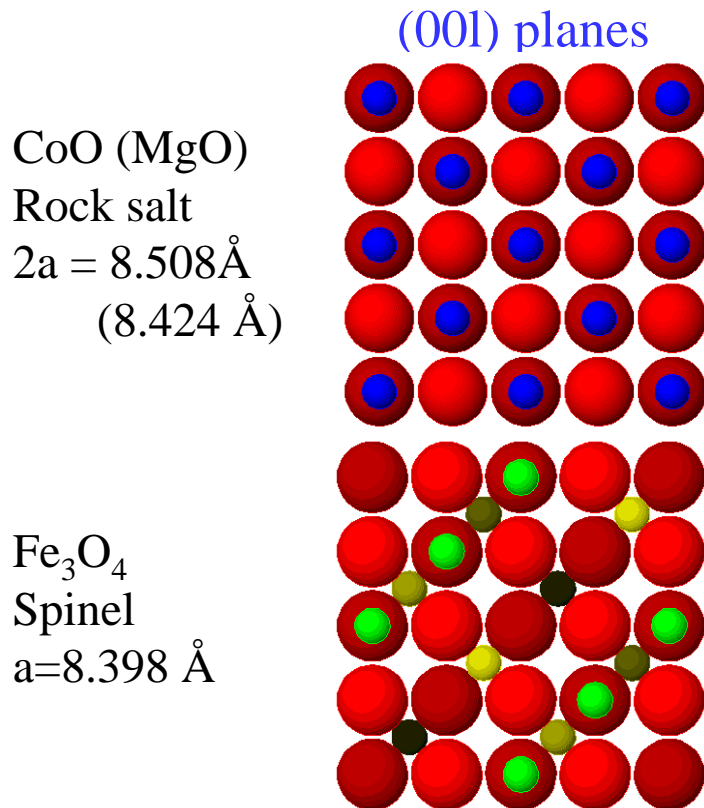
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- Many exchange biasing issues centered on either the antiferromagnet or the interface spins
  - orientation of spins
  - temperature evolution
  - nature of domains
- Techniques to probe the antiferromagnet, (buried) interface
  - use large single crystals of AFM/companion samples
  - image with spin-polarized STM
  - x-ray magnetic circular dichroism
  - Neutron scattering
- Approach: to use neutron diffraction and reflectivity techniques along with other magnetization probes to correlate behavior

# Fe<sub>3</sub>O<sub>4</sub>/CoO system

- Good growth due to structural match

- (Fe<sub>3</sub>O<sub>4</sub> 100 Å)/(CoO 17-100 Å)<sub>x50</sub>  
 -(CoO 30 Å/MgO 30 Å)<sub>x333</sub> on MgO



- Magnetic properties

- Bulk **CoO** orders AFM at 291K, planes alternate in <111> directions, 3.9 μ<sub>B</sub> on Co<sup>+2</sup>  
 - Bulk **Fe<sub>3</sub>O<sub>4</sub>** orders ferrimagnetic at 858 K, net moment 4.2 μ<sub>B</sub>

- Composite system shows bias



# Scattering geometry

- Measure AFM reflection or FM reflection or low angle reflectivity

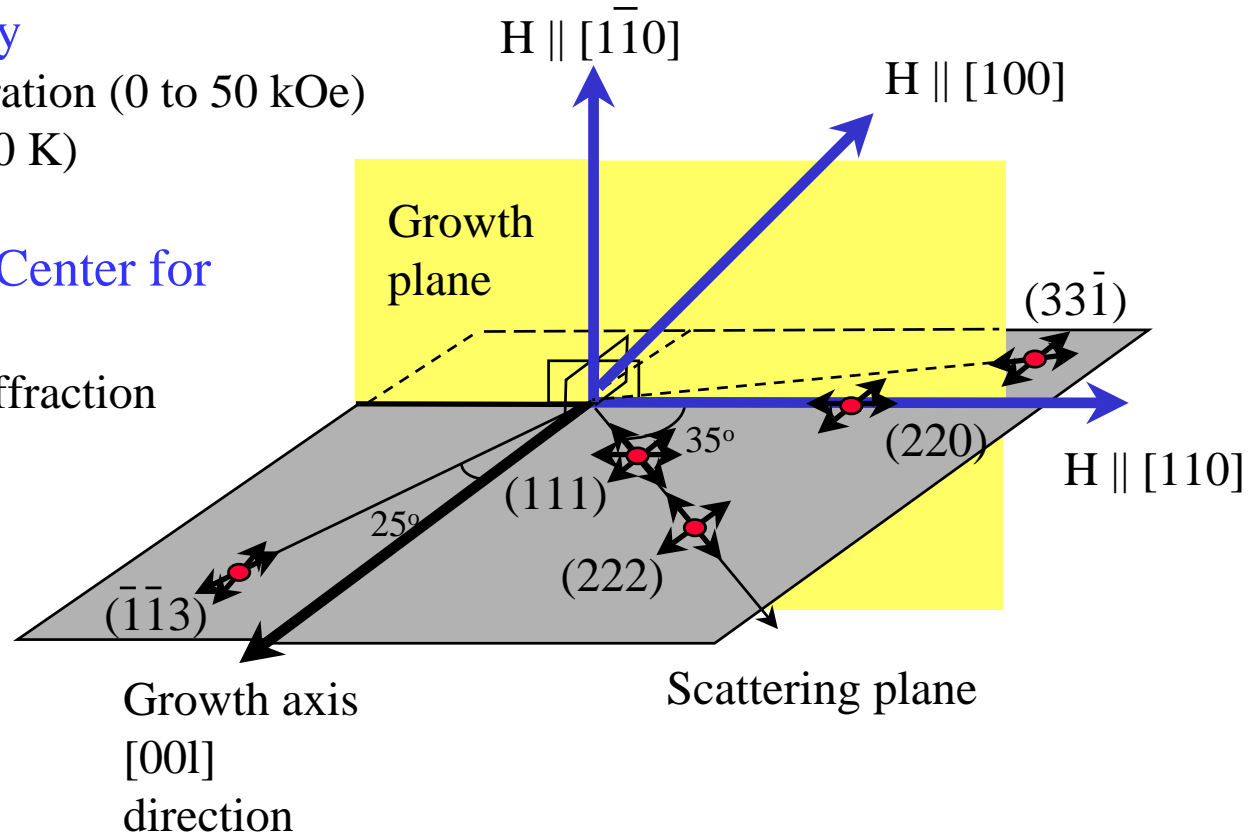
-vary field cooling preparation (0 to 50 kOe)

-vary temperature (10-550 K)

- Experiments at NIST Center for Neutron Research

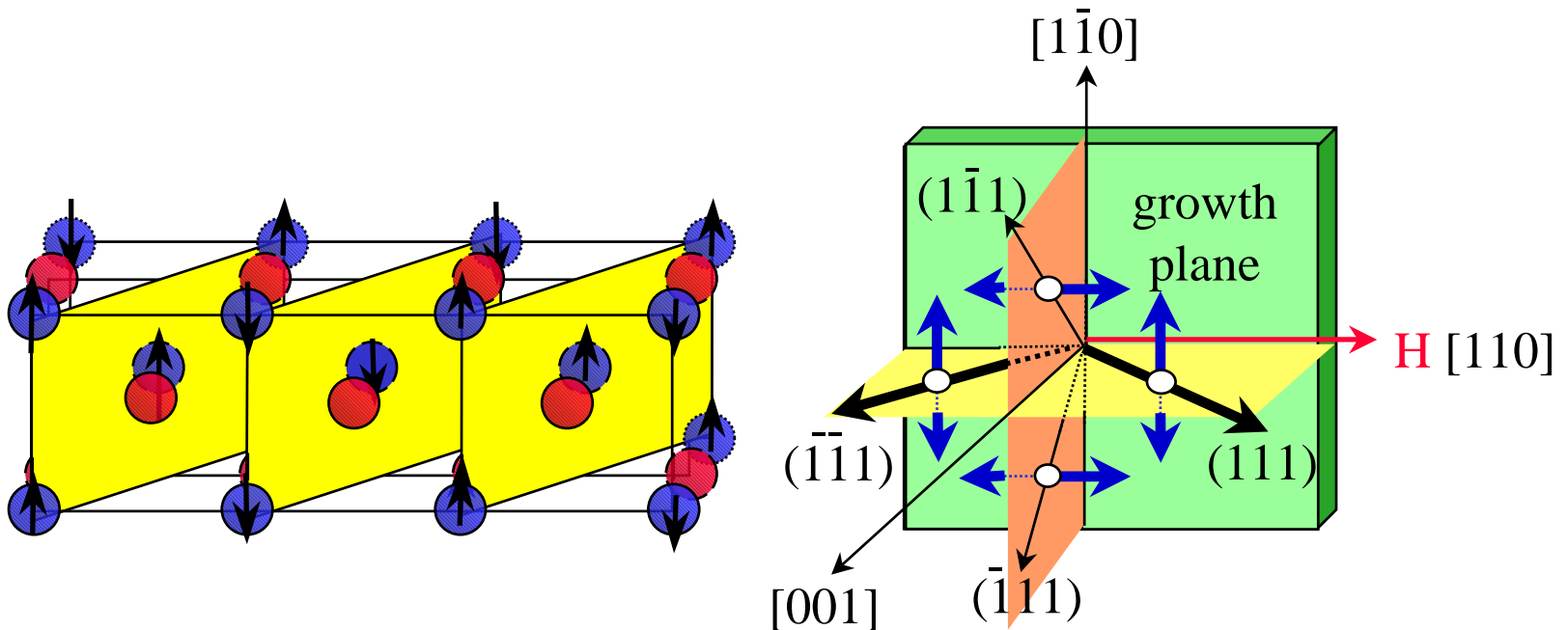
-BT2, BT9, SPINS for diffraction

-NG1 for reflectivity



# AFM spin directions for CoO

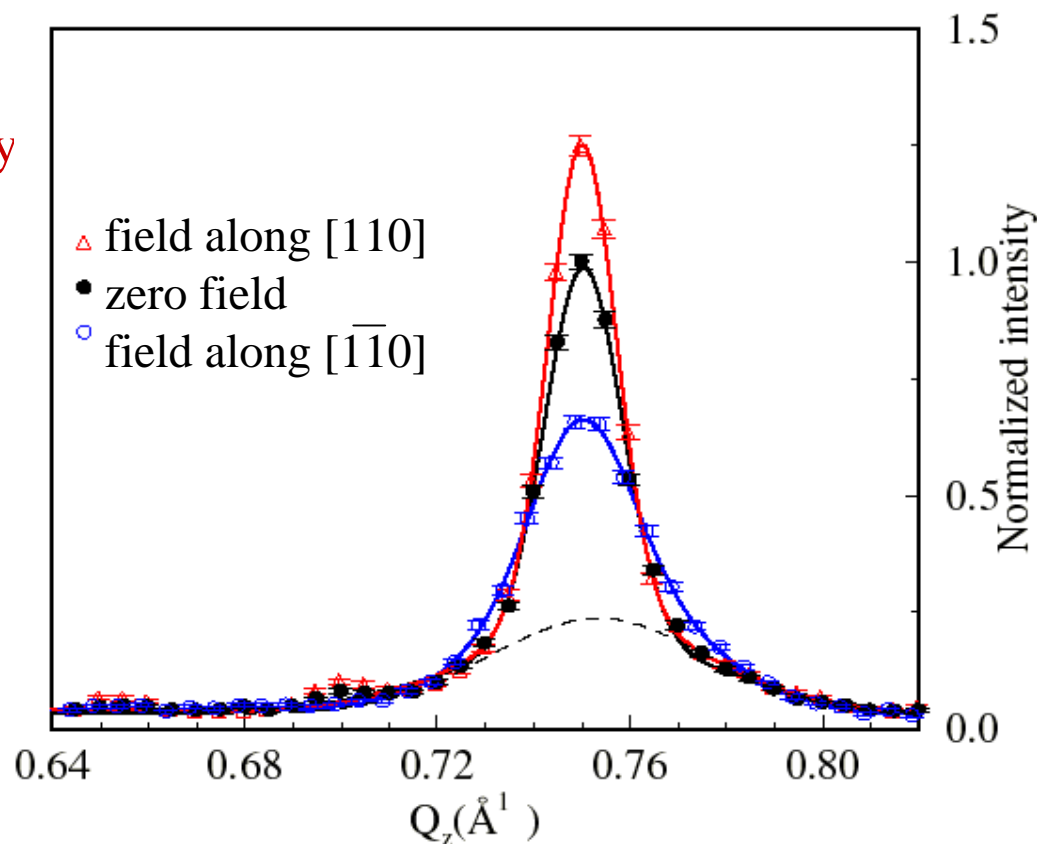
- New moment directions, substantially different from bulk
- Spins constrained within sample growth plane
- Observed for both  $\text{Fe}_3\text{O}_4/\text{CoO}$  and  $\text{CoO}/\text{MgO}$  superlattices



Ijiri, Borchers, Erwin, Lee, van der Zaag, Wolf, *PRL* **80** (608), 1998

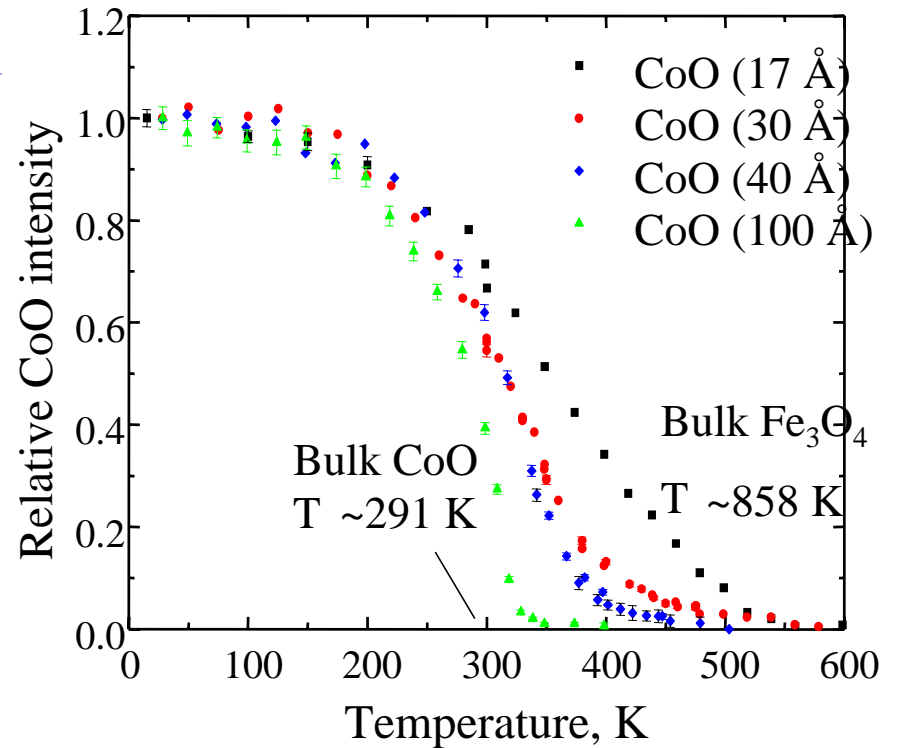
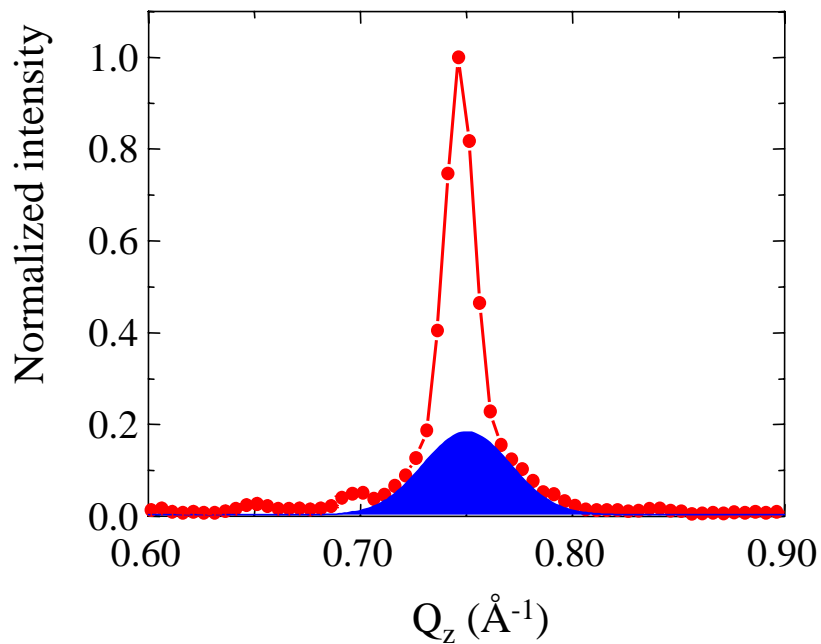
# AFM/FM perpendicular alignment

- AFM spins are preferentially perpendicular to FM spins
- Effect not observed for CoO/MgO superlattice



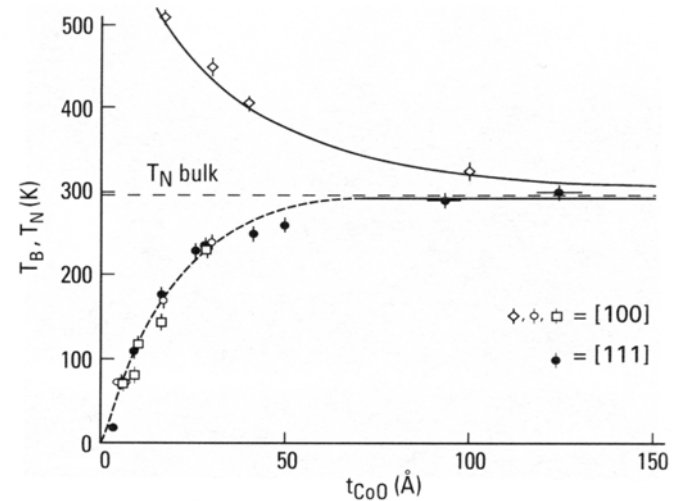
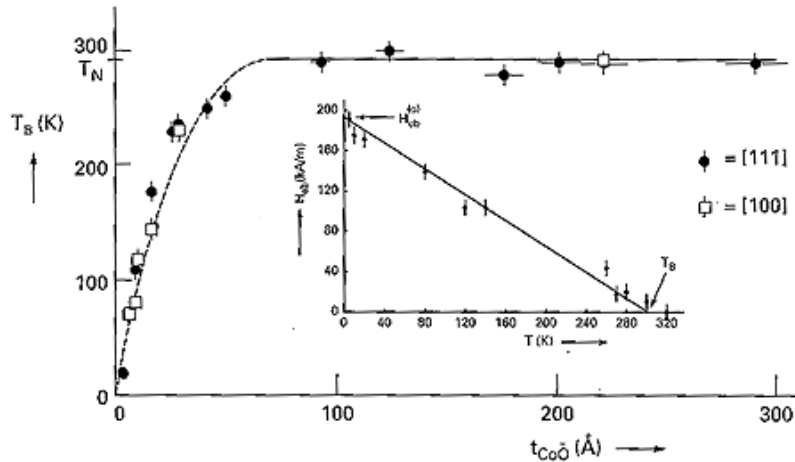
# Data to extract $T_N$

- 2 component line shape to reflection
  - Broad- $\text{Fe}_3\text{O}_4$  contribution
  - Narrow-CoO contribution



- $T_N$  increases with decreasing CoO thickness

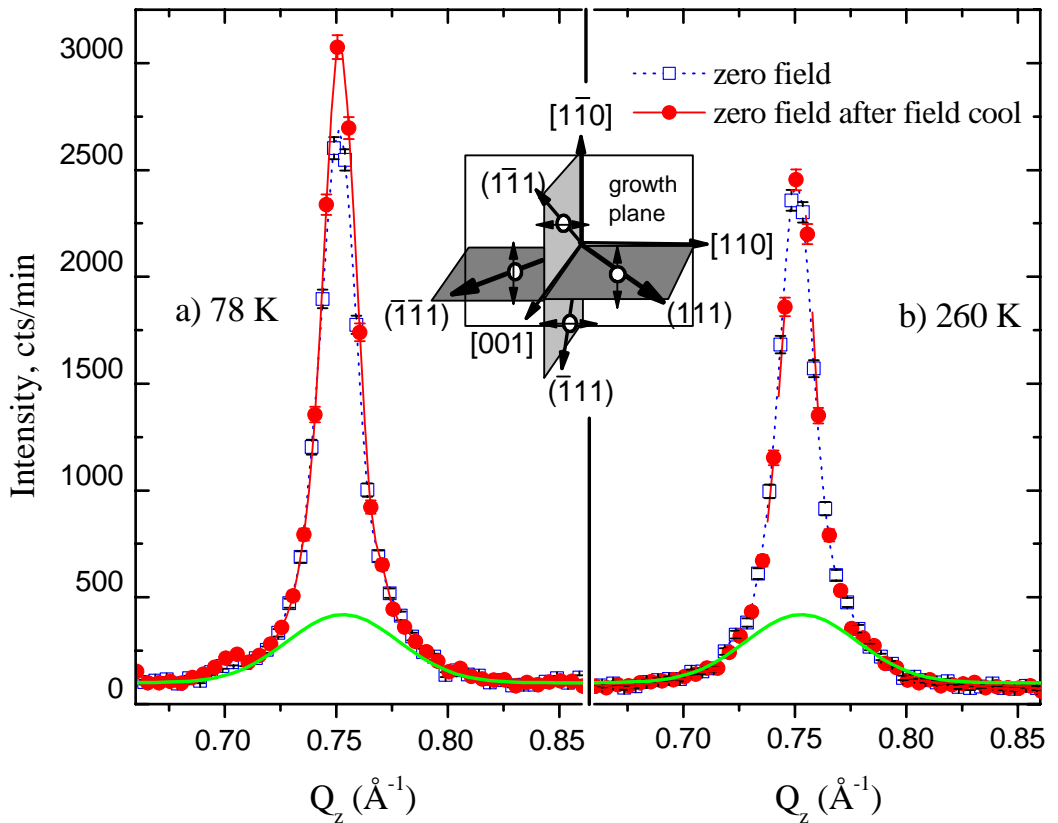
# Comparison of $T_N$ to $T_B$



- $T_B$  from SQUID magnetometry shows **opposite** trend to  $T_N$   
-van der Zaag, Ijiri, Borchers, Feiner, Wolf, Gaines, Erwin, Verheijen, *PRL* **84** (6102), 2000
- Reduced  $T_B$  not a finite size effect of  $T_N$

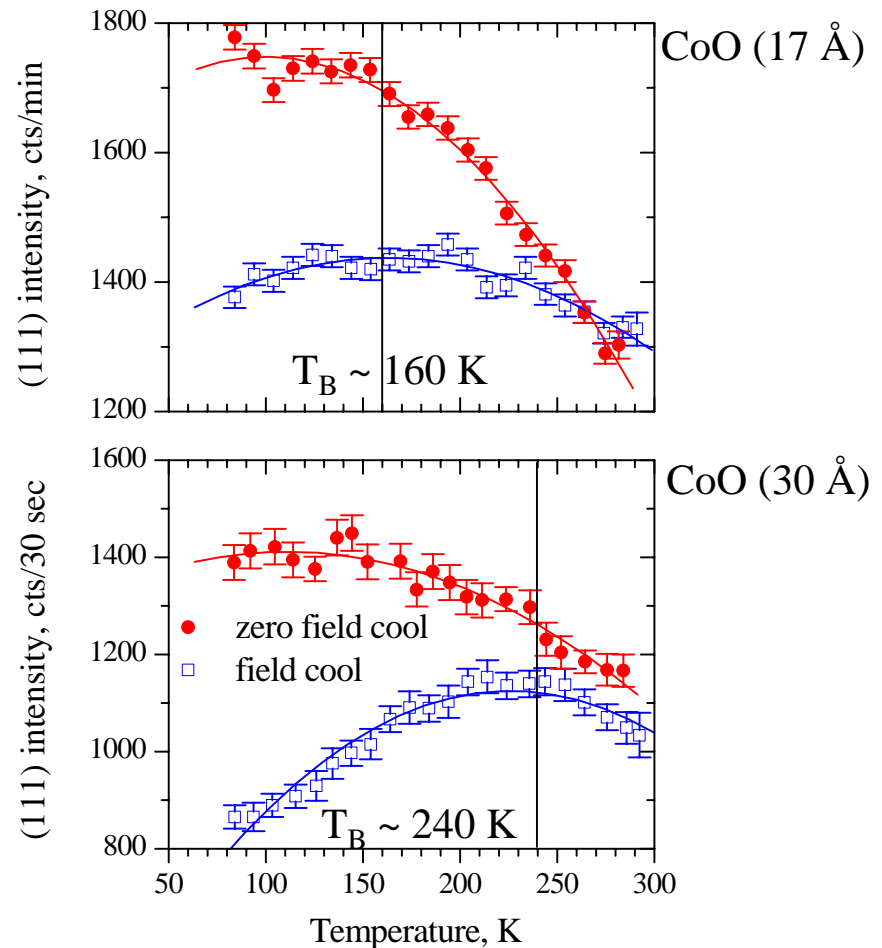
# AFM behavior associated with $T_B$

- Below  $T_B$ , preferred AFM directions appear locked in
- Above  $T_B$ , AFM directions randomized



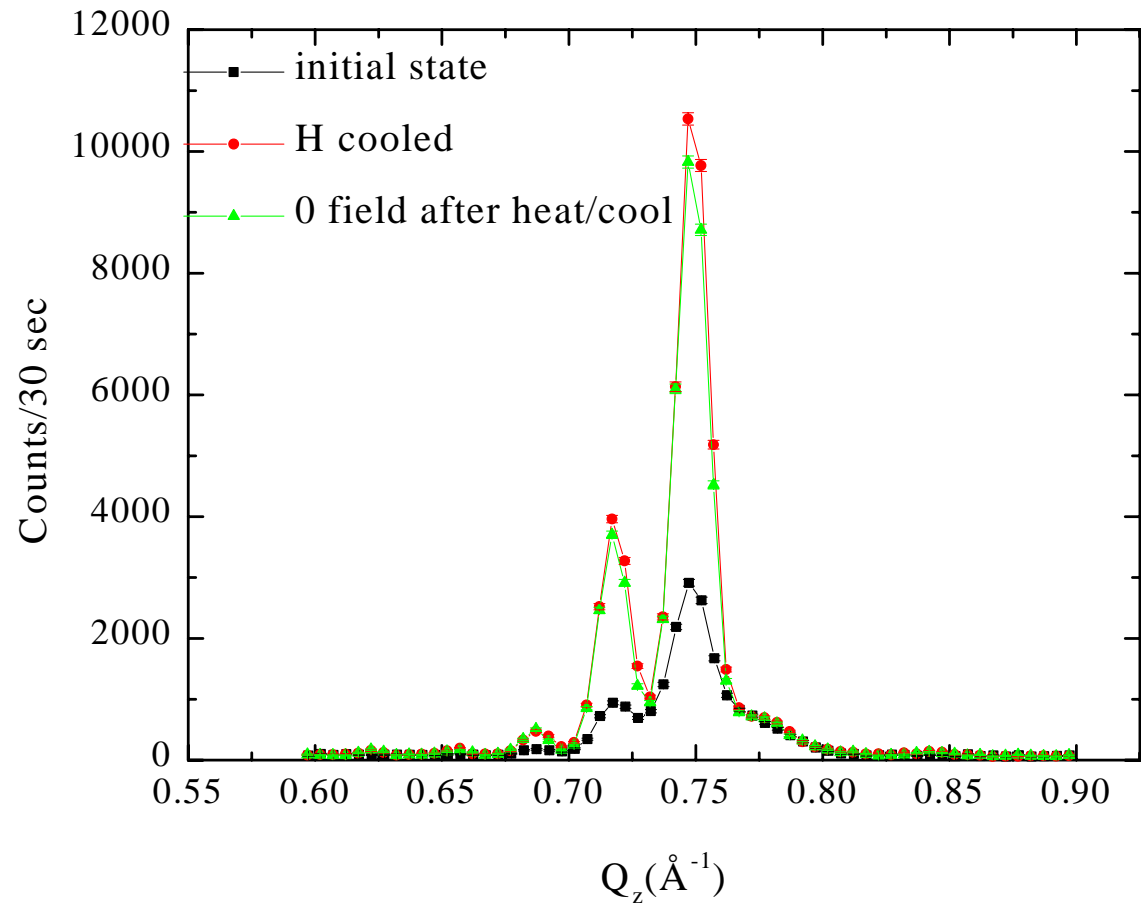
# Perpendicular coupling and $T_B$

- Track (111) intensity vs. temp. with and without field treatment
- Observe peak~ plateau corresponding to  $T_B$
- Unlocking of spins from preferential perpendicular coupling direction at  $T_B$



# Behavior for $T_B \sim T_N$

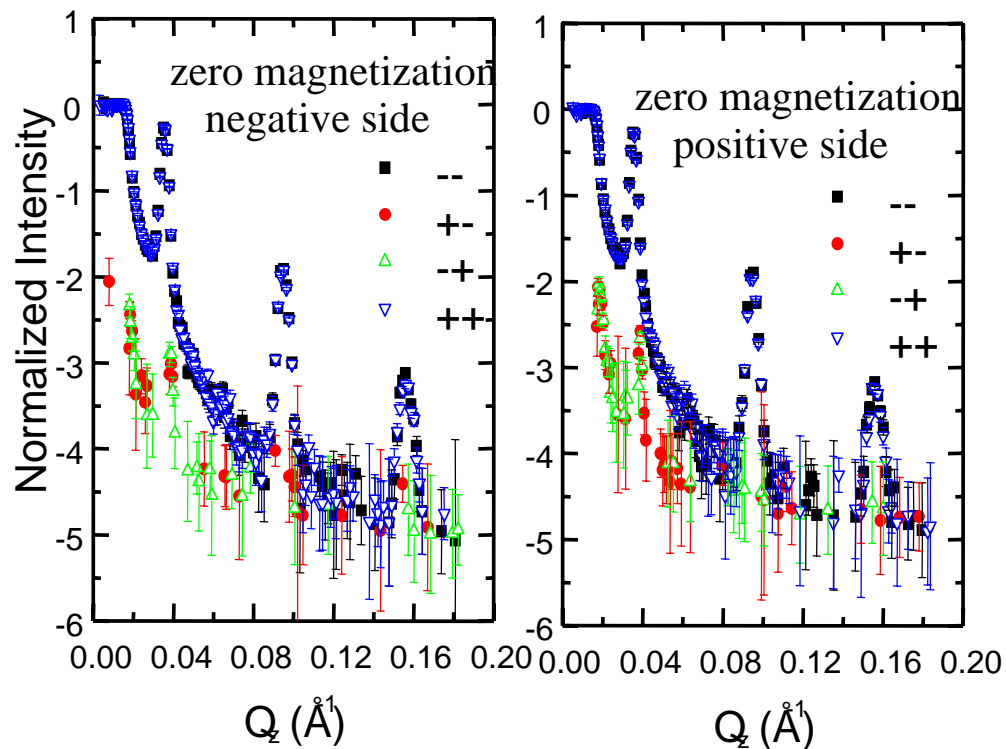
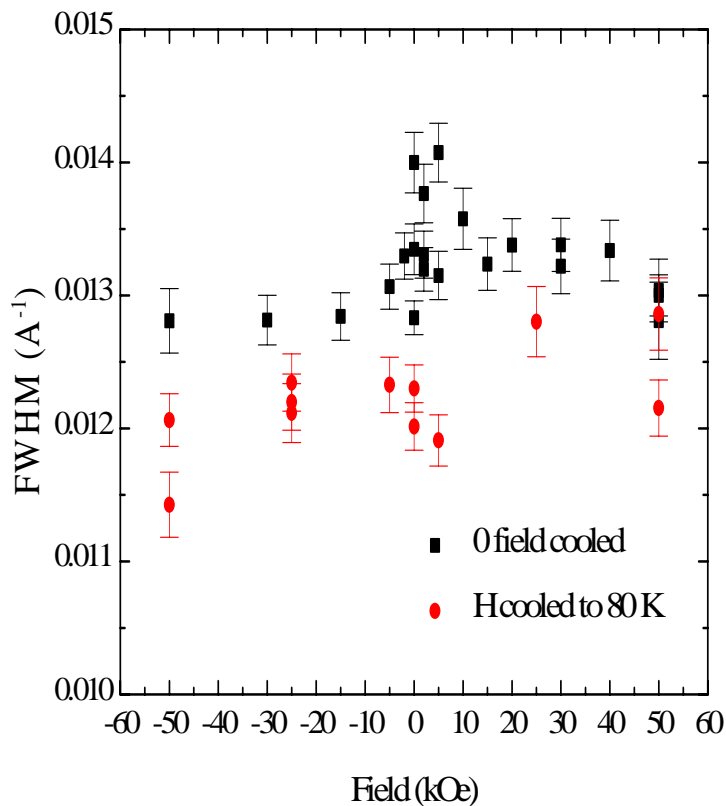
- Field preparation memory despite re-randomizing the CoO above  $T_N$
- Evidence of response to  $\text{Fe}_3\text{O}_4$





# Character of AFM, FM domains

- Few changes on field cycling



- Similar to random field model, little evidence of twists, changes in average domain sizes

# Theoretical understanding

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- Experimental results:

- Perpendicular coupling clearly associated with biasing-connection to  $T_B$
- Otherwise random field like



How to connect the two?

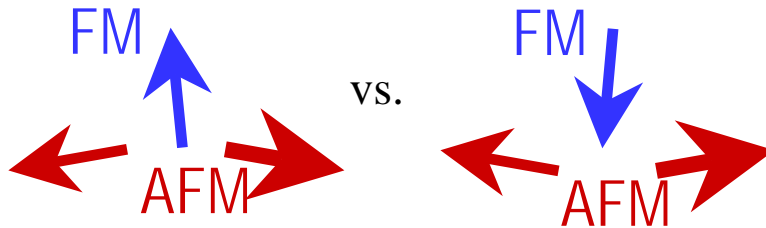
- Role of anisotropic exchange term?

- $E_{A-F} = -J_{A-F} \mathbf{S}_A \cdot \mathbf{S}_F + \mathbf{D}_{A-F} \cdot (\mathbf{S}_A \times \mathbf{S}_F)$
- Dzialoshinski, Sov. Phys. *JETP* **5**, 1259 (1957); Moriya, *Phys. Rev.* **120**, 91 (1960)
- D term from spin orbit coupling and superexchange interaction
- Nonzero for noncollinear spins
- Nonzero only for low/broken symmetry

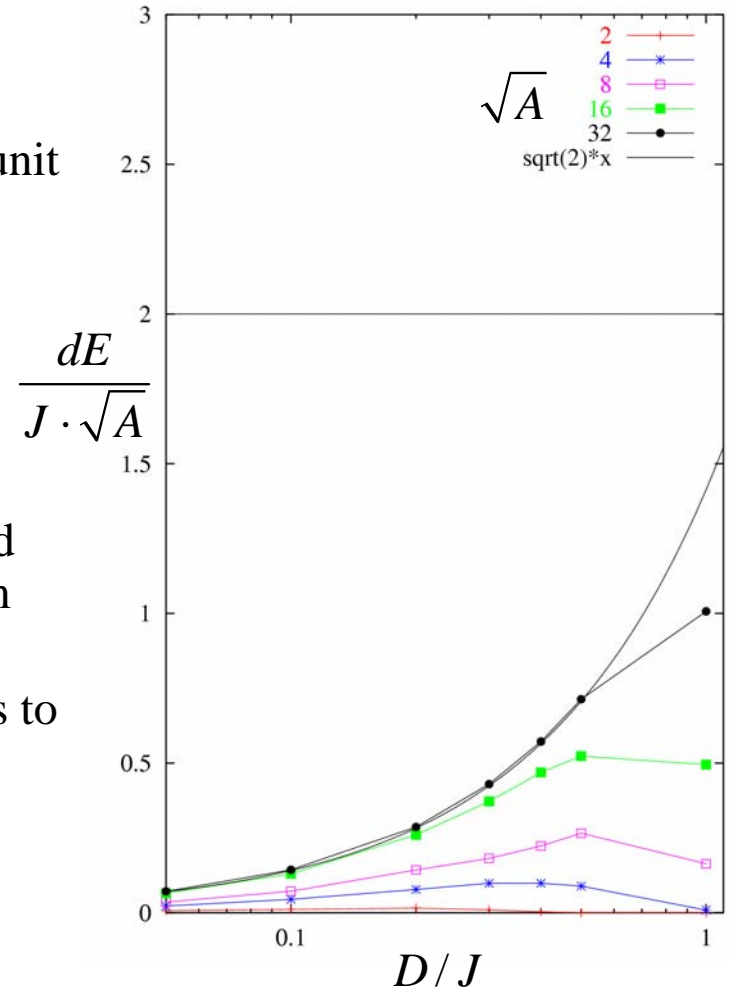
# D-M exchange for biasing

- Recent simulations

- Calculate energy difference for 2 configurations  $\sim 2(D^z_{\text{net}})\sin\theta$  for one unit

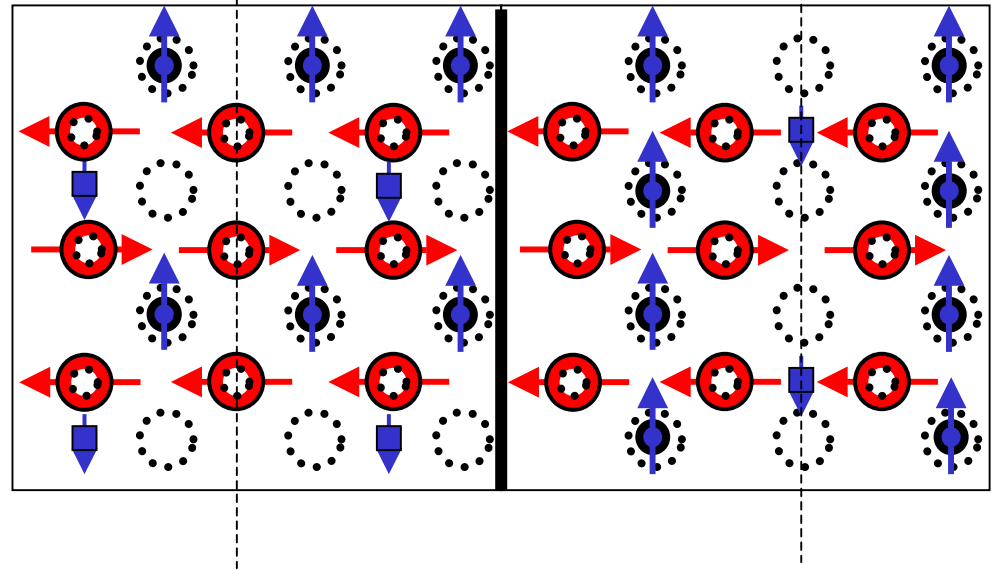


- $D/J \sim .3$ , Heisenberg spins, randomized bias comparable to Ising spins, random field model
- Size effect to coupling directions-leads to coercivity
- Schulthess, MRS Symp. Proc. **346**, 31 (2003).

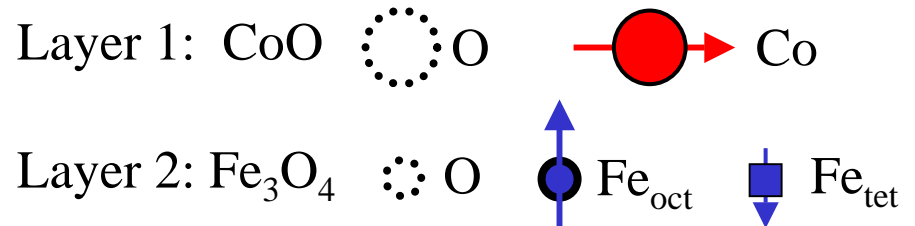


# DM exchange for Fe<sub>3</sub>O<sub>4</sub>/CoO

- Consider interface
  - CoO domains longer range
  - Fe<sub>3</sub>O<sub>4</sub> domains shorter due to antiphase boundaries
  - Hibma, *et al.*, *JAP* **85**, 5291 (1999)



- Tetrahedral irons can have significant DM exchange



# Conclusions and Further Work

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- Summary of main results for  $\text{Fe}_3\text{O}_4/\text{CoO}$  system
  - Preference for perpendicular coupling of FM and AFM spins
  - Inequivalence of  $T_N$  vs  $T_B$
  - Association of  $T_B$  with the unfreezing of perpendicular coupling
  - Results consistent with a model of anisotropic exchange
- Implications of work
  - Interfacial spins can be very different from the bulk
  - Need for more sophisticated exchange considerations
- Further work to explore model of anisotropic exchange
  - Quantitative match?
  - Density of antiphase domains, etc. vs. size of bias?