



# A New Paradigm for Exchange Bias in Polycrystalline Films

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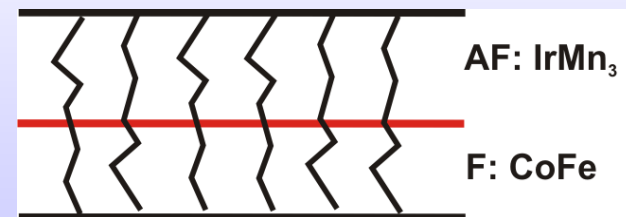
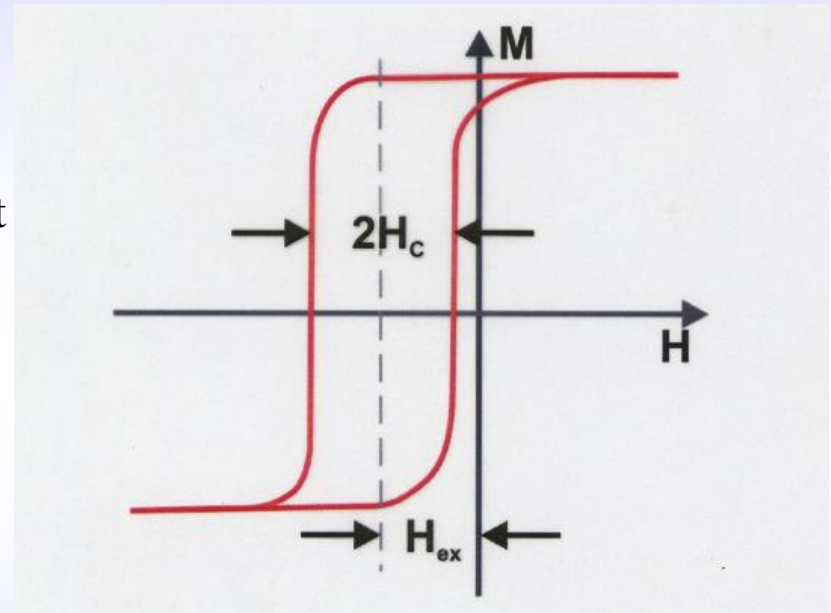
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# Exchange Bias

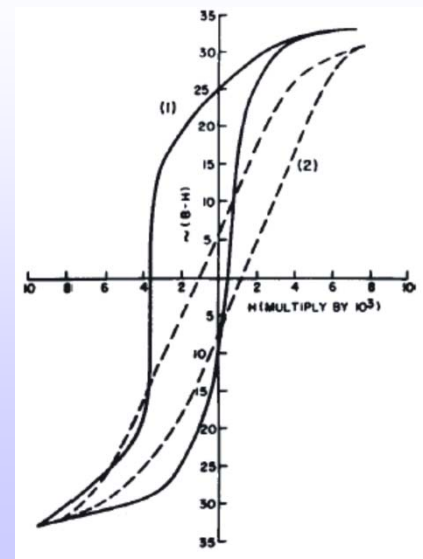
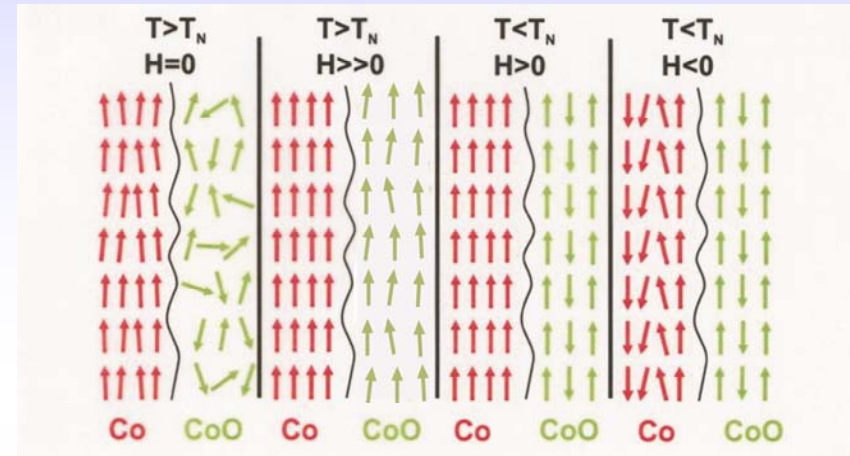
- Exchange bias was first reported in 1956 by Meiklejohn and Bean.
- The effect occurs when an antiferromagnet (*AF*) is in contact with a ferromagnet (*F*).
- This can be engineered in thin films.
- It can also occur if films or particles of Co or Ni are oxidised to give a surface of NiO or CoO.
- If the *AF* is field-cooled through  $T_N$  a shifted loop and increased  $H_c$  occur.



W. H. Meiklejohn and C. P. Bean, *Phys. Rev.* **102** p.1413 (1956).

# FM/AFM Coupling

- CoO is a classical superexchange-based *AF*.
- Neighbouring (111) planes align antiparallel with  $T_N \sim 290$  K
- Hence in oxidised small Co particles there is an interface between a *F* and an *AF* material.
- The loop shifts by  $H_{ex}$ .
- $H_c$  increases dramatically.



# Early Theories

- Meiklejohn and Bean calculated the shift assuming a perfect interface but were  $> 10x$  out.
- An uncompensated spin interface model by Néel also failed.
- There have been several complex models predicting domain walls in the AF with marginal success.
- The only successful model was a granular model due to Fulcomer and Charap based on Stoner-Wohlfarth theory.

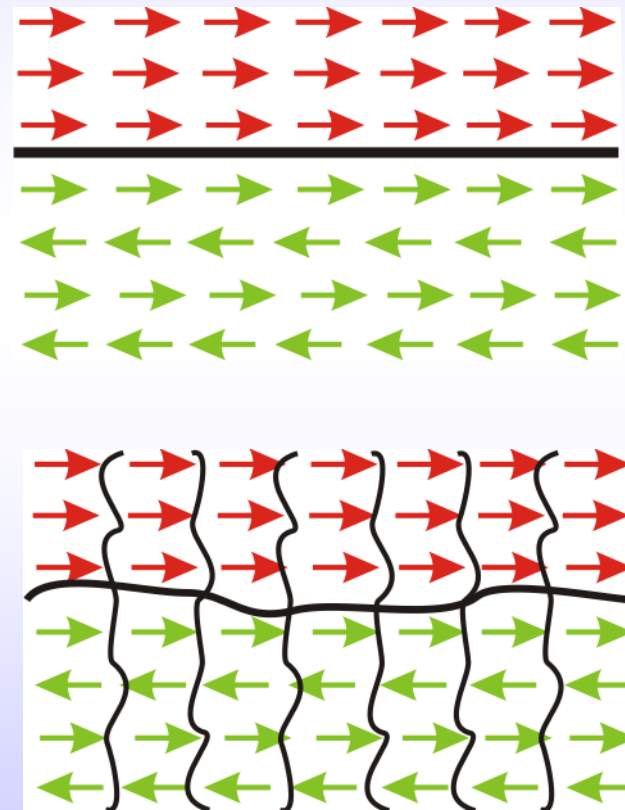
W. H. Meiklejohn and C. P. Bean, *Phys. Rev.* **102** p.1413 (1956).

L. Néel, *Ann. Phys. Paris* **2** p.61 (1967).

R. L. Stamps, *J. Phys. D: Appl. Phys.* **33** p.R247 (2000).

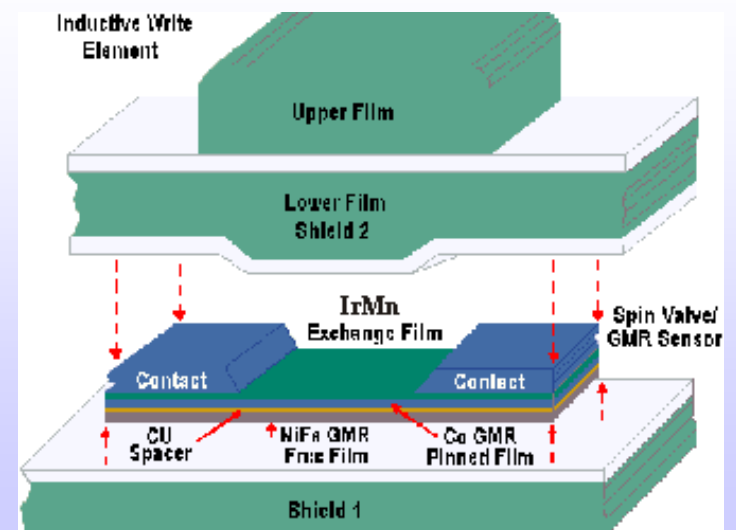
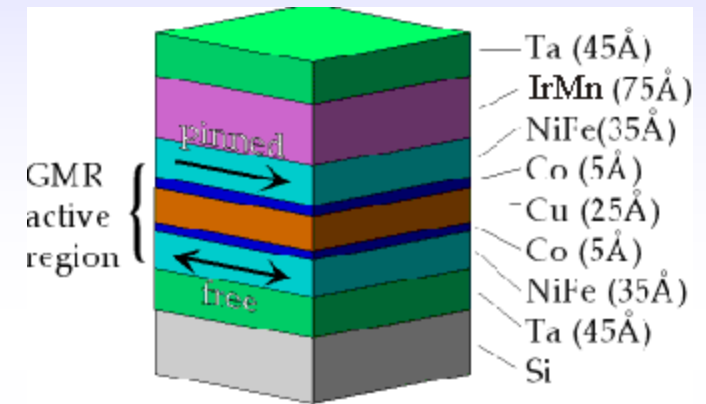
U. Nowak, K. D. Usadel, J. Keller, P. Miltényi, B. Beschoten and G. Güntherodt, *Phys. Rev. B* **66** 014430 (2002).

E. Fulcomer and S. H. Charap, *J. Appl. Phys.* **43** p.4190 (1972).



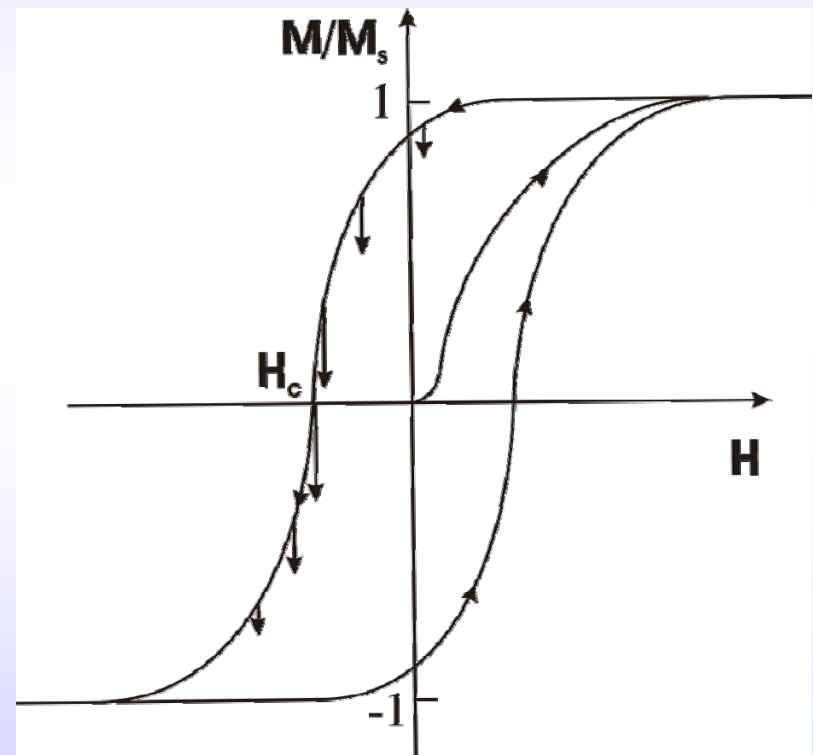
# Technical Importance

- From 1956 to ~1990, Exchange Bias (EB) was of academic interest only.
- With the discovery of GMR and the development of spin-valves EB was used to align the pinned layer.
- An understanding of how EB works and can be controlled was then essential.
- For example, early spin-valve heads used NiO as the *AF* and had to be reset.
- All GMR, TMR and Spin Electronic devices require or will require Exchange Bias.



# Time Dependent Effects

- All magnetic materials exhibit time dependent effects because hysteresis is a non-equilibrium phenomenon.
- In a ferromagnet, time dependence occurs around the coercivity leading to a sweep-rate dependence of  $H_c$ .
- Time dependence occurs because of thermal activation over energy barriers.
- The energy barriers are due to anisotropy effects or domain wall pinning.



# Nature of Relaxation

- Relaxation over a single barrier is described by the Néel Arrhenius law

$$\tau^{-1} = f_0 \exp\left(\frac{-\Delta E}{k_B T}\right) \quad f_0 = \text{attempt frequency}$$

- Note that the origin of the barrier is not specified.
- In real materials there is a distribution of barriers so that approximate  $\ln(t)$  behaviour is observed.

$$M(t) = M_0 \pm S(H) \ln t$$

- $S(H)$  depends on the value of the energy barrier distribution  $f(\Delta E)$  at the critical value being activated.

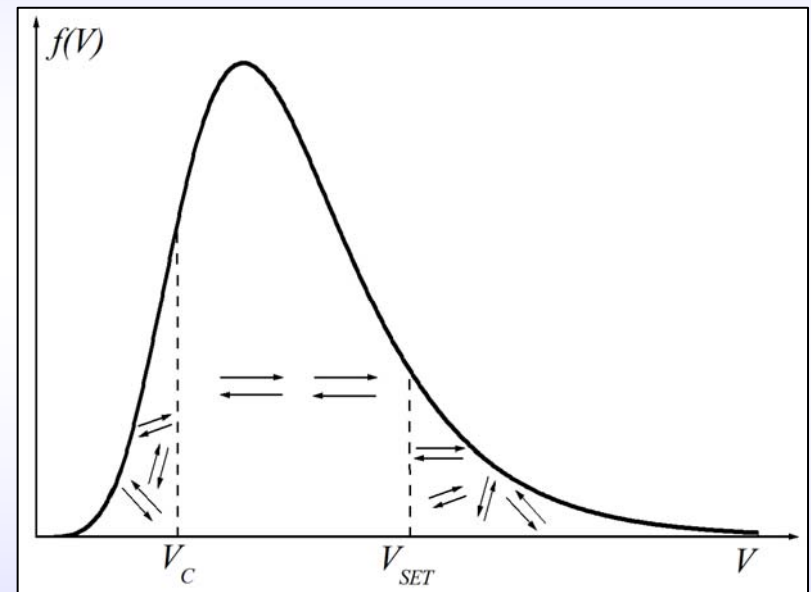
$$S(H) = 2M_s k_B T f(\Delta E_c(H, T))$$

R. Street, and J. C. Wooley, *Proc. Phys. Soc.*, **A62** p.562 (1949)

P. Gaunt, *J. Appl. Phys.*, **59** p.4129 (1986)

# Time Dependence in *AF*s

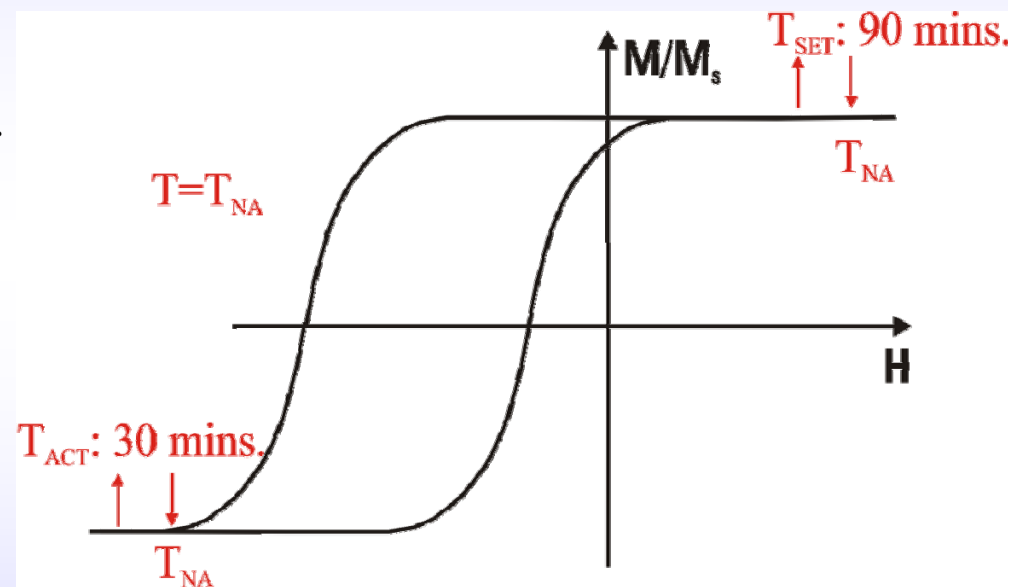
- In polycrystalline films the *AF* is ‘set’ below  $T_N$  to avoid damage to the structure.
- The *AF* is ordered by the exchange field from the F layer.
- This is done by field annealing using thermal activation giving an  $\ln(t)$  process.
- Temperature causes parts of the *AF* to disorder and only the stable grains cause  $H_{ex}$ .
- Also in IrMn ( $T_N=690K$ ) the ‘setting’ of the *AF* is often incomplete.



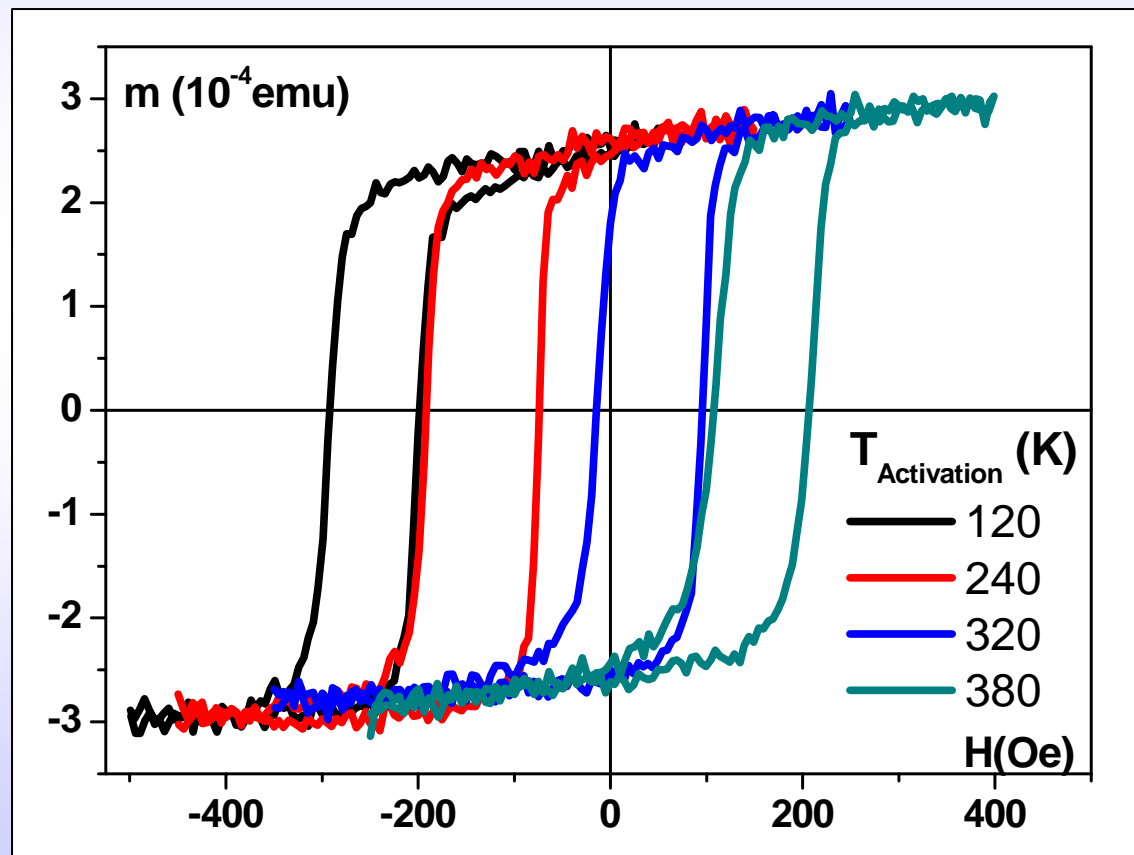


# York Protocols

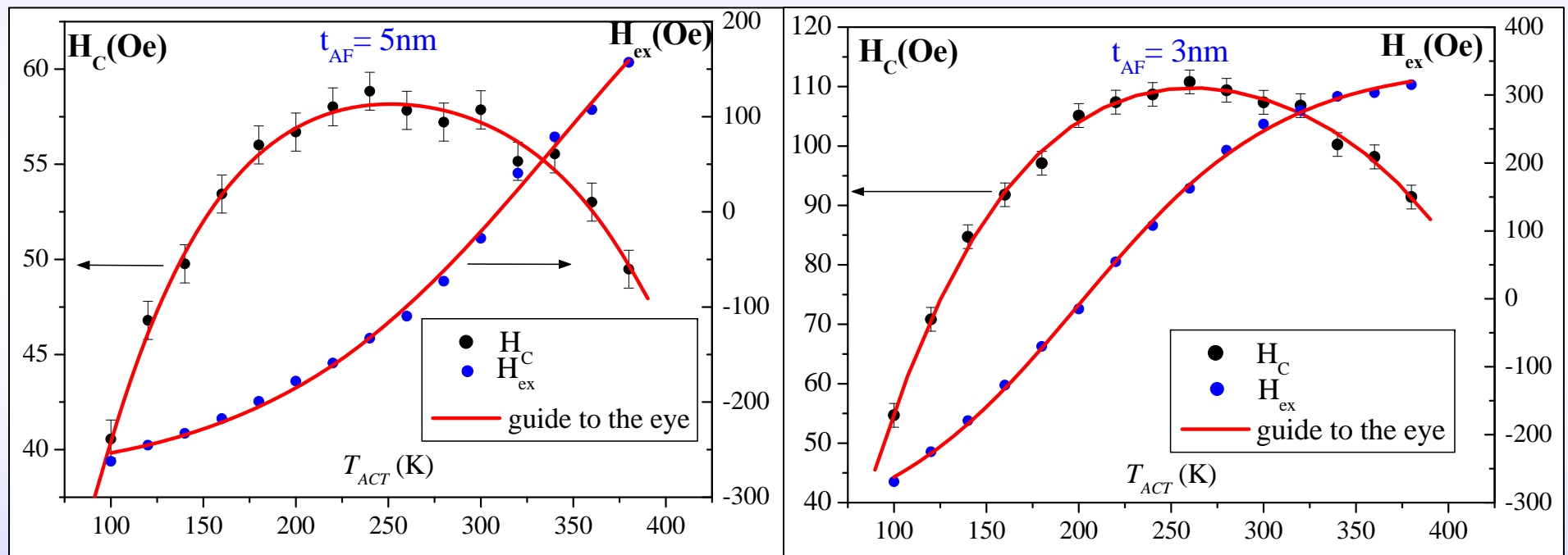
- The  $AF$  is set at  $T_{SET}$  for 90 minutes.
- Sample cooled to  $T_{NA}$ .
- Sample heated to  $T_{ACT}$  for 30 mins.
- Sample measured at  $T_{NA}$ .



# Reversal in IrMn(5nm)/CoFe(10nm)



# $H_C$ and $H_{ex}$ IrMn( $t_{AF}$ )/CoFe(10nm)

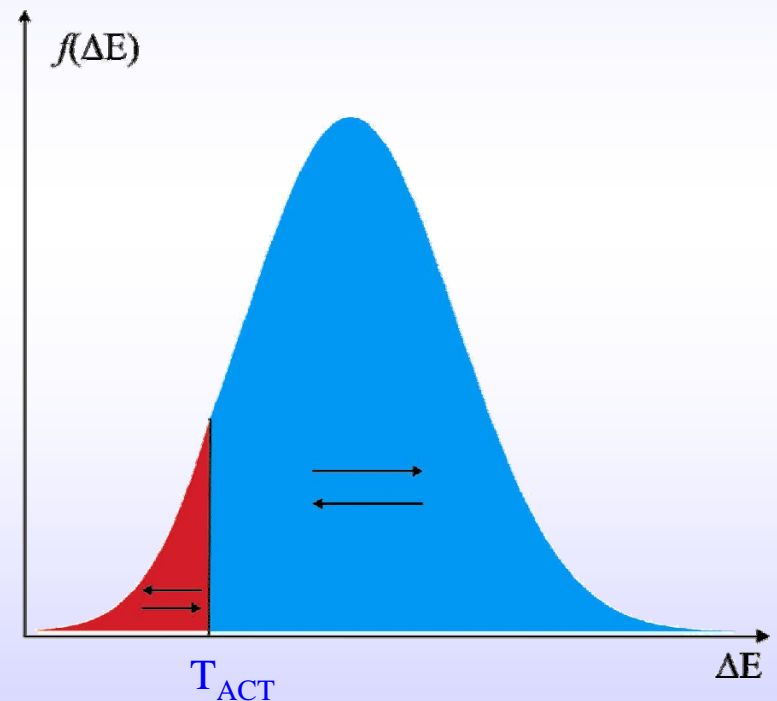


# Features of Exchange Bias

- $H_{ex}$  is controlled by the ordered  $AF$ .
- $H_{ex}$  and  $H_c$  are not related.
- Ordering is controlled by thermal activation.

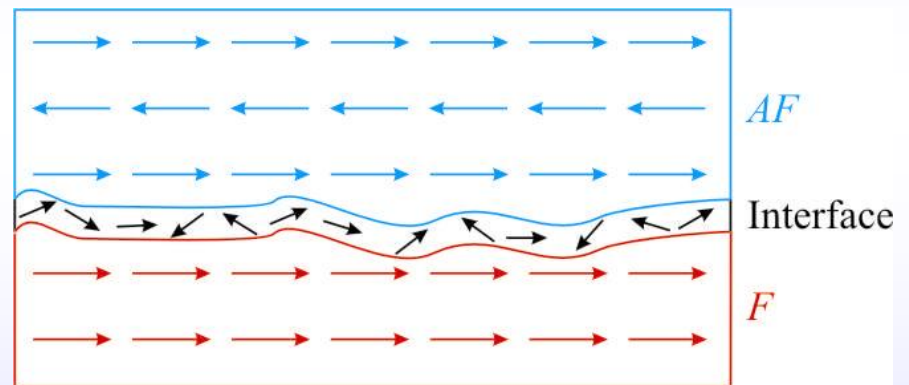
$$H_{ex} \propto \int_{\Delta E_c(T_{ACT})}^{\infty} f(\Delta E) d\Delta E - \int_{\Delta E_c(T_{NA})}^{\Delta E_c(T_{ACT})} f(\Delta E) d\Delta E$$

- $f(\Delta E)$  appears to scale with the grain volume.



# Nature of the Energy Barrier

- For several years there has been no clear model of  $\Delta E$  in *AF*s.
- Some, but not all, features can be explained using *AF* domain structures and computer models.
- All models predict  $H_{ex}$  values too large by orders of magnitude.
- We proposed a simple model based on granular reversal of  $H_{ex}$  modulated by disordered interfacial spins.



$$\Delta E = K_{AF} V$$

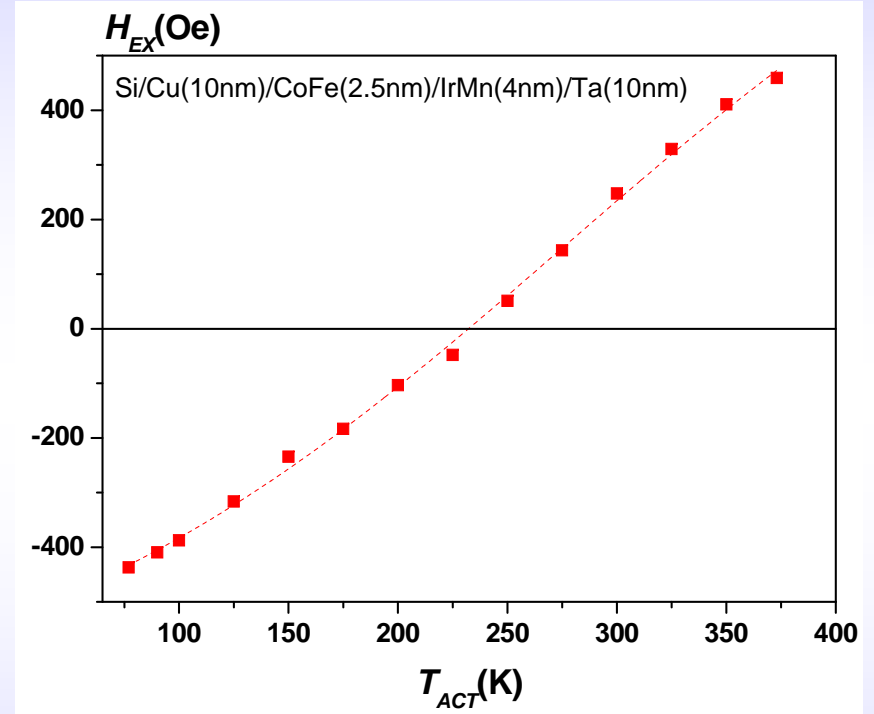
# Determination of $K_{AF}$

- We have calculated  $K$  from the zero point in the  $H_{ex}(T)$  data.
- The factor 1800 arises due to activation for 30 minutes and  $\langle V \rangle$  is obtained from TEM analysis of >500 grains.

$$K_{AF}(T_B) = \frac{\ln(1800 f_0) k_B T_B}{\langle V \rangle} \quad f_0 = 10^9 \text{ s}^{-1}$$

- The value of  $K(T)$  was obtained using:

$$K_{AF}(T) = K_{AF}(0) \cdot \left[ 1 - \frac{T}{T_N} \right]$$



$$\langle T_B \rangle : K_{AF}(236\text{K}) = 6.3 \times 10^6 \text{ ergs/cc}$$

$$RT : K_{AF}(295\text{K}) = 5.5 \times 10^6 \text{ ergs/cc}$$

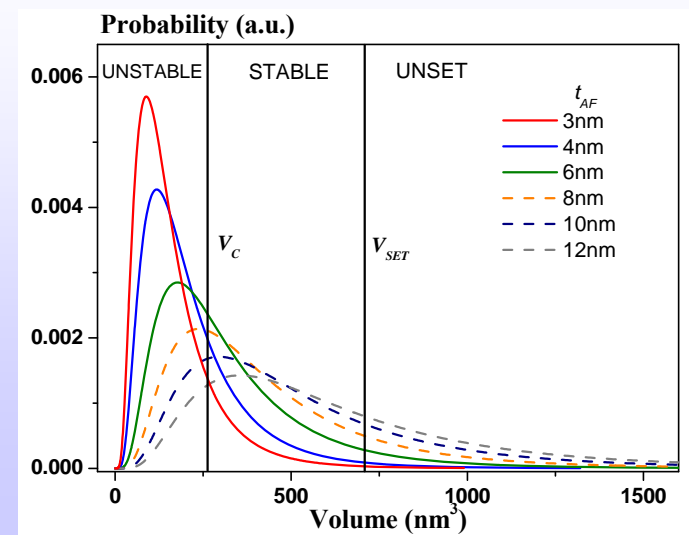
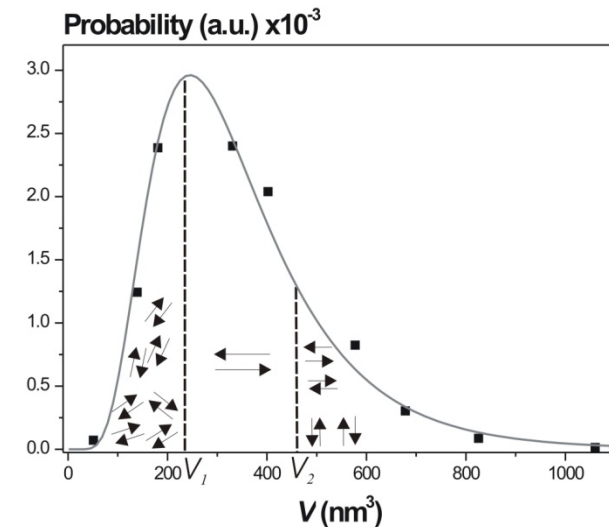
$$T_{SET} : K_{AF}(373\text{K}) = 4.4 \times 10^6 \text{ ergs/cc}$$

# Grain Size Dependence of EB

- Since the  $AF$  is 'set' by thermal activation all large grains may not be set at  $T < T_N$ .
- Small grains will be disordered by thermal energy above  $T_{NA}$ .
- Hence  $H_{ex}$  will be due to the stable and set fraction at finite temperatures.

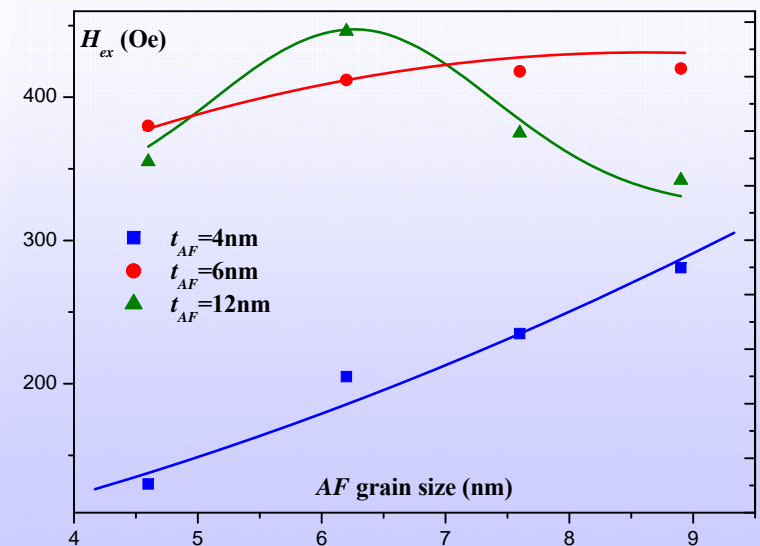
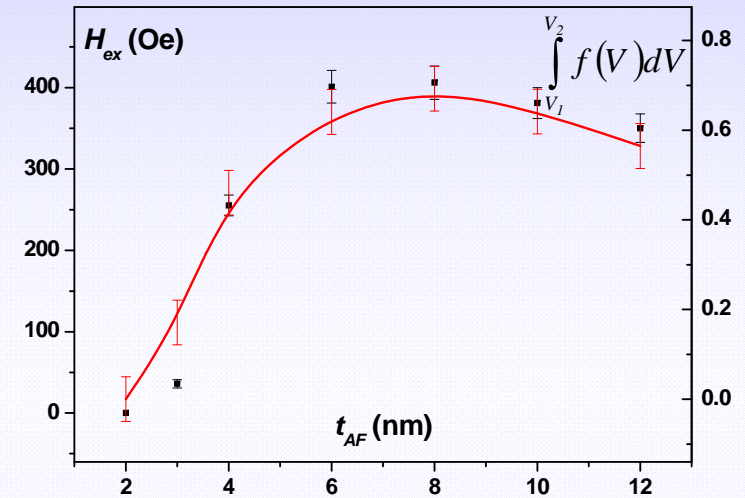
$$H_{ex} \propto \int_{V_C(T_{meas})}^{V_{set}(T_{set})} f(V) dV$$

- Grain volume was varied via the  $AF$  thickness  $t_{AF}$  and the grain size.



# $H_{ex}$ and Grain Volume

- At room temperature it is known that  $H_{ex}$  can increase or decrease with  $t_{AF}$ .
- This is due to the “stable and set” fraction of the volume distribution changing.
- The grain volume distribution can account for both variations.
- The fit between the integral and  $H_{ex}(t_{AF})$  is excellent.
- The fitting is better than that from domain wall models.





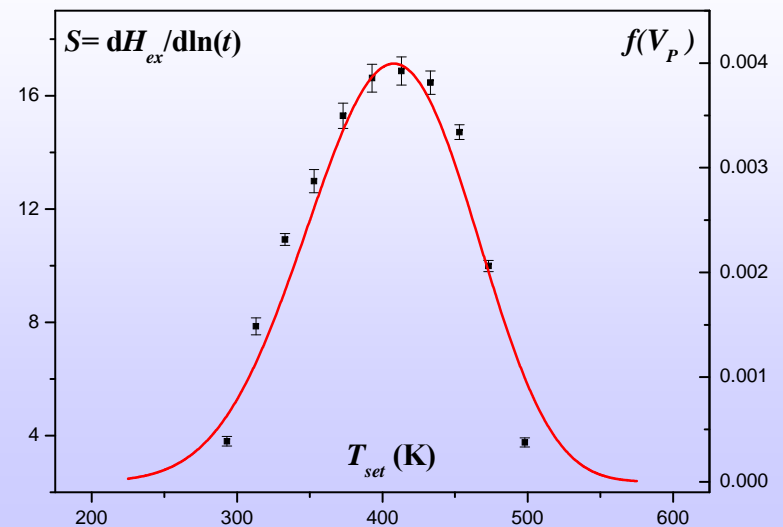
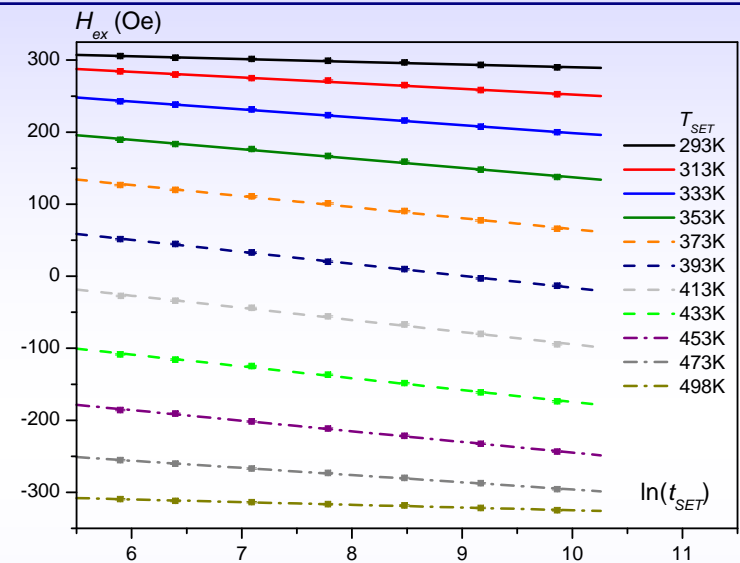
# Other Predictions

- The fit of the simple grain model to size and thickness curves is not sufficient to validate the model.
- We have also looked at the setting process because of its importance in applications.
- Because of the form of the time dependence we predict

$$S \propto f(\Delta E_C)|_T \propto f(V_P)|_T$$

- Again the simple model works and gives the correct form.

G. Vallejo-Fernandez, N.P. Aley, L.E. Fernandez-Outon and K. O'Grady, *J. Appl. Phys.* **104** 033906 (2008).

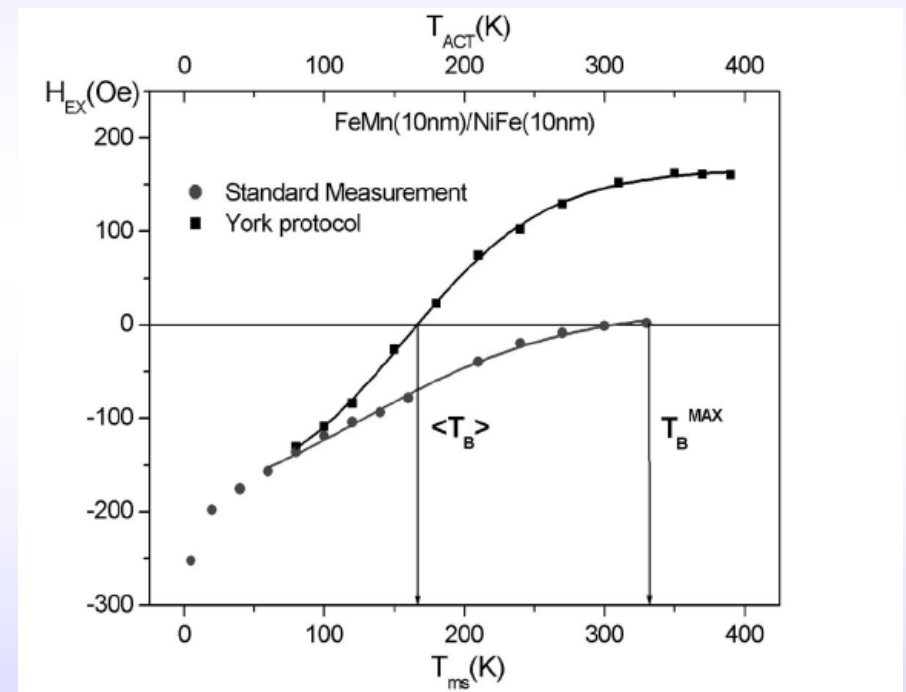


# Interfacial Spin Order

- We have observed a dramatic increase in  $H_{ex}$  at low temperatures.
- As the bulk of the  $AF$  is stable, this must be due to changes at the interface.

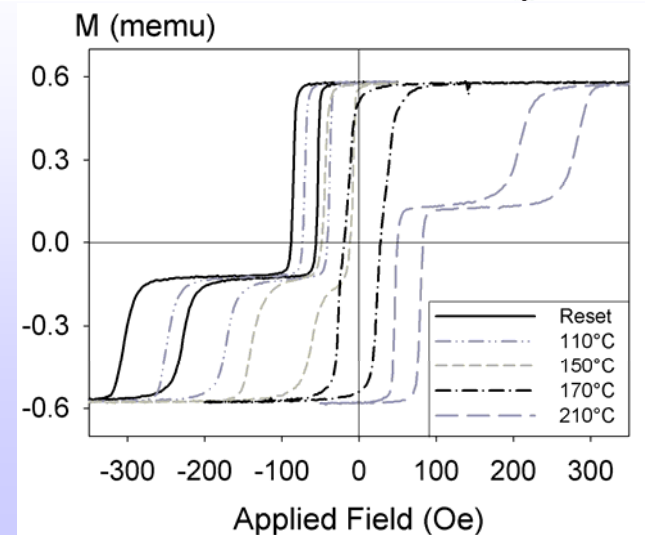
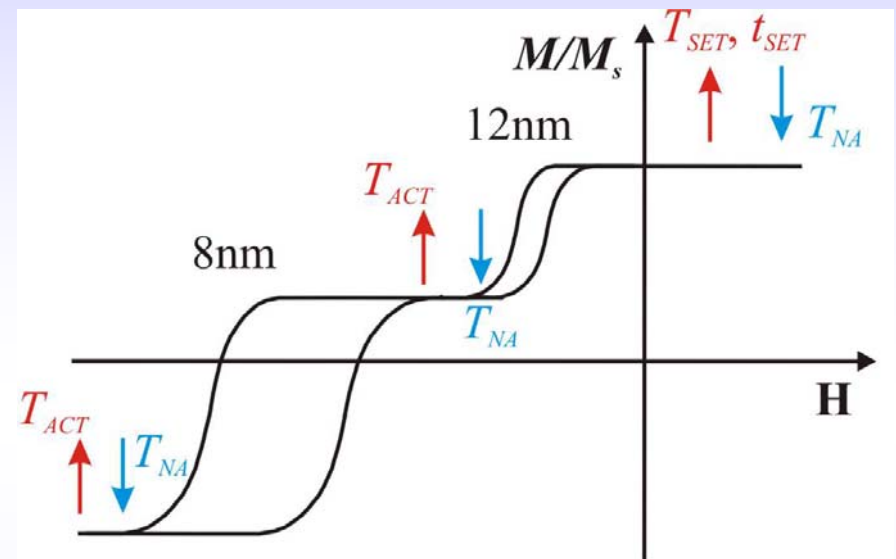
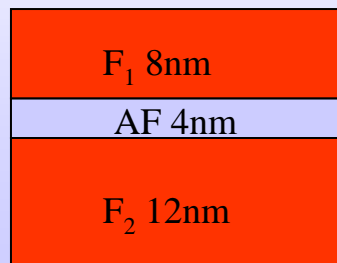
$$H_{ex} \propto C^*(?,?) \int_{V_c}^{V_{set}} f(V) dV$$

- We know that  $C^* < 1$  to fit the results.
- The low temperature data indicate a change in order similar to that in spin glass freezing.



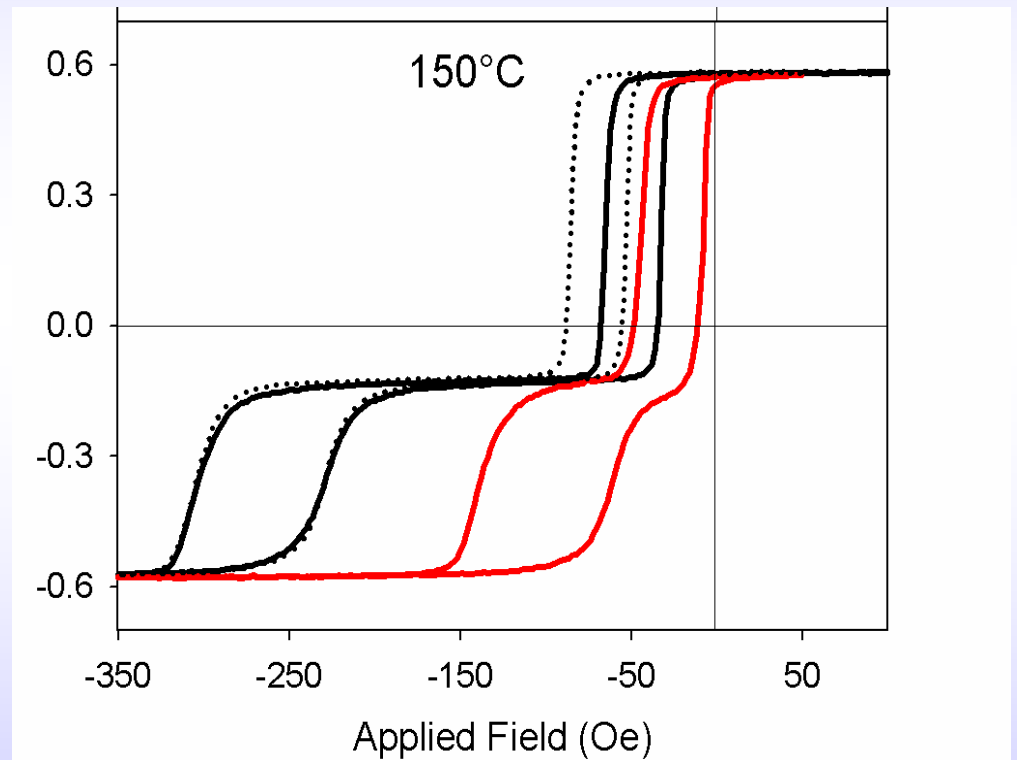
# Interfacial Spin Effects

- We have done an experiment on trilayer systems with different  $F$ .
- This systems produces two distinct hysteresis loops.
- The  $AF$  can be thermally activated as before.
- Both loops shift as the bulk and interfaces are reordered.



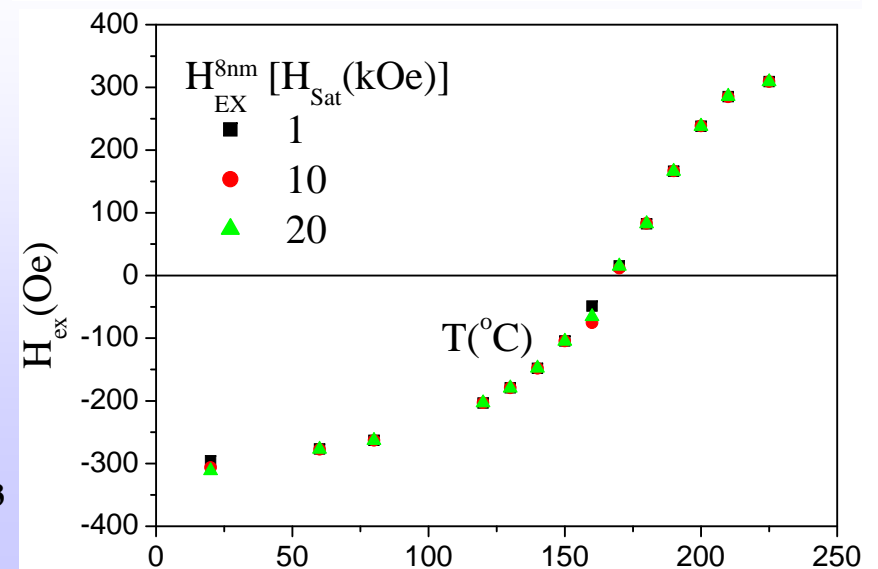
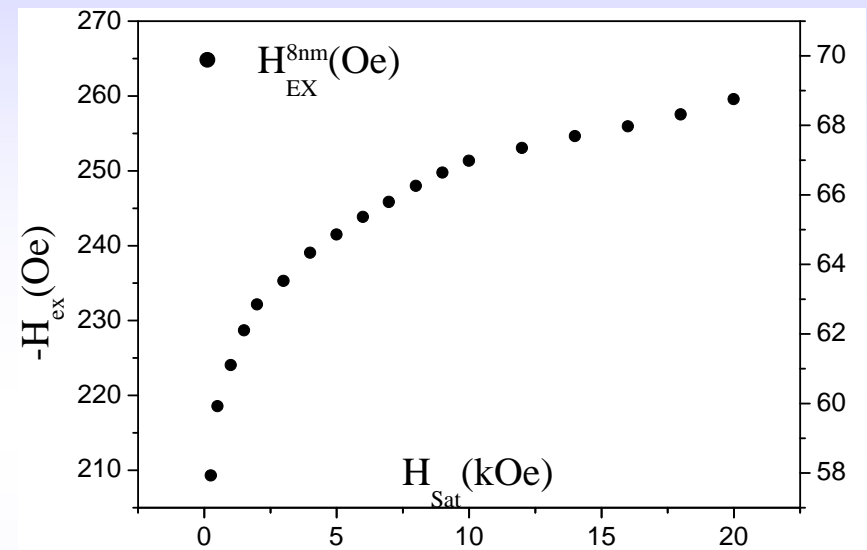
# Single Interface Activation

- We can also activate the *AF* at one interface only so that only the 12 nm layer is measured.
- This shifts only the loop for the 12nm layer and to a lesser degree.
- The loop for the 8 nm layer is not moved at all.



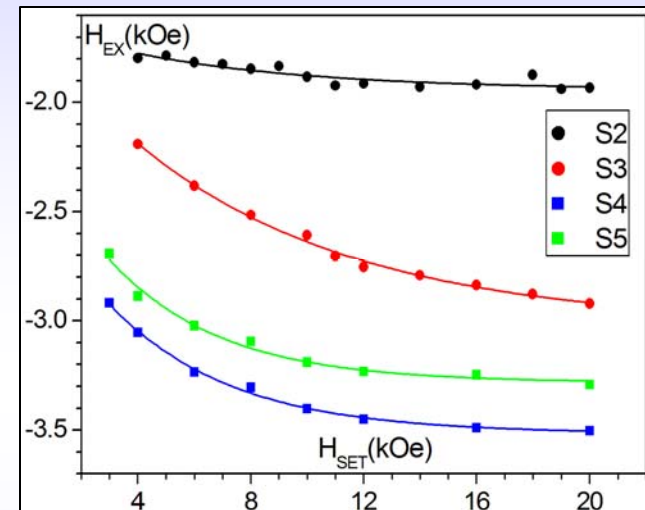
# Field ‘Setting’ of Interfaces

- Interfacial spins can also be set by high fields.
- At low fields ( $H < 500$  Oe) the exchange field from the F layer aligns the interfacial spins.
- Higher fields increase  $H_{ex}$  due to improved interfacial spin alignment.
- We know this is not a bulk  $AF$  effects since  $f(T_B)$  is unaffected.



# Interfacial Spin Effects

- The state of order of the interfacial spins, represented by  $C^*(H,T)$ , is altered by the application of  $H_{set}$ .
- The F/AF coupling is due to the order of the interfacial spins.
- When the coupling is strong  $H_{ex}$  is larger and  $H_c$  smaller.
- Increasing  $H_{set}$  increases  $H_{ex}$  but does not change  $\langle T_B \rangle$ .
- $H_c < 20\% H_{ex}$  for our samples. (3% for S2)



$H_{ex}$ (kOe) measured after setting at $H_{set}$	SAMPLE			
	S2	S3	S4	S5
$H_{ex}(4\text{kOe})$	-1.796	-2.188	-3.537	-2.887
$H_{ex}(20\text{kOe})$	-1.936	-2.922	-3.700	-3.292
$\Delta H_{ex}$ (%)	8	37	14	14
$H_c$ (kOe)	0.055	0.540	0.200	0.130
$H_c/H_{ex}$ (%)	2.8	18.5	5.4	3.9

# Design of AFs

- The width of  $f(T_B)$  means that it is difficult to have a stable system that can be set.

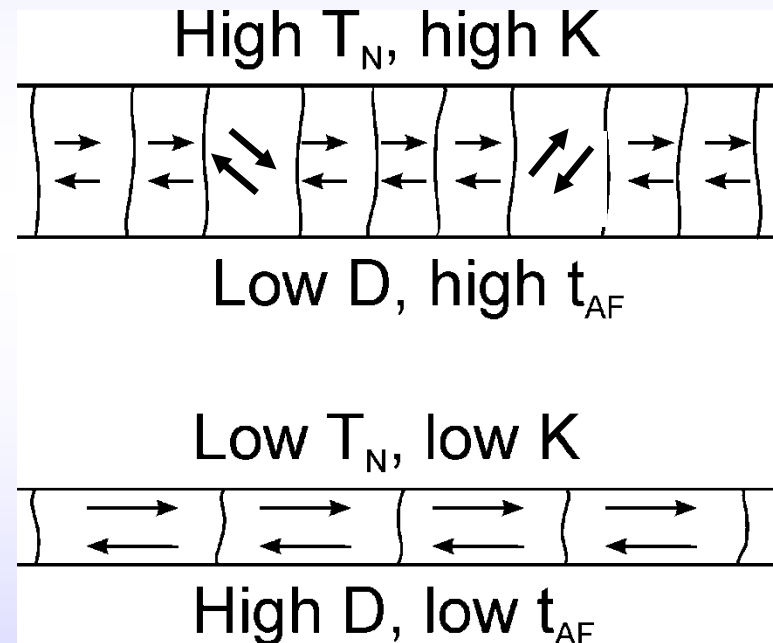
- It would be best to set at:

$$T > T_N$$

- That would lower  $K$  and  $T_B$  unless large grains were used.

$$\langle T_B \rangle \propto \Delta E = KV(1-H^*/H_K^*)^2$$

- The ideal would be low  $T_N$  and  $T_B$  with large grains and low  $t_{AF}$ .



# Conclusions

- We now have an understanding of the blocking process and interface required to get optimum  $H_{ex}$ .

$$H_{ex} = H_{ex}^i C^*(H_{set}, T) \int_{V_c}^{V_{set}} f(V) dV$$

- This is the first formulaic description of exchange bias.
- It is unlikely that a grain size distribution could be made sufficiently narrow to meet the setting and stability requirements.
- This can be overcome by setting above  $T_N$  and using big grains to avoid thermal instability.
- It is not yet clear how interfacial spin order can be controlled other than by field setting.