30th N R Kamath & Mrs. Ruzena Kamath Oration (2014)

In the Centenary year of Professor N R Kamath (b: 8.9.1914)

New Functional Materials for Spintronics and Magnetic STM Imaging

A. K. Grover*

Vice Chancellor
Panjab University, Chandigarh, 160014
and
DCMPMS, School of Natural Sciences,
Tata Institute of Fundamental Research, Mumbai, 400005

***Based on work done in association with Post-Doctoral Fellows (in particular, Dr. Prasanna D Kulkarni, S.Venkatesh and Dr. Swati Pandya) and other colleagues (S. K. Dhar, A. Thamizhavel, S. Ramakrishnan and Ulhas Vaidya) at TIFR, Mumbai.**

Sixty Seventh Annual Meeting of Institution of Chemical Engineers (CHEMCON 2014) , P U Auditorium, Chandigarh 160014

Story of a contemporary topic in Magnetic Materials

Motivation

* Extend the scope of STM Imaging for magnetic Imaging

(STM : Nobel Prize, 1986)

* Simplify structure of Giant Magneto-Resistance (GMR) effect based Spin Valve Device in the READ HEADS of Hard Disks of Computers

(GMR : Nobel Prize, 2007)

Spin Valve Devise that reads the magnetic information stored on the Hard Disk belongs to realm of Spintronics

What is Spintronics ?

Spintronics, also known as magneto-electronics, is an emerging technology that exploits the intrinsic spin of the electron and its associated magnetic moment, in addition to its fundamental electronic charge in solid-state devices

In such devices a distinction is made for the (polarized) electrical current in two spin channels depending on their spin orientation with respect to some axis such as an applied magnetic field.

A small digression

History of nomenclature 'Magnet'

"It was probably the Greeks who first reflected upon the wondrous properties of magnetite, the magnetic iron ore $FeO\text{-}Fe_2O_3$ and famed lodestone (leading stone, or compass). This mineral *is believed* to have been mined in the province of Magnesia" *

The magnet's name the observing Grecians drew

From the Magnetick region where it grew[1]

[1]English translation by Th. Creech (1974, London),

from Lucretius Carus, De Rerum Natura, 1st century B. C.

***First para of Chapter 1:** **"The Theory of Magnetism" by Daniel C. Mattis**
HARPER & ROW Publishers Inc. (New York, 1965)

Plan of Story

Background Information :

* Primer on atomic characteristics of Rare Earth elements in the Periodical Table, in particular, the special attributes of element Samarium

* Explain rationale for NET Zero-Magnetization (ZM), possible via *Spin-Ferromagnetism* (SF) that prevails in admixed Rare Earth (RE) systems.

Highlights of Studies at TIFR in recent years :

* Discovery of tunable Exchange Bias as a new functionality in ZMSF.

* Elucidation of *(long awaited)* self-magnetic compensation in Samarium based Ferromagnets.

* New physics issues relating to magnetic compensation in Ferri-magnets.

A long Prologue to the story

An extract from a text book titled

(Cambridge University Press, UK, 2011)

"Magnetic Materials: Fundamental and Applications"

by **Nicola A Spaldin** ***(recipient of APS McGroddy Prize for New Materials)***

Section 9.4: Half-metallic Anti-Ferromagnets

(oxymoron?)

Half-metallic **Anti-Ferromagnets** are a class of materials which have been **predicted theoretically** **but not yet synthesized**. We include them here in part for **some light entertainment**, but also to illustrate that there is **still great potential in the search for new magnetic materials** with **novel** and possibly **technologically relevant** properties.

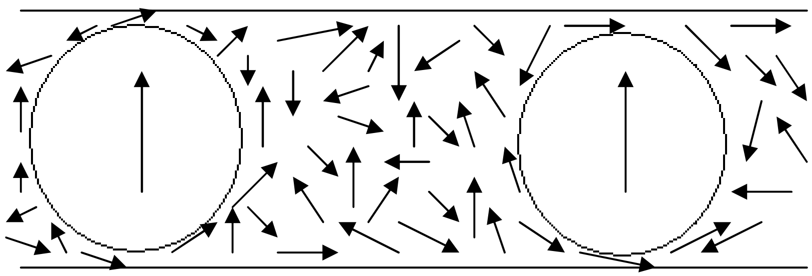
“Magnetic Materials: Fundamental and Applications”

Half-metallic materials are defined as those which are **metallic for one spin** direction (up-spin, say) but **insulating for the other spin** channel (down-spin).

Such a situation can happen in **Ferromagnetic intermetallics** or alloy systems, where well separated large **Local Magnetic Moments** are **coupled** via the **polarization** of the **conduction band electrons**.

The *RKKY (Rudermann-Kittel-Kasuya-Yoshida)* Interaction:

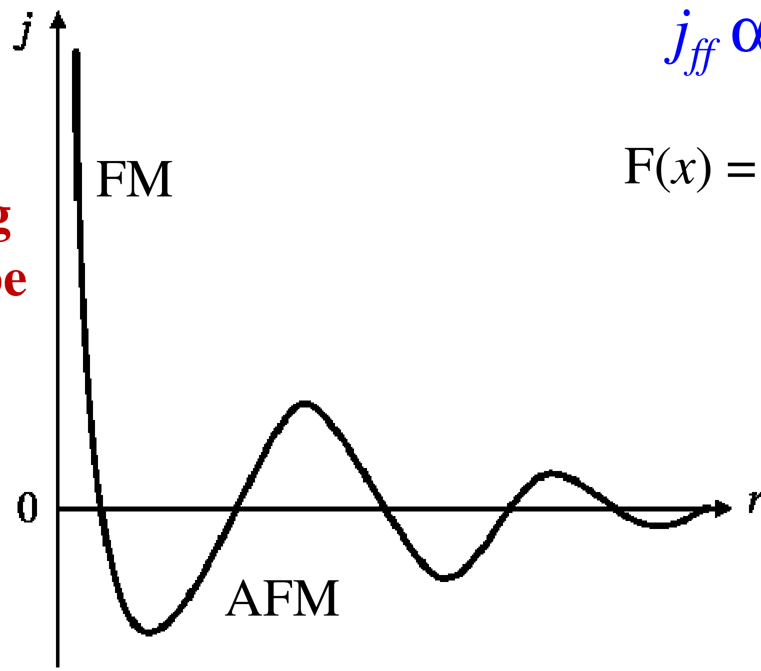
Governs coupling between Spins of Magnetic Ions



Indirect Exchange: mediated by local ***s-f*** interactions (J_{sf}) by the conduction electrons.

Spin of an Ion

Spin of another ion



$$j_{ff} \propto -J_{sf}^2 \, \Sigma \, F(x)$$

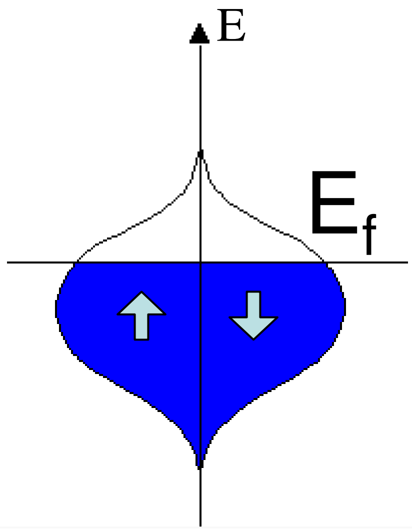
$$F(x) = (x \cos x - \sin x) \, / \, x^4$$

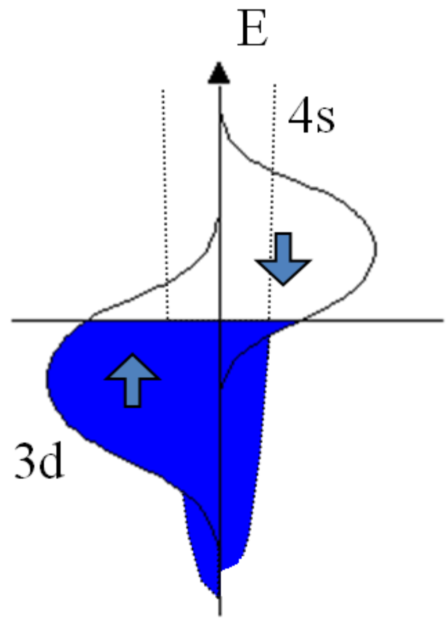
$$x = k_F r$$

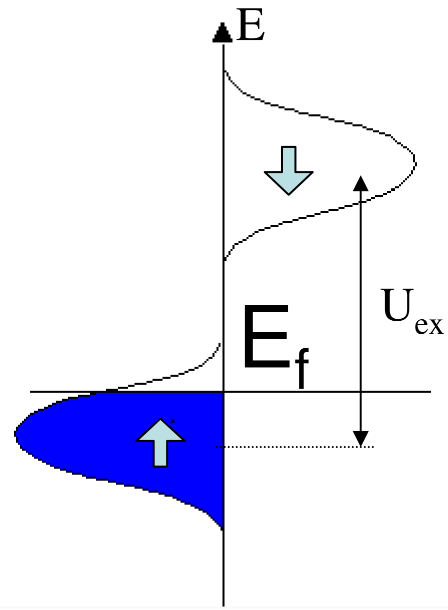
Materials comprising local ionic spins can be Ferromagnetic or Anti-Ferro depending on $k_F r$

Variation of coupling constant j_{ff}, with distance (r) from an ionic spin.
It is oscillatory and damped in spatial variation.

Schematic band diagrams







Non-magnetic Metal

(equally populated and Metallic in both spin directions)

3d-4s Ferromagnet

(Unequally populated in s and d-channels , but metallic in both spin directions)

Half-Metallic system (It is indeed a Ferro-magnet)

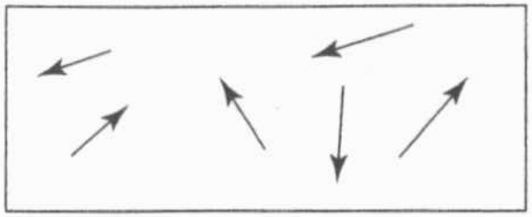
(But what is desired is a material with (near) Zero Net Magnetization, like, in an Anti-Ferromagnet)

Half-Metallic Anti-Ferromagnets

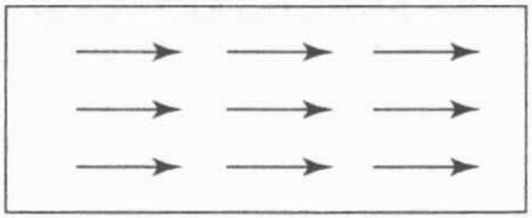
- The properties of half-metallic Anti-Ferromagnets are **unusual**. …… since there is **no net internal magnetization**, half-metallic Anti-Ferromagnets would **not generate a magnetic field, in spite of their fully magnetized currents**. This would be a particularly **desirable** property, for example in **spin-polarized Scanning Tunneling Microscopy (STM)** , …**Currently such experiments are complicated** by the existence of a **permanent magnetic tip** (*required to produce the spin-polarized electrons*) **close to the magnetic surface being investigated.**

* In the context of **Half-metallic Anti-Ferromagnets, realistically** one can conceive of ***pseudo-Ferrimagnets,*** in which the magnetizations of the **two different magnetic sub-lattices can** (*somehow*) **cancel out** (*atleast over some temperature region*).

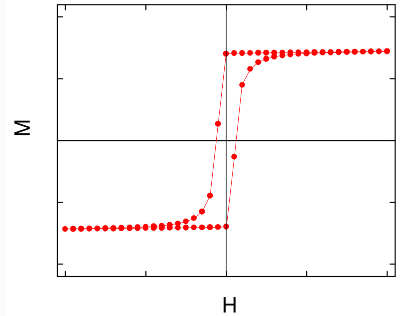
Schematic M-H curves of different arrangements

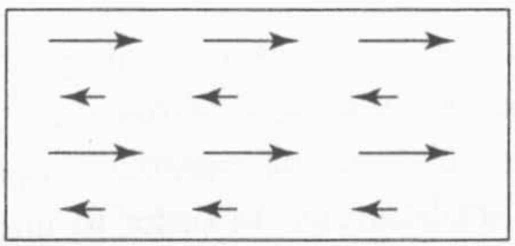


(a) Paramagnetic

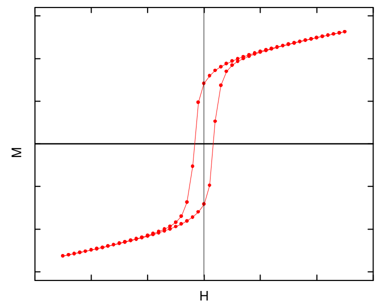


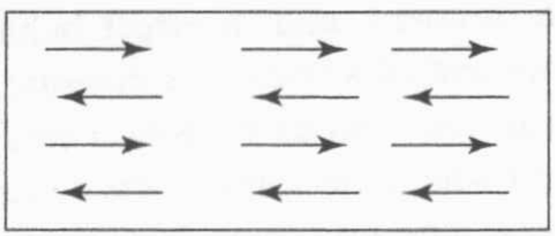
(b) Ferromagnetic



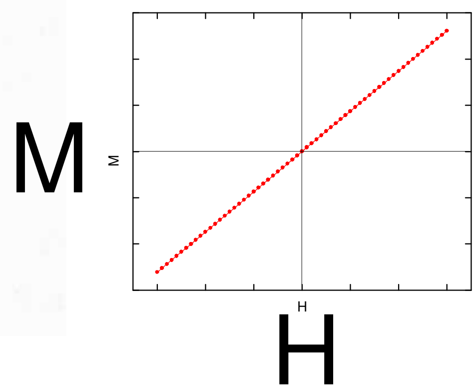


(c) Ferrimagnetic

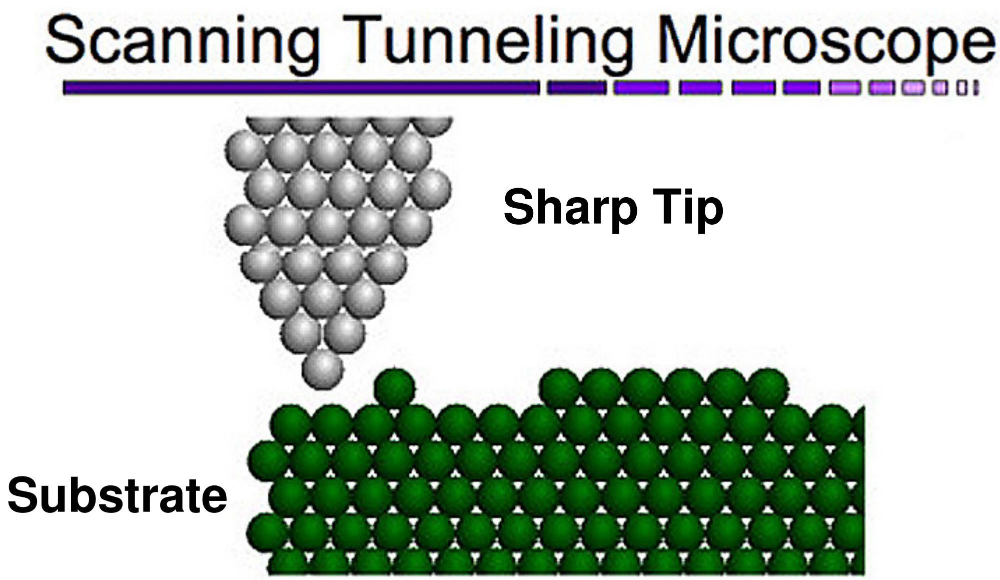


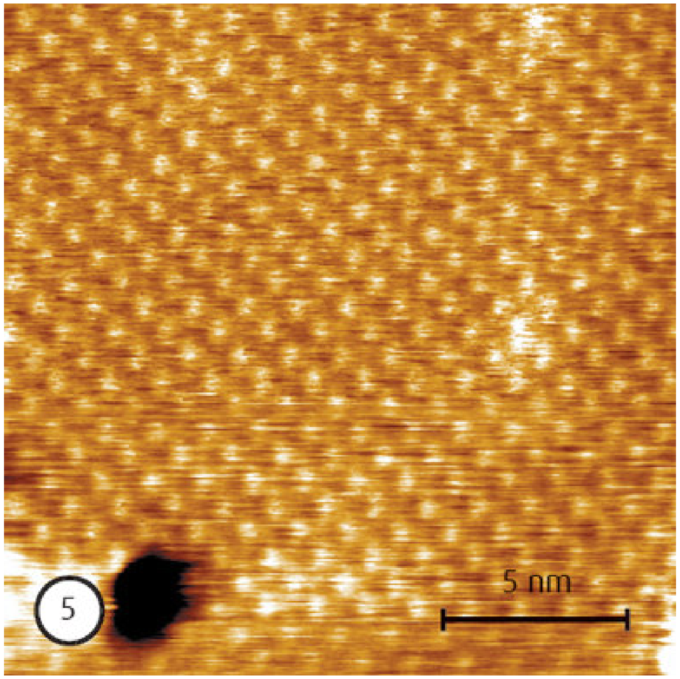


(d) Antiferromagnetic



STM : Nobel Prize 1986 (Gerd Binning and Heinrich Rohrer)



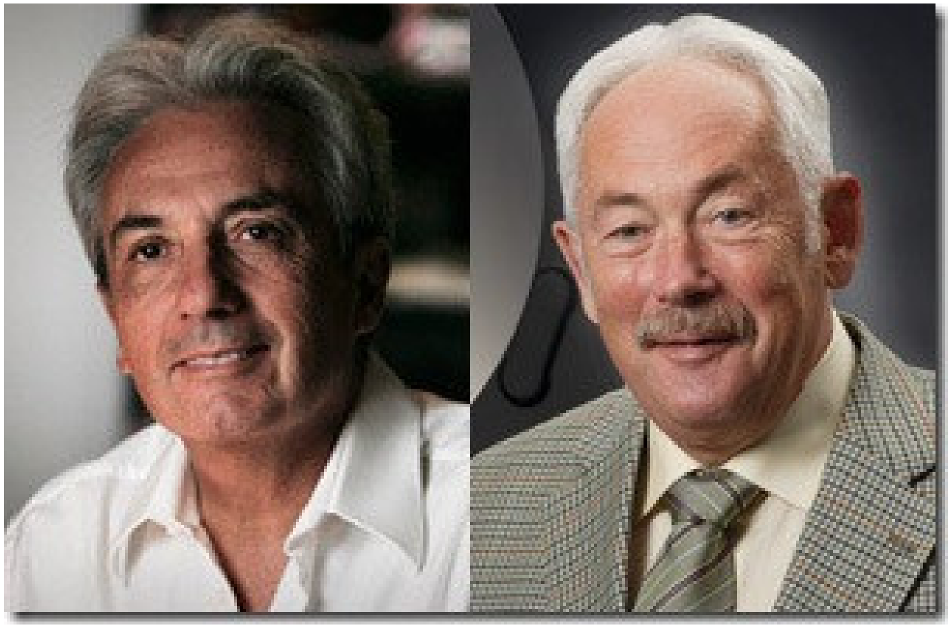


STM image of evaporated silver film

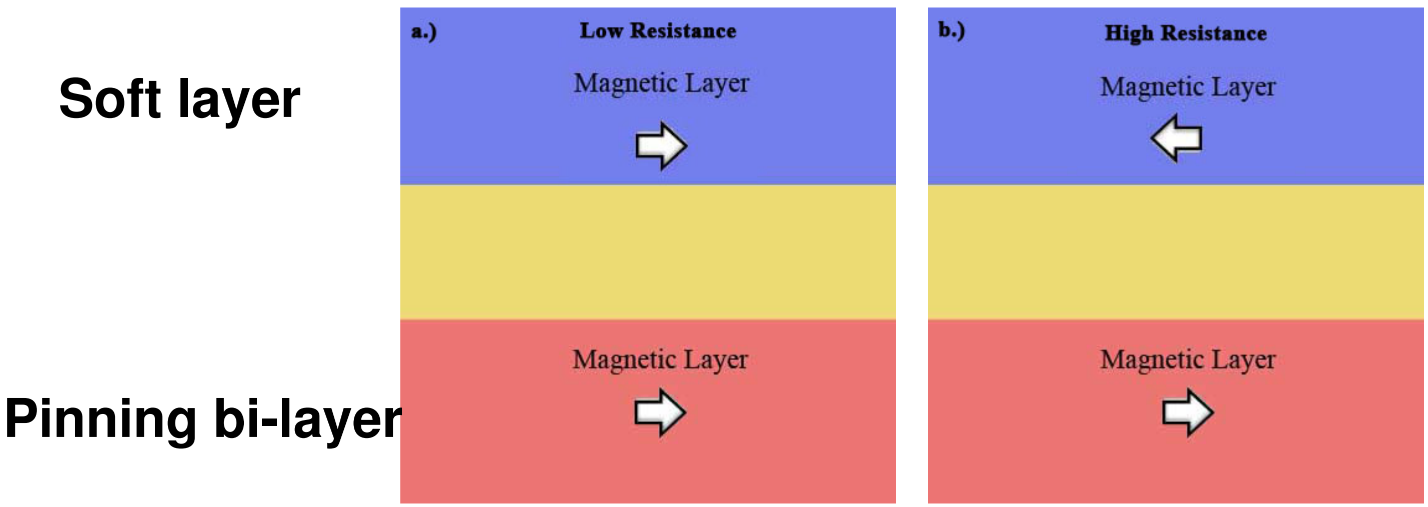
Heinrich Rohrer and Gerd Binning were recognized for developing the powerful microscopy technique, which can form an image of individual atoms on a metal or semiconductor surface by scanning the tip of a needle over the surface at a height of only a few atomic diameters.

GMR: Nobel prize in Physics, 2007

Giant Magneto-Resistive Effect was discovered in 1988 in Fe/Cr/Fe trilayers by a research team led by Peter Grünberg of the Jülich Research Centre (DE), and independently in Fe/Cr multilayers by the group of Albert Fert of the University of Paris-Sud (FR).

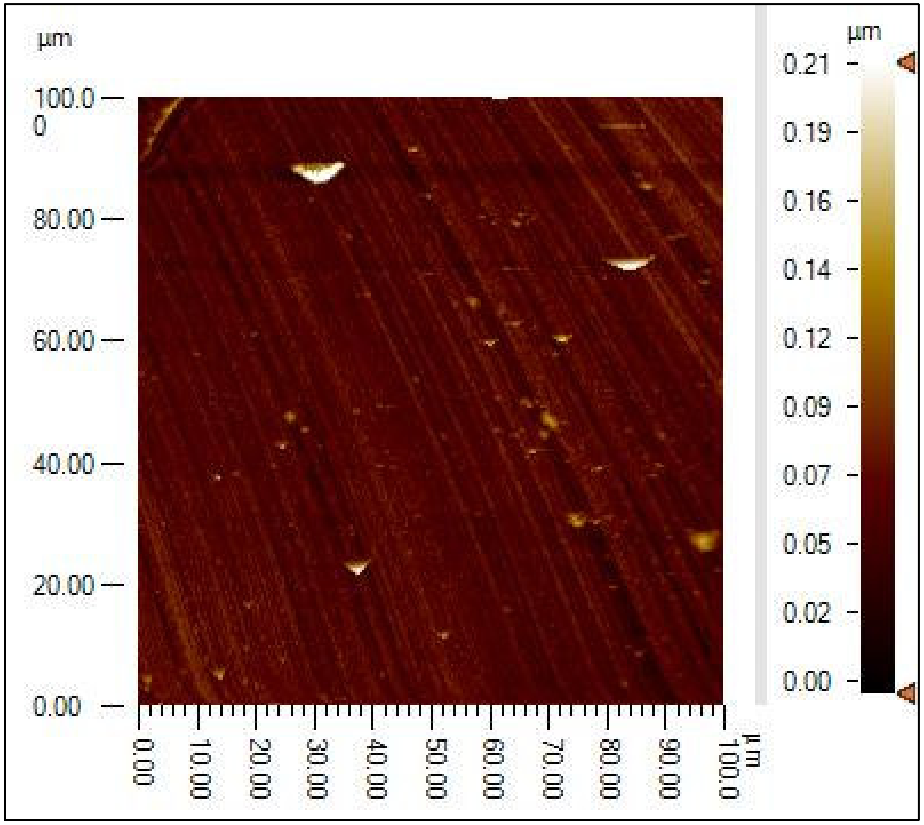


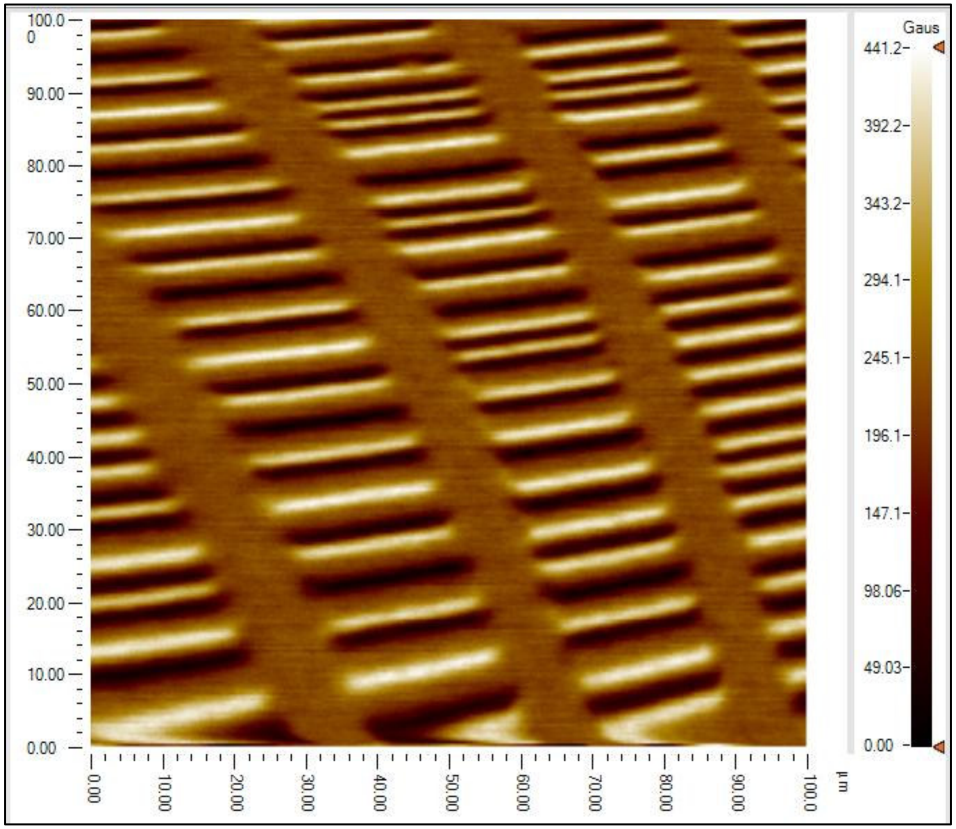
Two states of a magnetic bit yield different resistance values



GMR Effect forms the basis of functionality of multi-layer Spin Valve Device, which yield different resistance values for two states of a magnetic 'bit' in every day used Hard Disc.

Hard disk image

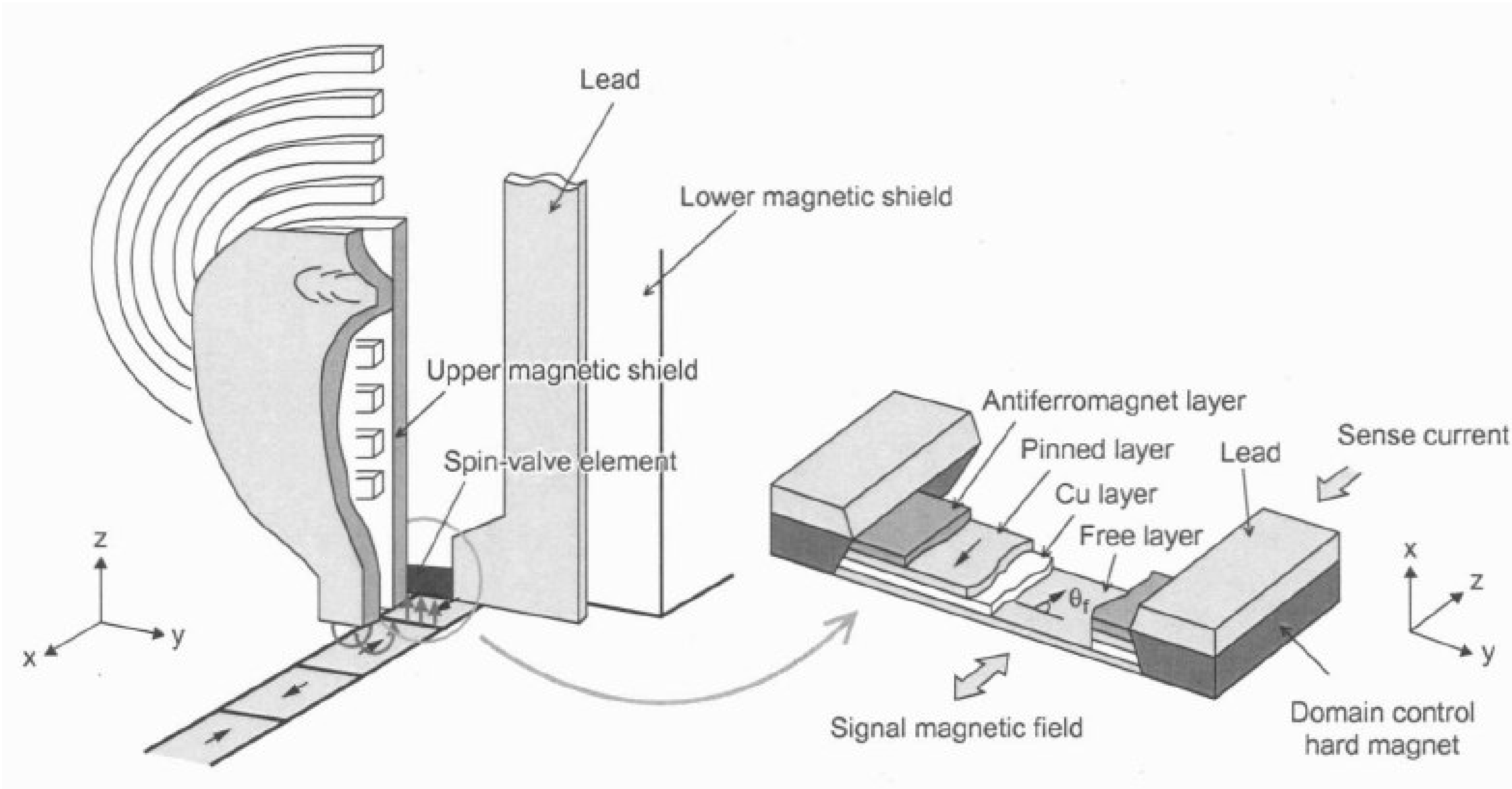




Tracks on the Surface

Stored Magnetic information

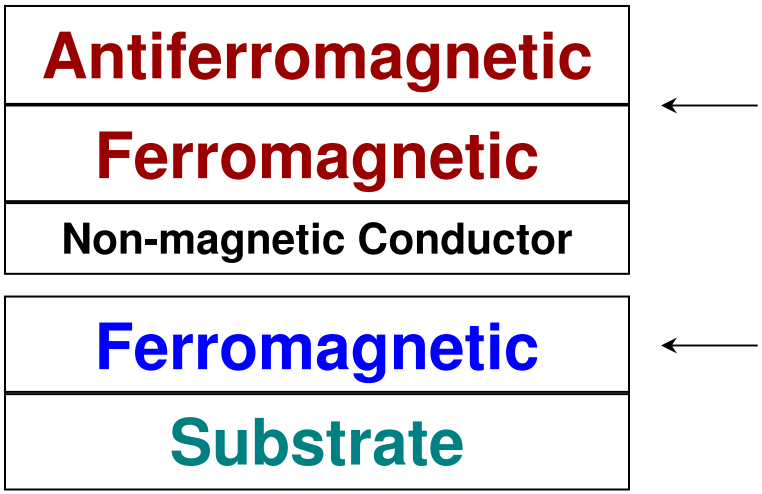
Different components of a Spin valve read head



H. Kanai et al, Advanced Spin-Valve GMR Head, FUJITSU Sci. Tech. J., 37, 174-182 (2001)

SPIN-VALVE DEVICE

A Spin-Valve device consists of layers of two (or more) conducting magnetic materials.

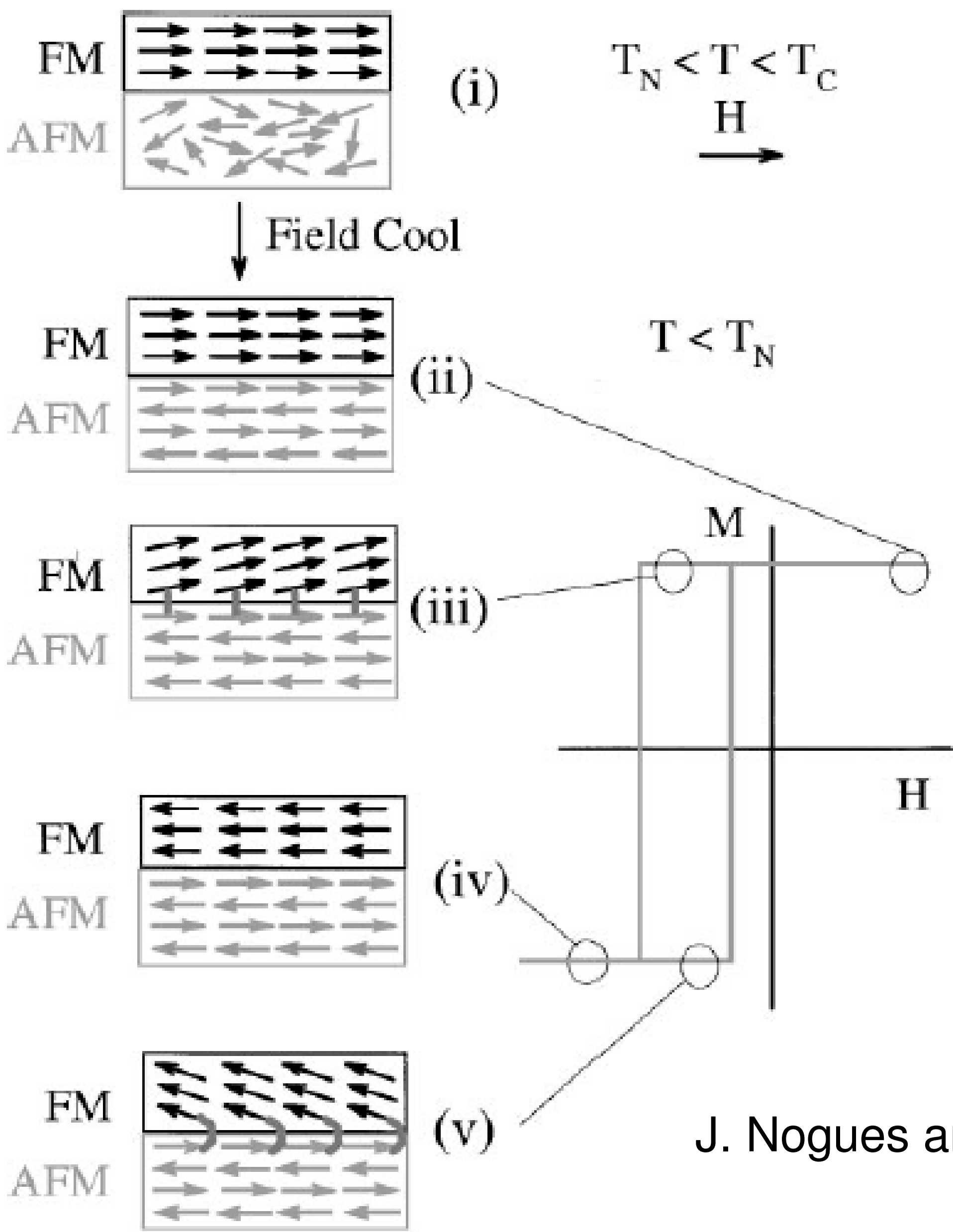


Pinned layer with an Exchange Bias, i.e., Shifted Hysteresis Loop

Soft layer with no Bias,It faithfully aligns with the direction of magnetic bit

It alternates its electrical resistance (from low to high or high to low) depending on the alignment of the magnetic layers, while exploiting the Giant Magneto-Resistive (GMR) effect.

EXCHANGE BIAS ATTRIBUTE IN FM/AFM BI-LAYER OF A SPIN VALVE



SPIN VALVE

Antiferromagnetic
Ferromagnetic
Conductor
Soft-Ferromagnet
Substrate

J. Nogues and I. K. Schuller, *JMMM* **192,** 203 (1999).

A possible material for Magnetic STM Imaging

letters to nature

....

A ferromagnet having no net magnetic moment

(should be read as 'no net-magnetization')

H. Adachi*[2] & H. Ino*[3]

* Department of Materials Science, the University of Tokyo, Tokyo 113, Japan

[2] Institute of Materials Structure Science, **KEK**, Tsukuba, Ibaraki 305, Japan

[3] College of Engineering, Hosei University, Koganei, Tokyo 184, Japan

NATURE |VOL 401 | 9 SEPTEMBER 1999 | www.nature.com

Ferromagnet $SmAl_2$ on replacement of few percent of Sm by Gd loses its magnetization, ***and when this happens ….***

….it exhibits the seemingly incompatible properties **of large spin polarization, but no bulk magnetization.**

…. attributes should be **generic** to ferromagnets containing trivalent **Samarium ions.**

…. potential application in, for example, **spin-resolving devices for charged particles.**

US Patent : 6558481 B1

Hiromichi Adachi, Hiromitsu Ino & Hirosi Miwa

Filed: Sept.17,1999 ;   Granted: May 6, 2003

BASED ON THEIR TWO PAPERS

(i) Effect of conduction-electron polarization on the magnetism of Samarium metal
(Phys. Rev B 56, 349, 1997)

(ii) Separation of the 4*f*-spin, 4*f*-orbital, and conduction-electron magnetization … for ferromagnetic Sm intermetallics
(Phys. Rev. B 59, 11445, 1999)

US Patent : 6558481 B1

<u>Hiro</u>michi Adachi, <u>Hiro</u>mitsu Ino & <u>Hiro</u>si Miwa

Patent title: ‘Method of controlling properties of a ferro-magnetic Sm based material and a spin resolving device’

Metallic materials with no self-stray field are useful:

(i) To generate a spin polarized charge-particle beam.

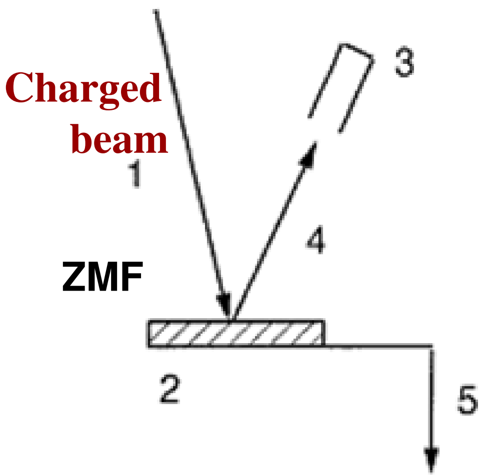
(ii) To measure spin polarization/structure on a surface (e.g., magnetic STM imaging).

(iii) To polarize or analyze the spin for charged-particle flow.

Schematics:

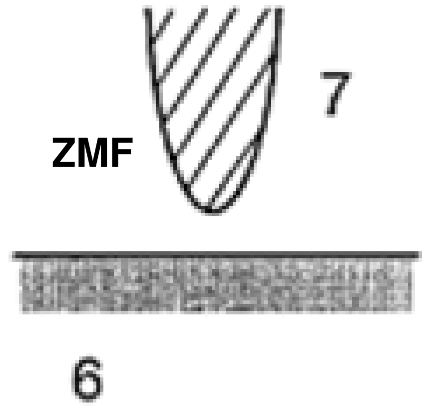
(Extracted from sheet #4 of U.S. patent: 6558481B1)

(a) To measure spin polarization of charged beam



Spin asymmetry in the scattering or diffraction intensity of beam 4 or that in the observed electric current (5)

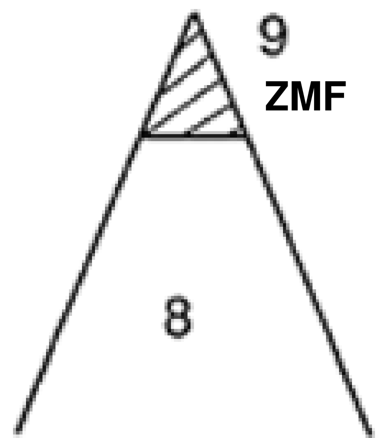
(b) To measure spin structure of surface by tip made up of ZMF



Spin asymmetry in the tunneling electric current on exchange force between the surface of the magnetic sample (6) and the ZMF tip could make it possible to observe surface spin structure

Magnetic STM Imaging

(c) Spin polarized electron gun



ZMF: Zero Magnetization Ferromagnet

Newer Functionality of Zero-Magnetization materials as a Pinning Layer (via Exchange Bias field) in SPIN VALVE DEVICES

 # Shift in centre of gravity of the M-H loop is termed as an Exchange Bias (EB) in the literature

 # Noted in 2% Gd doped $SmAl_2$ by C W (Paul) Chu's group in 2005 , see, Phys. Rev. B 72, 054436 (2005).

PHYSICAL REVIEW B **82**, 174421 (2010)

Using a zero-magnetization ferromagnet as the pinning layer in exchange-bias systems

M. Ungureanu,[1] K. Dumesnil,[1] C. Dufour,[1] N. Gonzalez,[1] F. Wilhelm,[2] A. Smekhova,[2] and A. Rogalev[2]

SA / SGA Bilayer assembly displays EB
SA: $SmAl_2$ (Ferromagnet)
SGA:$Sm_{0.972}Gd_{0.028}Al_2$
(Zero-Magnetization Spin-Ferromagnet)

2010: First results towards use of doped Sm-Ferromagnets as a pinning layer

Exchange Bias is a *Generic* attribute of Rare Earth based Zero-Magnetization systems

2902 IEEE TRANSACTIONS ON MAGNETICS, VOL. 45, NO. 7, JULY 2009

Exchange Bias and Its Phase Reversal in Zero Magnetization Admixed Rare-Earth Intermetallics

Prasanna D. Kulkarni, S. Venkatesh, A. Thamizhavel, V. C. Rakhecha, S. Ramakrishnan, and A. K. Grover

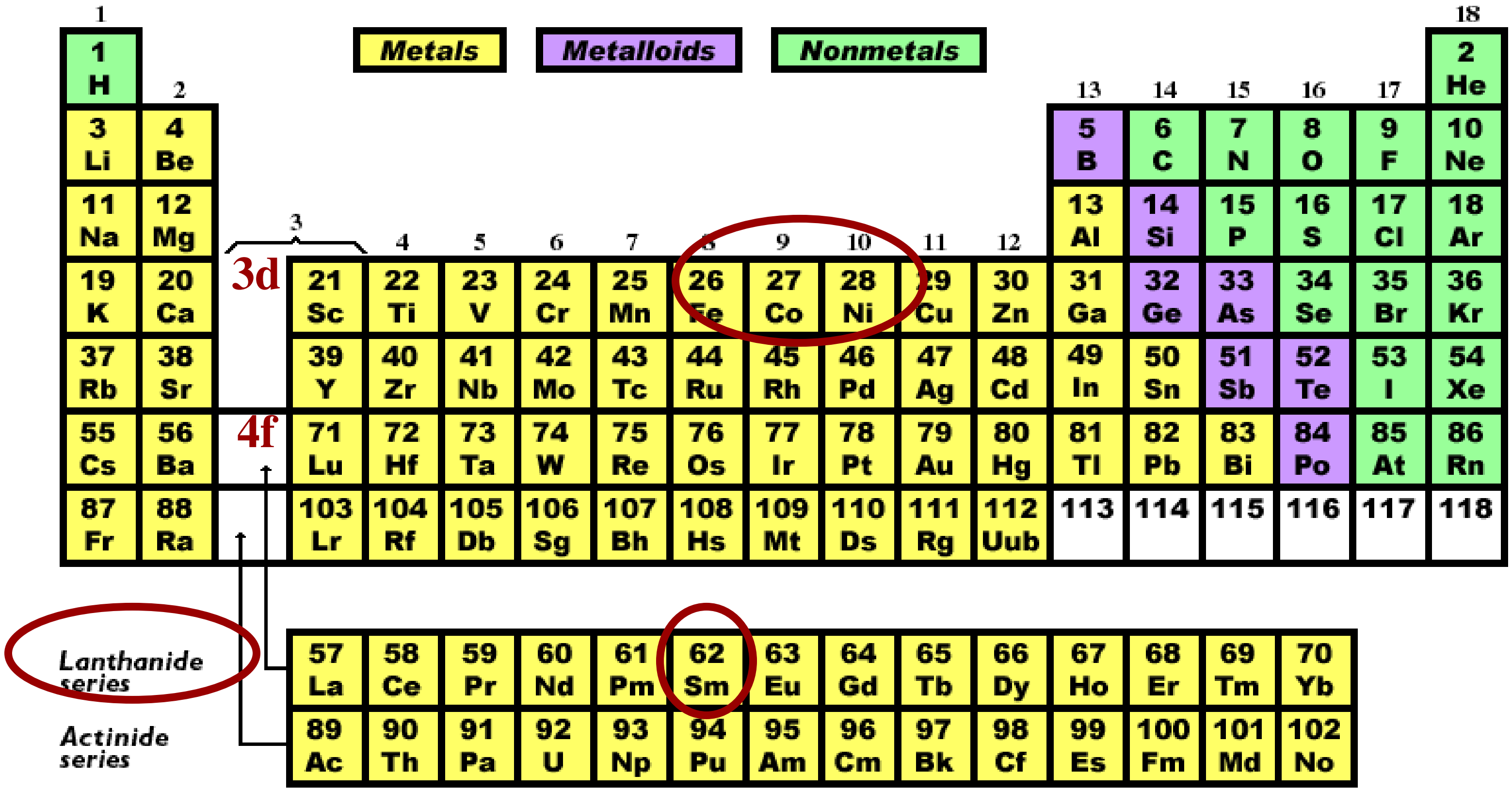
Magnetic compensation phenomenon and the sign reversal in exchange bias in a single crystal of $Nd_{0.75}Gd_{0.25}Al_2$

P D Kulkarni e*t al*., *Euro Physics Letters 86, 47003, 2009*

Premise of studies at TIFR

Special attributes of net zero-magnetization materials derived from ferromagnetic Samarium compounds are not specific to Sm ions but these are generic to admixed rare earth alloys, formed by mixing together any two ferromagnetic rare earth intermetallic compounds , belonging to two different halves of the 4f-Rare Earth series.

The Periodic Table



Magnetic properties of some elements

Material	Curie Temperature T_c (K)	Bohr Magnetons per Atom (μ_B)
Fe (3d)	1040	2.22
Co (3d)	1395	1.72
Ni (3d)	630	0.61
Gd (4f)	290	7.1
Sm (4f)	160	**~ 0.1**

Samarium is the 5th element of the 4f-Rare Earth series (At. no.: 58-71), having small moment for Sm metal

The ground state properties of the Rare Earth (RE^{3+}) ions

	m_L	$+3$	$+2$	$+1$	0	-1	-2	-3	S	L	J	g_J	$g_J J$
$4f^1$	Ce^{3+}	↑							$\frac{1}{2}$	3	5/2	6/7	15/7
$4f^2$	Pr^{3+}	↑	↑						1	5	4	4/5	16/5
$4f^3$	Nd^{3+}	↑	↑	↑					3/2	6	9/2	8/11	36/11
$4f^4$	Pm^{3+}	↑	↑	↑	↑				2	6	4	3/5	12/5
$4f^5$	Sm^{3+}	↑	↑	↑	↑	↑			5/2	5	5/2	2/7	5/7
$4f^6$	Eu^{3+}	↑	↑	↑	↑	↑	↑		3	3	0	–	0
$4f^7$	Gd^{3+}	↑	↑	↑	↑	↑	↑	↑	7/2	0	7/2	2	7
$4f^8$	Tb^{3+}	↑↓	↑	↑	↑	↑	↑	↑	3	3	6	3/2	9
$4f^9$	Dy^{3+}	↑↓	↑↓	↑	↑	↑	↑	↑	5/2	5	15/2	4/3	10
$4f^{10}$	Ho^{3+}	↑↓	↑↓	↑↓	↑	↑	↑	↑	2	6	8	5/4	10
$4f^{11}$	Er^{3+}	↑↓	↑↓	↑↓	↑↓	↑	↑	↑	3/2	6	15/2	6/5	9
$4f^{12}$	Tm^{3+}	↑↓	↑↓	↑↓	↑↓	↑↓	↑	↑	1	5	6	7/6	7
$4f^{13}$	Yb^{3+}	↑↓	↑↓	↑↓	↑↓	↑↓	↑↓	↑	$\frac{1}{2}$	3	7/2	8/7	4

The ground state is given by-
J = L - S, for **less** than **half** filled
J = L+ S, for **more** than **half** filled.

L - orbital angular momentum,
S - spin angular momentum and
J - total angular momentum

Lande's g_J-factor: $g_J = 1 + [J(J+1) - L(L+1) + S(S+1)]/2J(J+1)]$

$\mu_{eff} = g_J\,[J(J+1)]^{1/2}\,\mu_B$

(paramagnetic state)

$\mu_{sat} = g_J\,J\,\mu_B$

(ferromagnetic state)

Magnetic Moments of Free Rare Earths Ions

$<L_z> = J_z\,(g_J - 2),$

$< S_z> = J_z\,(1 - g_J)$

$<\mu_z> = -\mu_B\, g_J\, J_z$

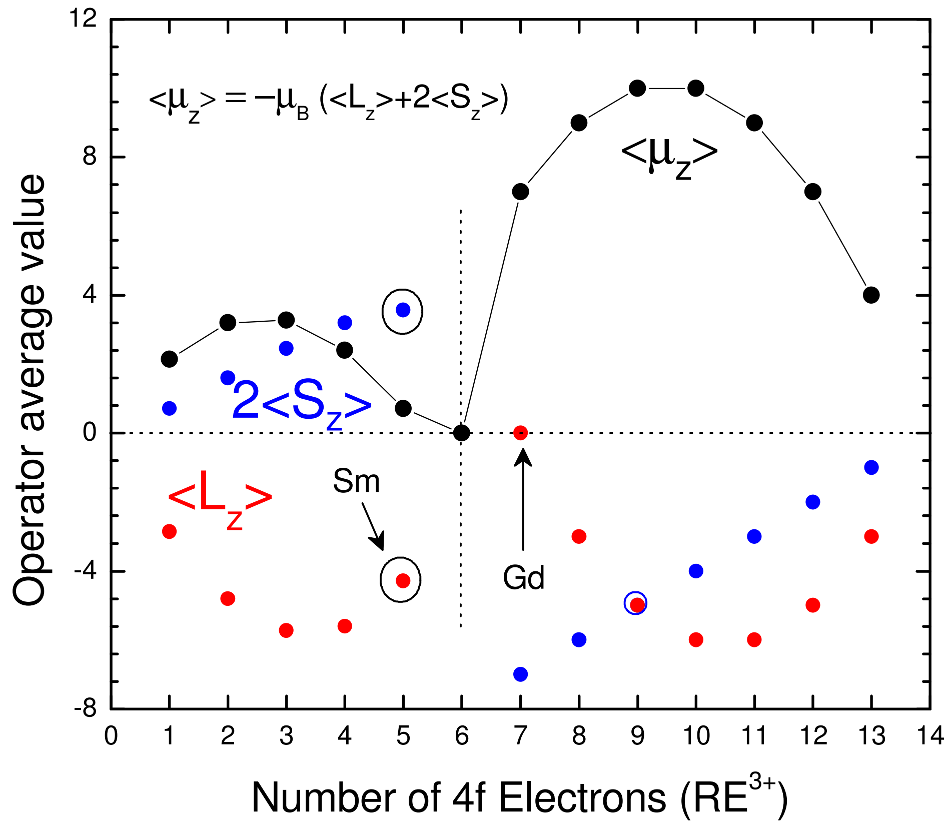
Sm^{3+}: L=5, S=5/2 ; J=5/2

$<L_z> = -30/7,\ \ 2<S_z> = 25/7$

$\mu_z(Sm^{3+}) = 5/7\ \mu_B$
(six fold degenerate)

In cubic systems:
$\mu_z(Sm^{3+})$ = 5/21 (doublet)
$\mu_z(Sm^{3+})$ = 11/21 (quartet)

Magnetic moment originating from $4f^n$ electrons has two parts: orbital and spin



Take any Ferromagnetic series of Rare Earth (R) compounds

For example: R Al_2, R atoms form a diamond lattice

1) $PrAl_2$:

Magnetic moment per Pr ion ~ 2.8 μ_B

Ferromagnetic Curie temperature, T_c ~ 35 K

2) $GdAl_2$:

Magnetic moment per Pr ion ~ 7.7 μ_B

Ferromagnetic Curie temperature, T_c ~ 170 K

Experimental fact (since 1960): $Pr_{0.7}Gd_{0.3}Al_2$ is a pseudo-Ferrimagnet

1960s: Mix two rare earth ferromagnetic compounds

$PrAl_2$: T_c ~ 35 K

μ / f.u. ~ 2.8 μ_B

$GdAl_2$: T_c ~ 170 K

μ / f.u. ~ 7.7 μ_B

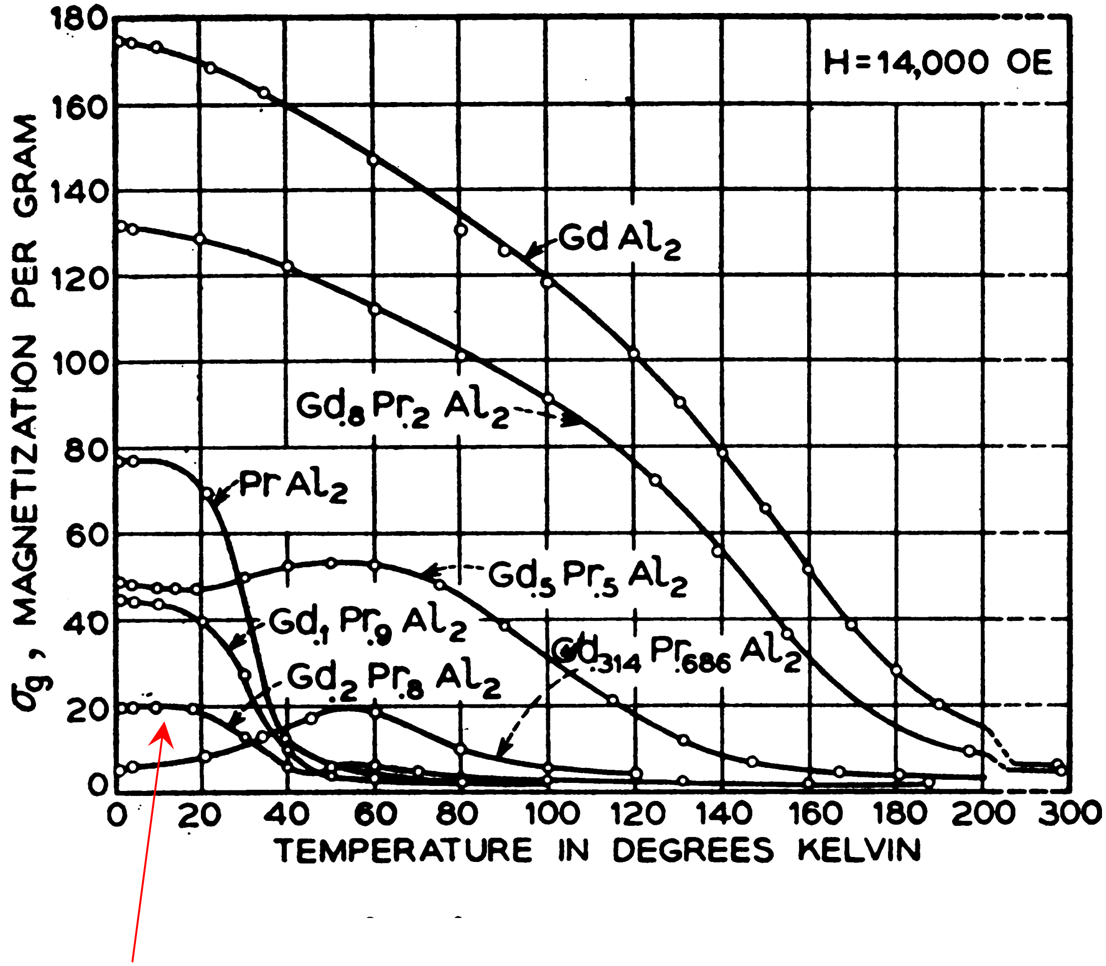
- Zero Magnetization stoichiometry realized at

x* ~ 0.3, *quasi-antiferro response in M vs. T

- **Magnetic compensation** phenomenon at x ~ 0.2

T_c ~ 80 K, $\mathbf{T_{comp}}$**~ 40 K**

Magnetization is minimum at $\mathbf{T_{comp}}$



$Pr_{1-x}Gd_xAl_2$

x = 0.2

H. J. Williams *et al.* J. Phys. Soc. of Japan **17 (Suppl. 1)**, 91 (1962)
Presented at ICM, Tokyo, 1961

Another example of mixing two Rare Earth ferromagnets

Magnetic compensation phenomenon in **polycrystalline $Nd_{1-x}Ho_xAl_2$: x ~ 0.25**

First-half : $NdAl_2$: T_c ~ 70 K

Second-half : $HoAl_2$: T_c ~ 35 K

$\mu_{Nd} / f.u. \sim 2.5\ \mu_B$

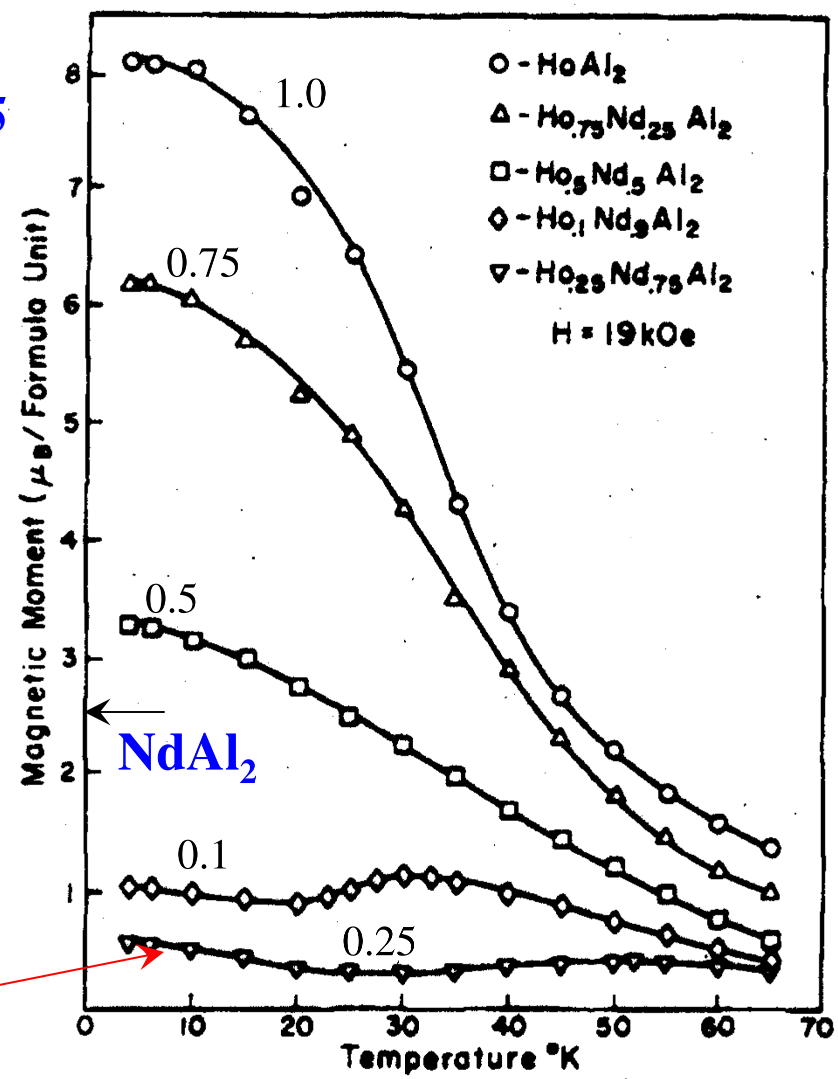
$\mu_{Ho} / f.u. \sim 8.5\ \mu_B$

$Nd_{1-x}Ho_xAl_2$

$x = 0.25$

$T_c \sim 65$ K

$T_{comp} \sim 25$ K



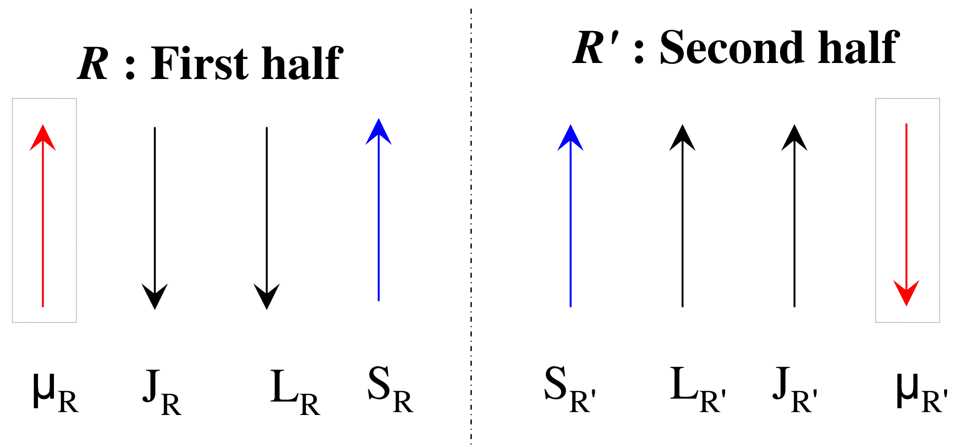
W. M. Swift *et al.*, J. Phys Chem. Solids 29, 2053 (1968)

Understanding the magnetic behavior of the admixed rare earth systems: SPIN FERROMAGNETS

1) Recall: J = L – S, for first half of Rare Earth series and J = L + S, for second half

2) When spins remain parallel in the admixed systems, the magnetic moments are compelled to align antiparallel

$R_{1\text{-}x}R'_xX_n$



Admixed alloys: Spin- ferromagnets (SF)

Zero-Magnetization SF : $(R_{1\text{-}x}R'_xX_n)$: $(1 - x)\,\mu_R \approx x\,\mu_{R'}$

(ZMSF)

X : Non-magnetic element(s)

Hyperfine fields in $Sm_{1-x}Gd_xAl_2$ alloys - **Microscopic evidence** for **ferromagnetic coupling** between rare earth **spins**

J. Appl. Phys. Volume 50, pp. 7501-7503 (1979)

(Cited amongst eleven references by H Adachi and H Ino, Nature 401, 148, 1999)

A. K. Grover, S. K. Malik and R. Vijayaraghavan

Tata Institute of Fundamental Research, Bombay 400 005, India

K. Shimizu

Faculty of Education, Toyama University, Toyama, Japan

Abstract : *The results of hyperfine field studies on $Sm_{1-x}Gd_xAl_2$ alloys for $0<x<0.5$ are reported. The hyperfine field at Al, H(Al), has been measured to be +32.5 kOe in $SmAl_2$ and –47 kOe in $GdAl_2$. In $Sm_{1-x}Gd_xAl_2$ alloys, we find that the* ***magnitude of H(Al) increases with increasing x and further H(Al) becomes negative even with small replacement of Sm by Gd****. H(Al) in these compounds is proportional to the average value of spin per rare earth ion. The observed behaviour can be understood in terms of a ferromagnetic coupling between the spins of Sm and Gd.*

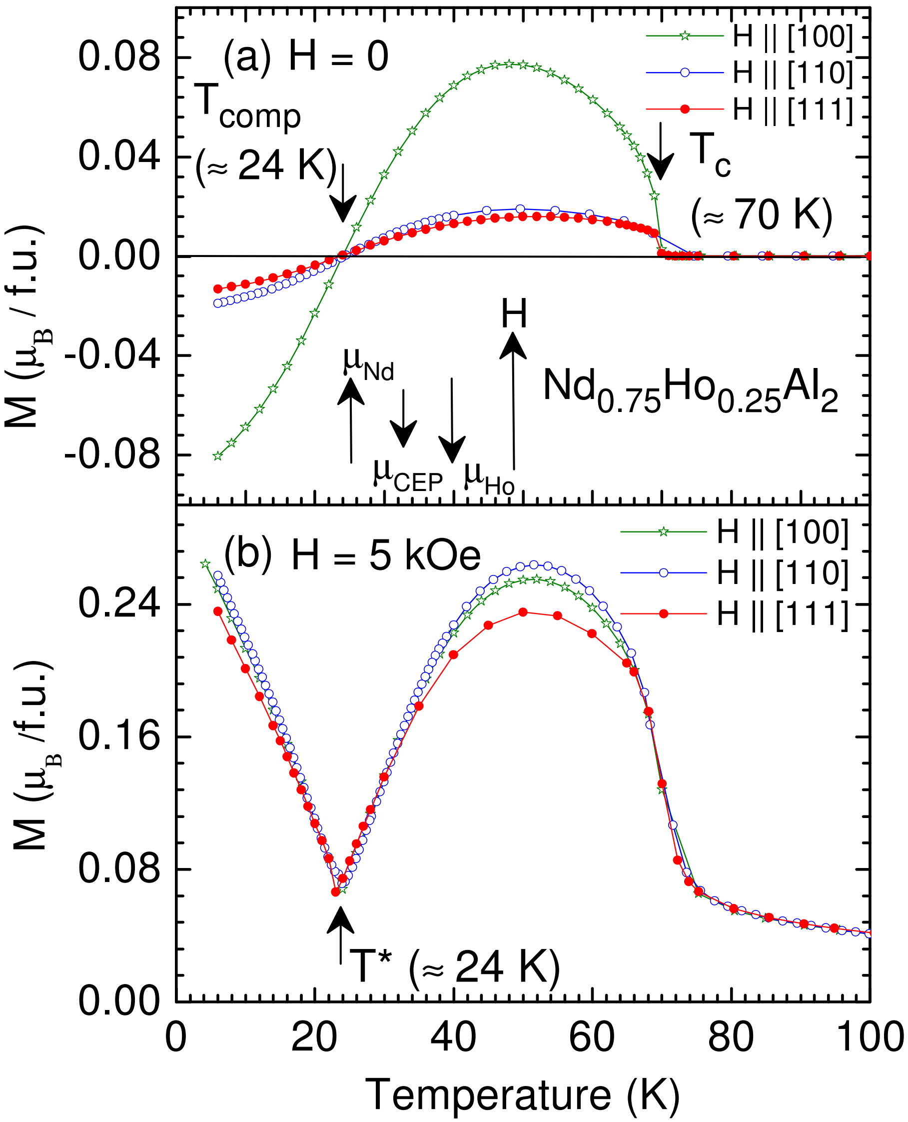
10% replacement of Sm by Gd changed the sign of Hyperfine Field at Al nuclei, implying that sample magnetization got dictated by Gd ions.

Our strategy in recent years

 # Admix Ferromagnetic compounds belonging to two different halves of Rare Earth series

 # Grow single crystals of them to re-explore physics issues, *in systems which were first studied in 1960s.*

STUDIES AT TIFR SINCE 2007

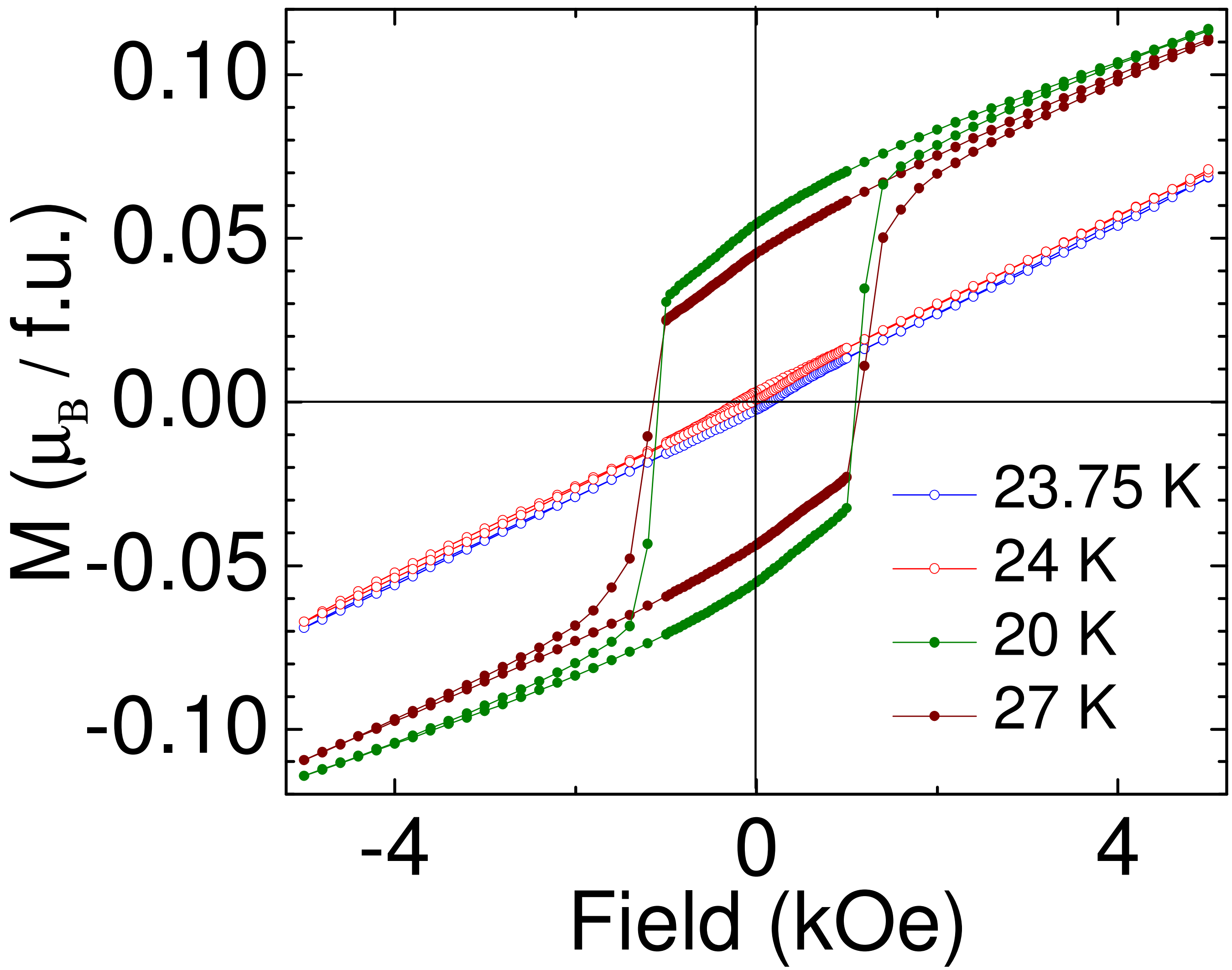


EXPERIMENTS IN SINGLE CRYSTALS OF $Nd_{0.75}Ho_{0.25}Al_2$

Magnetic compensation phenomenon established in all orientations of field in a single crystal of $Nd_{0.75}Ho_{0.25}Al_2$

Prasanna D. Kulkarni *et al., Euro Physics Letters 86,47003 (2009)*

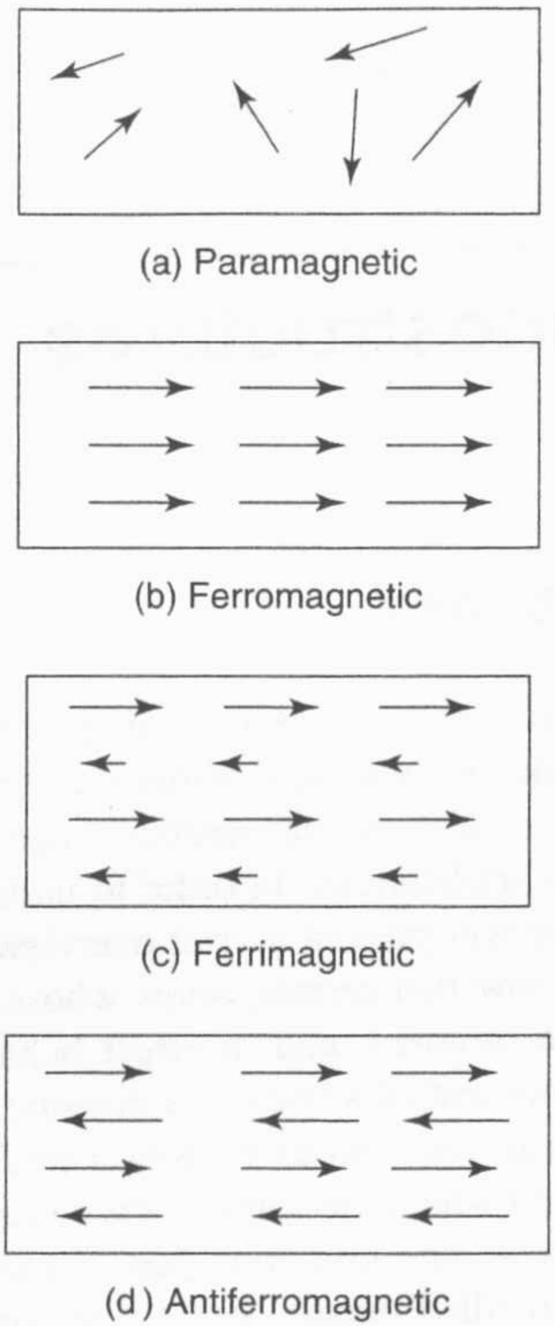
Magnetization Hysteresis Loops : $Nd_{0.75}Ho_{0.25}Al_2$

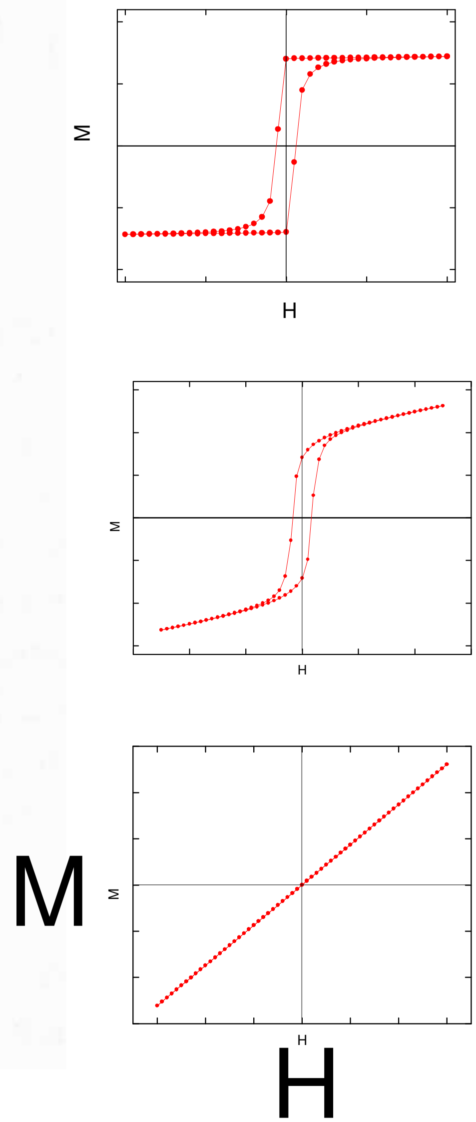


M-H **Response is quasi-antiferromagnetic very close to T_{comp}**

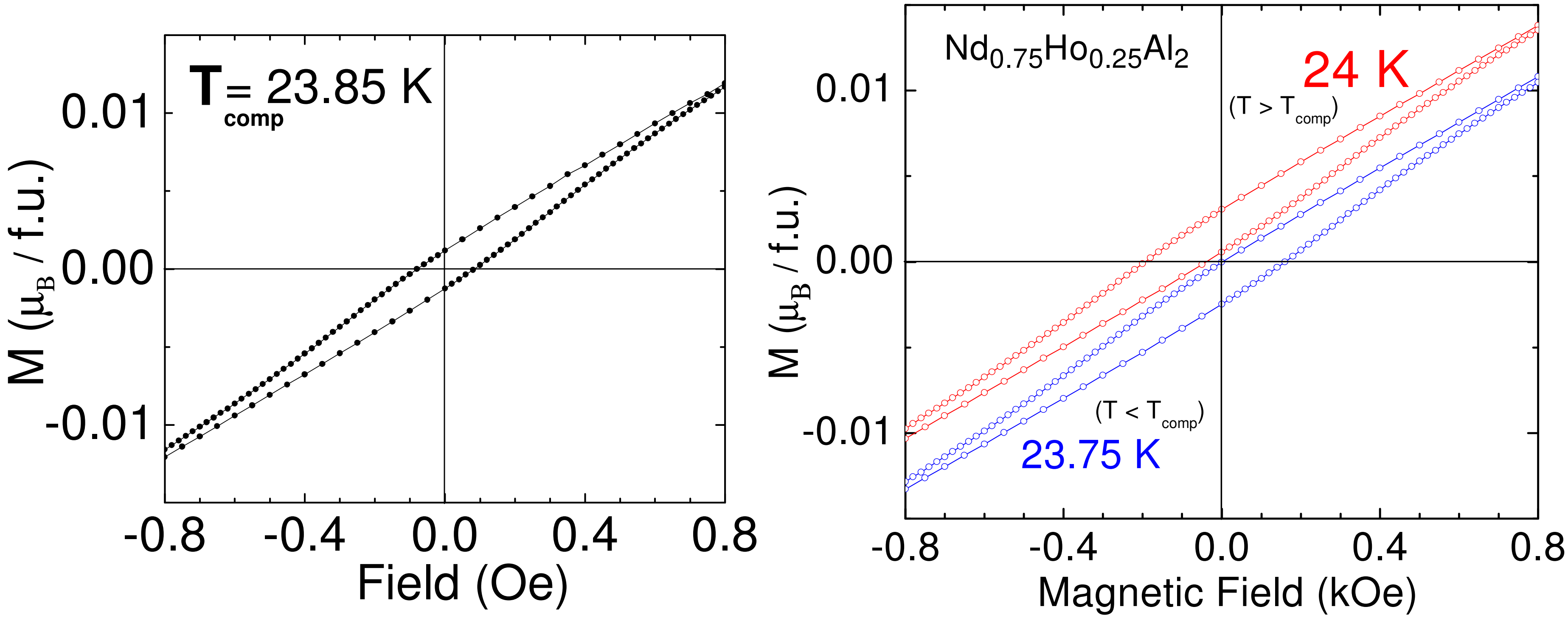
Prasanna D.Kulkarni ***et al., Euro Physics Letters 86,47003 (2009)***

Schematic M-H curves of different arrangements





Magnetization Hysteresis Loops in very close vicinity of T_{comp}



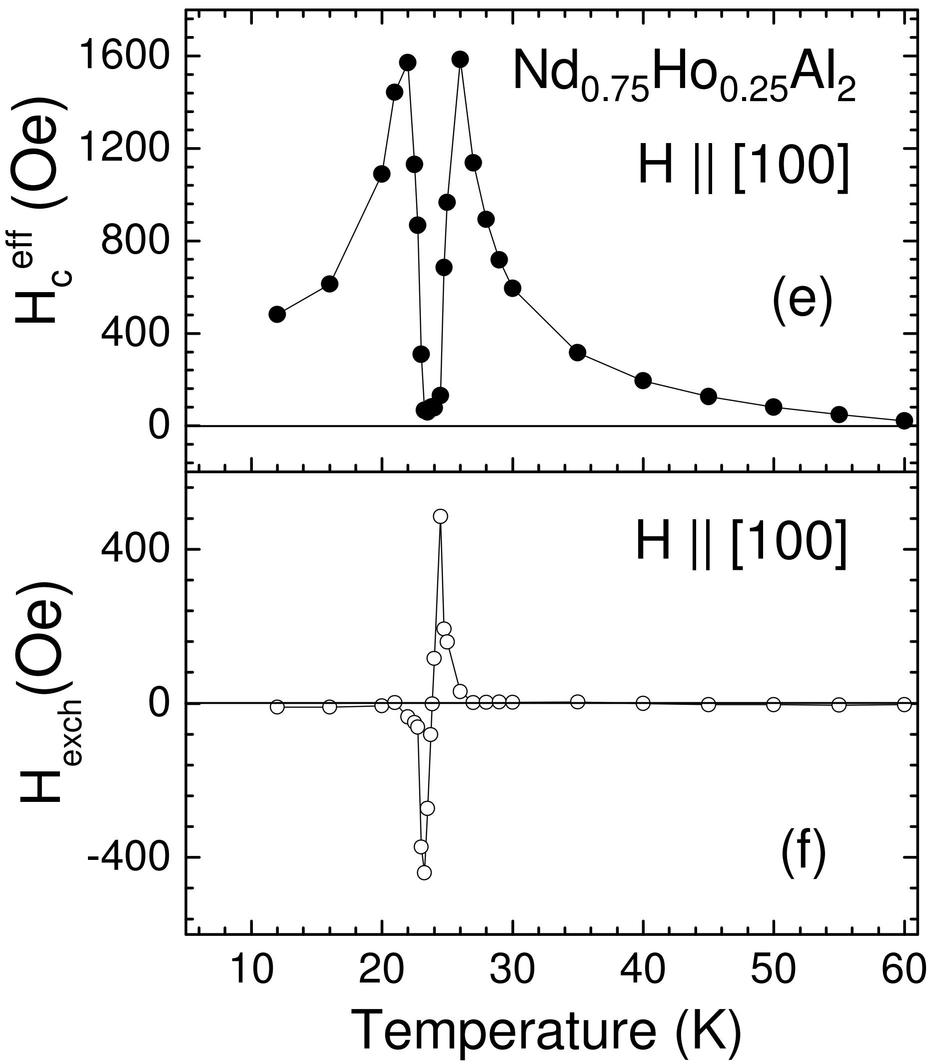
- Note the visible shift in the *M-H* loop at 23.75 K and 24 K and the symmetric *M-H* loop at 23.85 K.
- (Left/Right) Shift in M-H loop is called Exchange Bias Field

Prasanna D. Kulkarni *et al., Europhysics Letters 86,47003 (2009)*

Exchange Bias in $Nd_{0.75}Ho_{0.25}Al_2$

Half-width of the M-H loop

Shift in the M-H loop

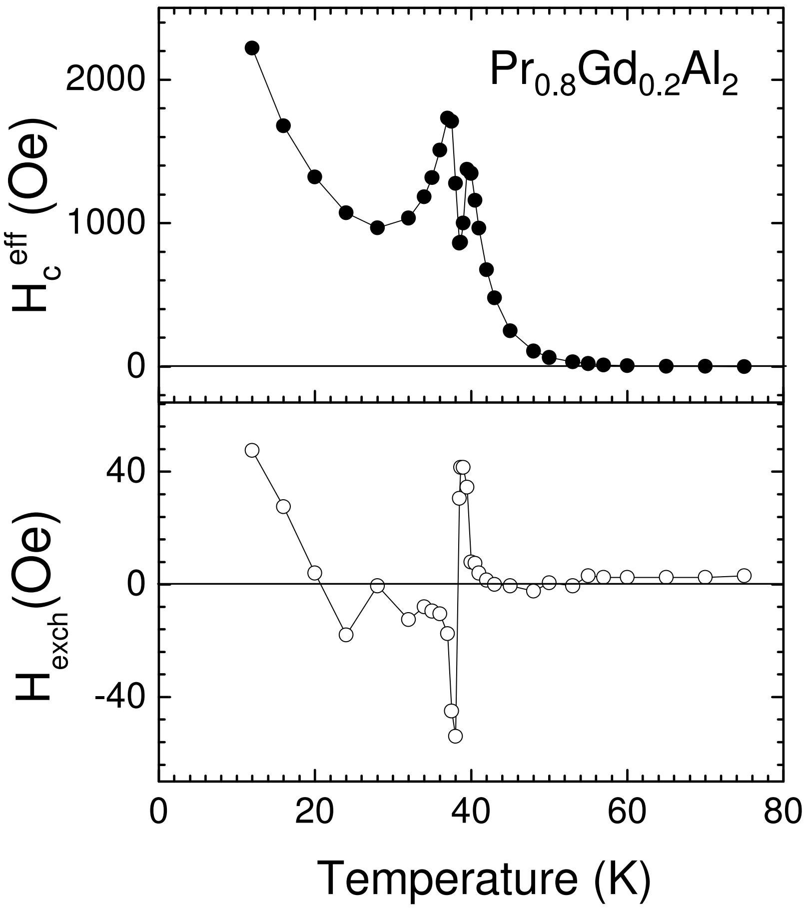


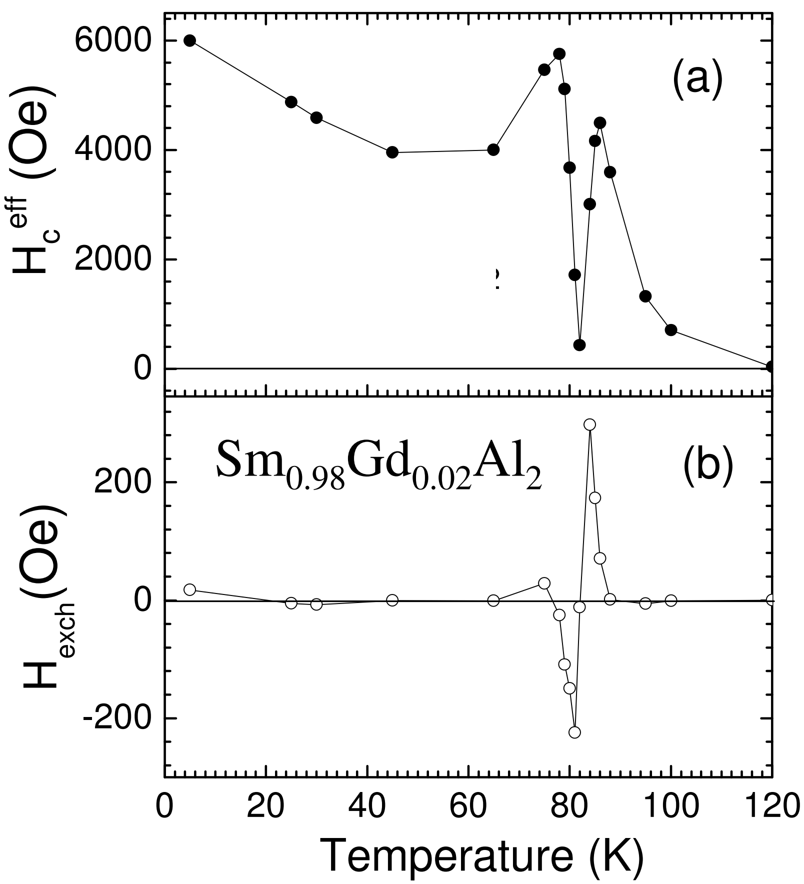
- The notion of exchange bias is observed in the admixed rare earth intermetallics near T_{comp}

P. Kulkarni *et al., Euro Physics Letters 86,47003 (2009)*

P. Kulkarni *et al., IEEE Trans. Magn. 45, 2902 (2009)*

Confirmation of EB and its phase reversal in close vicinity of T_{comp} in $Pr_{0.8}Gd_{0.2}Al_2$ and $Sm_{0.98}Gd_{0.02}Al_2$

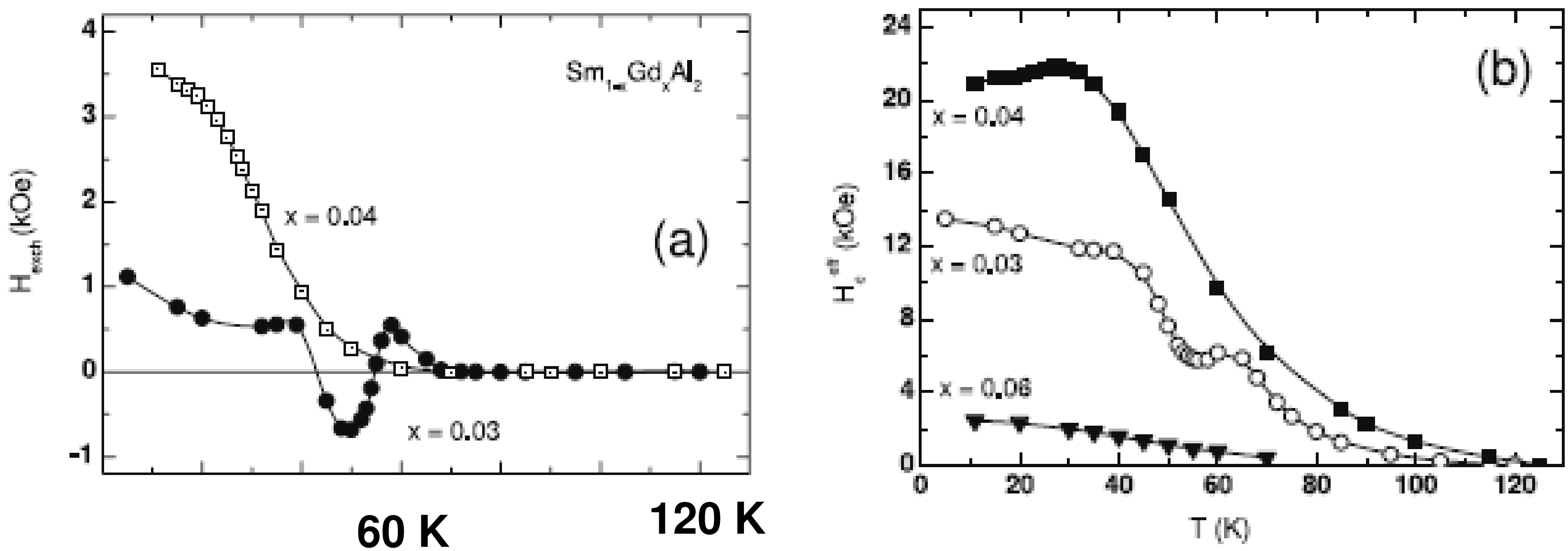




EXCHANGE BIAS FINGERPRINTS COMPENSATED LOCAL MOMENTS COUPLED TO POLARIZED BAND

P. D. Kulkarni *et al., IEEE Trans. Magn. 45, 2902 (2009)*

Tuning of exchange bias and coercive field in $Sm_{1-x}Gd_xAl_2$



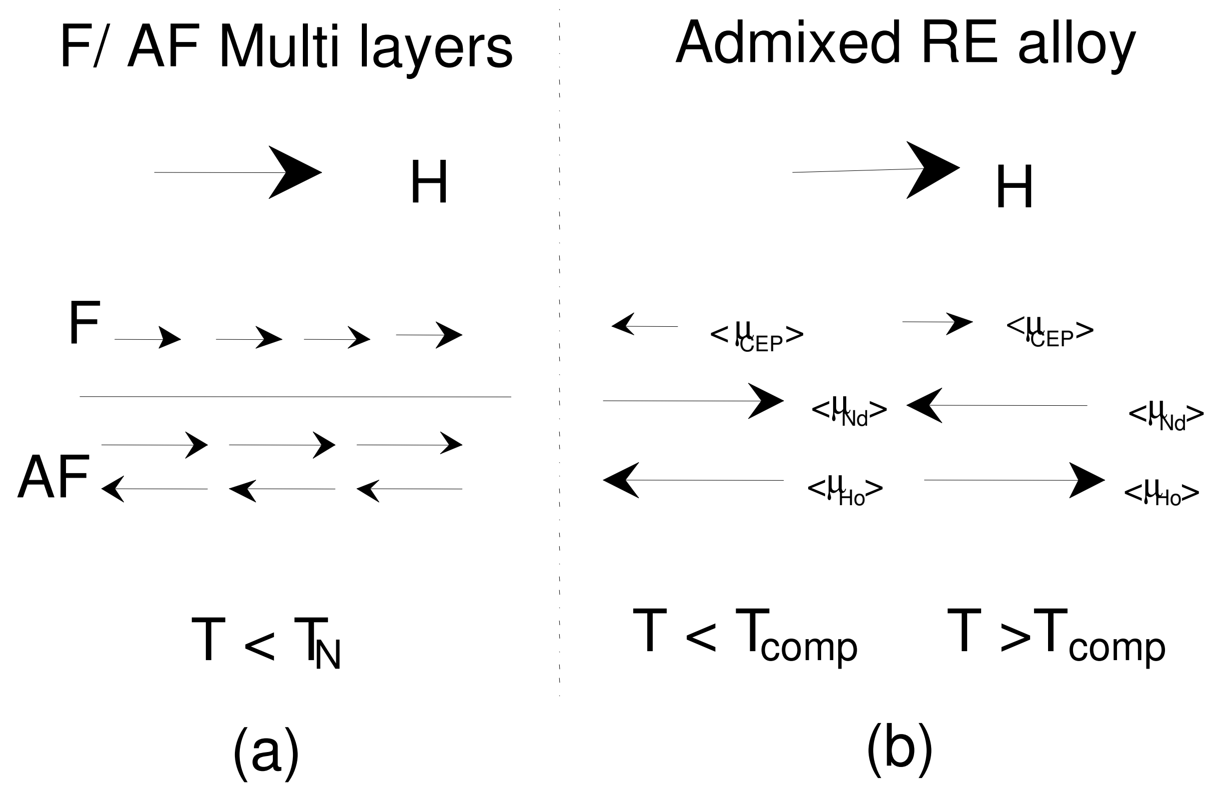
Exchange bias enhances from x = 0.1 to 0.4,

therafter it collapses at x = 0.06 and H_c^{eff} reduces.

At x= 0.06, Gd magnetization dominates over that from Sm.

S. Venkatesh *et al*, J. Phys.: Condens. Matter 22 (2010) 496002

Comparison between the pinned F/AF bi-layer structure and the admixed RE alloys



- The soft conduction electron contribution could assume a role of the soft ferromagnetic layer near the magnetic compensation region.
- However, both, the polarized electrons and the compensated local moments reverse their orientations across the compensation temperature.

P. D. Kulkarni *et al., Euro Physics Letters 86,47003*

Separation of the 4*f*-spin, 4*f*-orbital, and conduction-electron magnetization … for ferromagnetic Sm intermetallics

(H. Adachi *et al* ., *Phys. Rev. B*, 59, 11445 (1999))

Magnetization signal in a Sm system	=	Orbital contribution from Sm^{3+}	+	Spin contribution from Sm^{3+}	+	Cond. Electron Polarization contribution (CEP)
		+		−		−

Usually : Orbital contribution > contributions from spins

" **Orbital Surplus**" system

\# In the case of Sm ions, higher J-states easily admix into the ground J-state in presence of strong Exchange field and crystalline electric field .

\# 'Spin' and 'Orbital' parts of the Samarium moments typically start to follow **different** temperature dependences.

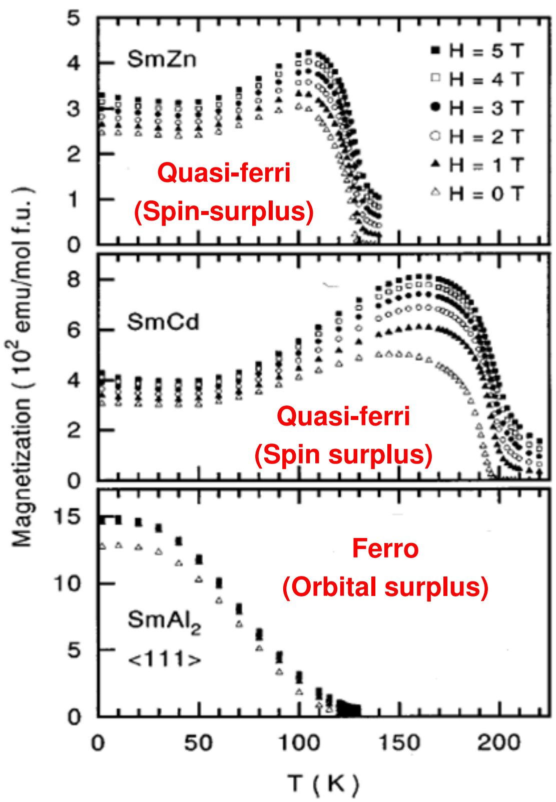
Occassionally : Contributions from spins > Orbital contribution

" **Spin Surplus**" system

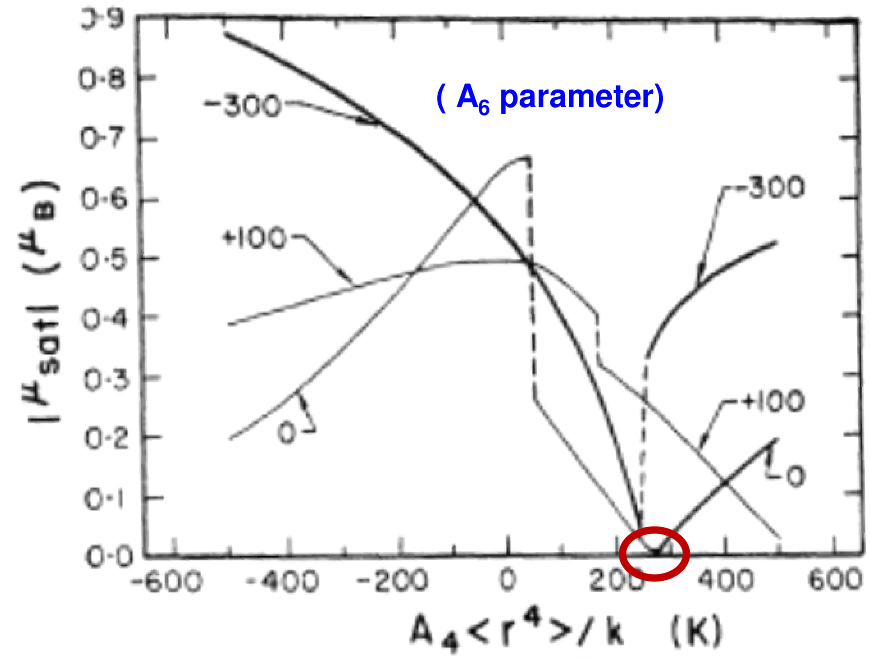
Calculated T = 0 values

	$M_{tot.}$ (μ_B)	m_{4f} (μ_B)	$-L_z$	$-2S_z$	m_{cond} (μ_B)
SmZn	.05	-0.36	-4.01	3.65	**0.41**
SmCd	.06	-0.34	-4.19	3.85	**0.40**
$SmAl_2$	0.26	**0.50**	4.37	-3.87	-0.24

H. Adachi *et al.*, *Phys. Rev. B* 59, 11445 ,1999



Calculations by S K Malik at TIFR (1973-74)



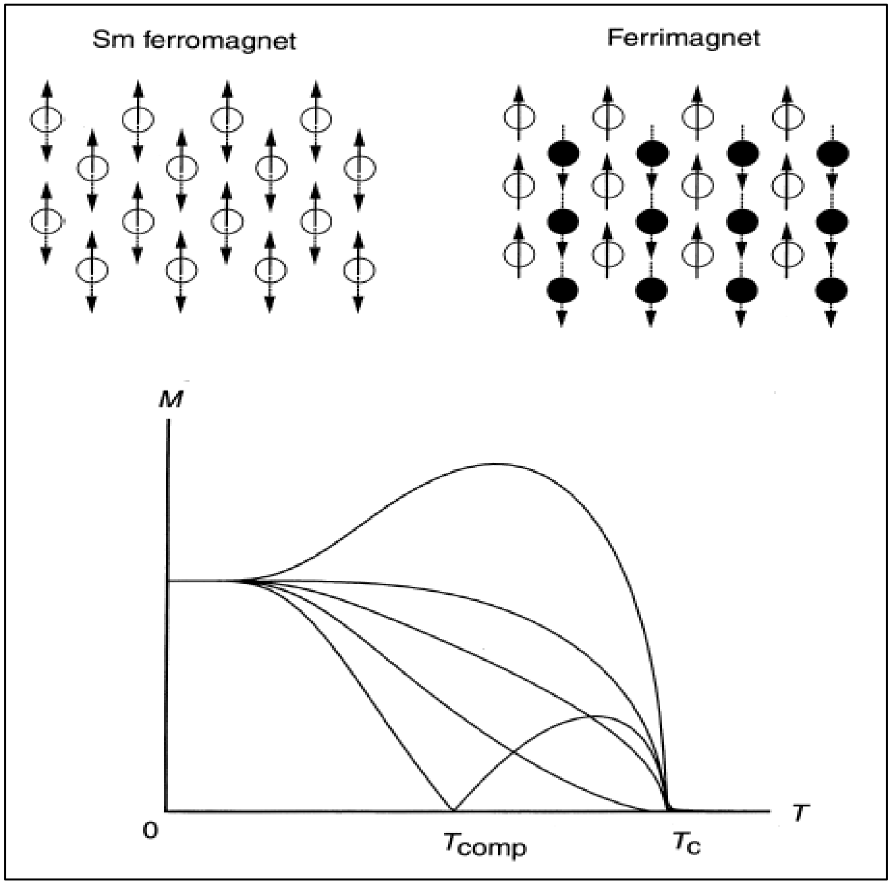
Pramana 3,122(1974)

S K Malik's zero-local moment implies an antiferromagnet at every ionic site, exchange coupled to conduction electrons to yield high CEP.

See, also, K H J Buschow *et al.* Phys. Rev. B 8, 5134 (1973)

“A ferromagnet having no magnetic moment”
H. Adachi, H. Ino *Nature*, **401**, 148 (1999)

S K Malik’s Local AF



Examples of Self-Compensation in pristine Sm based Ferro-magnets

M-T Curves in 10 kOe in Sm-Ferromagnets

Orbital-surplus

$SmCu_4Pd$

SmPtZn

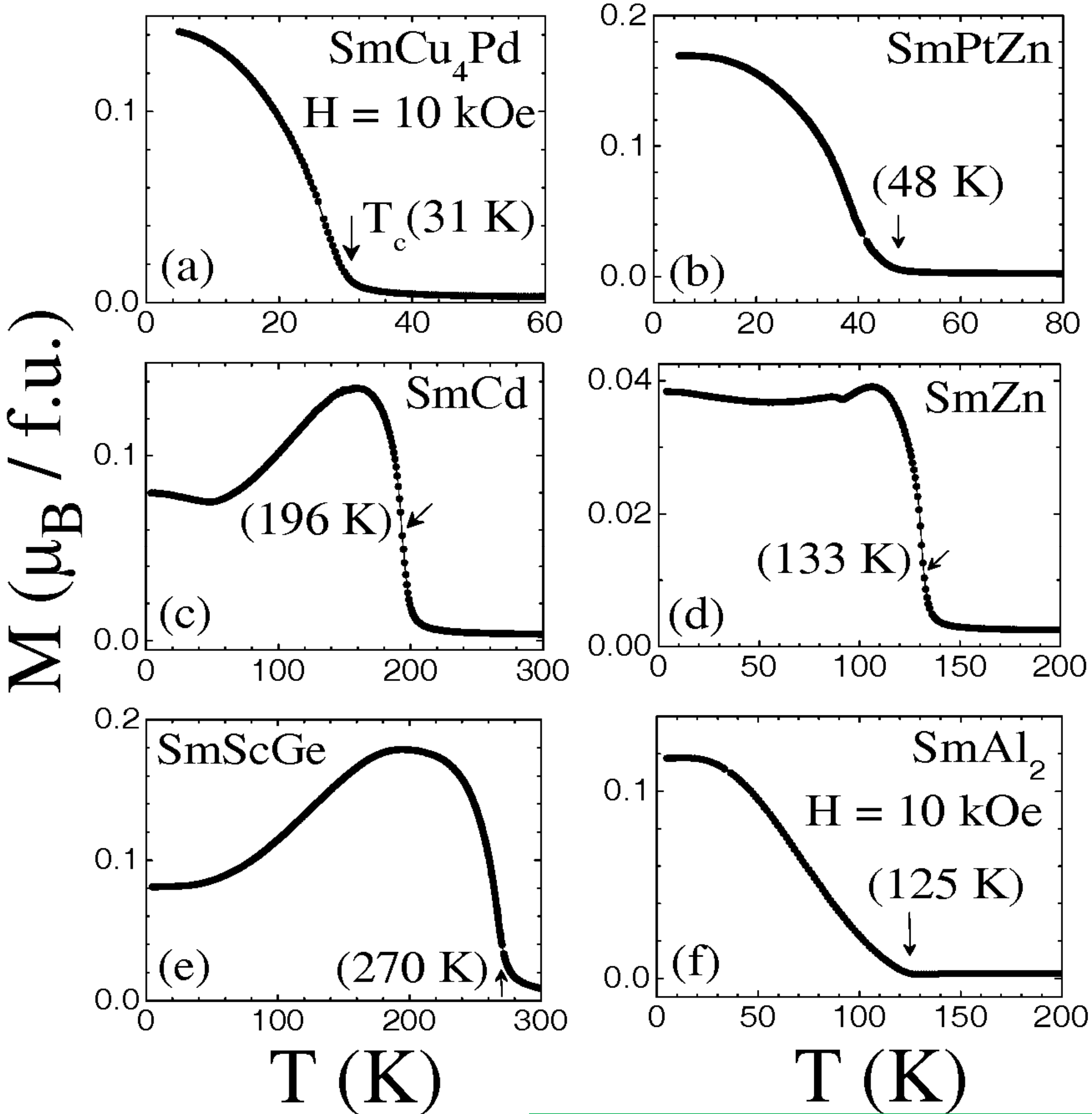
$SmAl_2$

Spin-surplus

SmCd

SmScGe

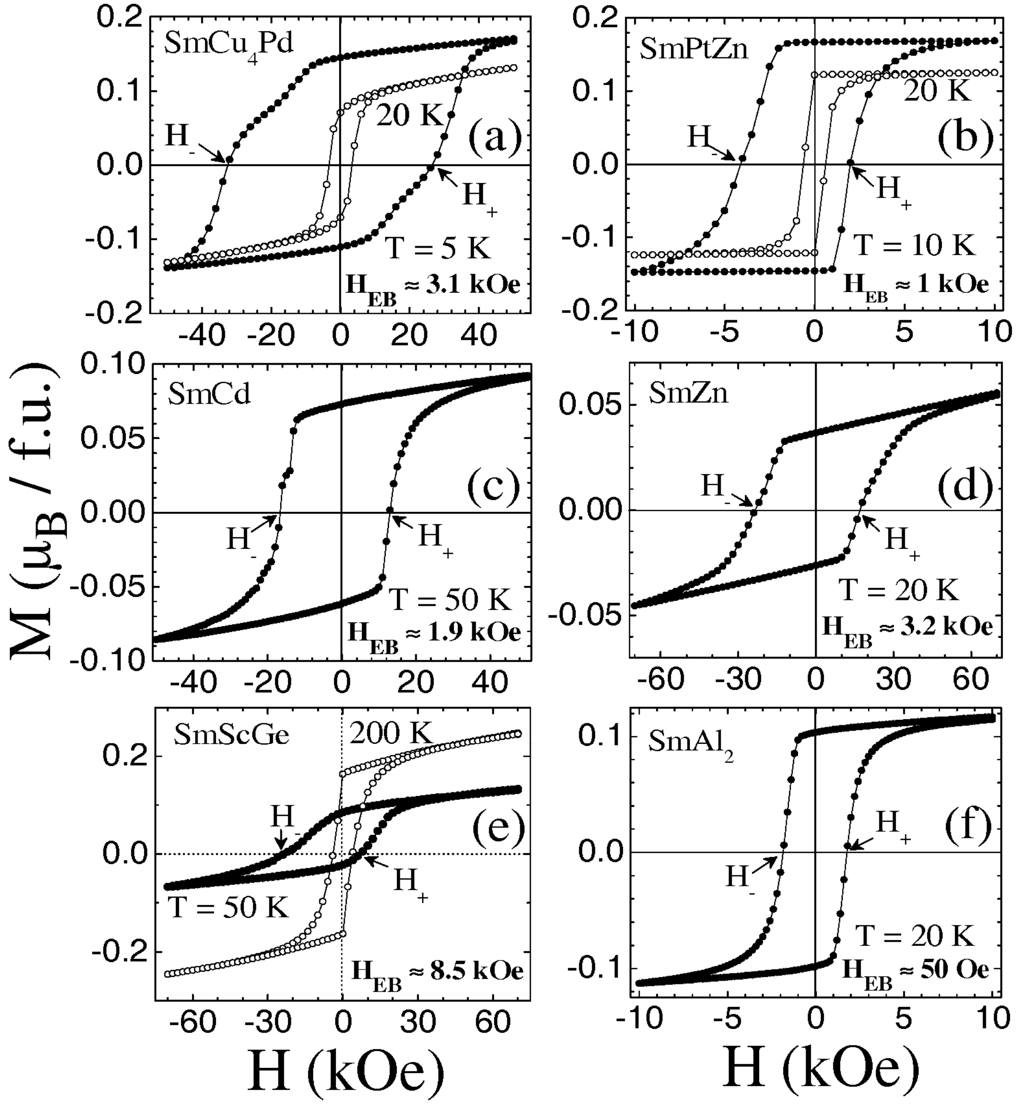
SmZn



P. D. Kulkarni *et al.*,
Phys. Rev. B 82,144411(2010)

Shifted M-H loops in Sm-Ferromagnets

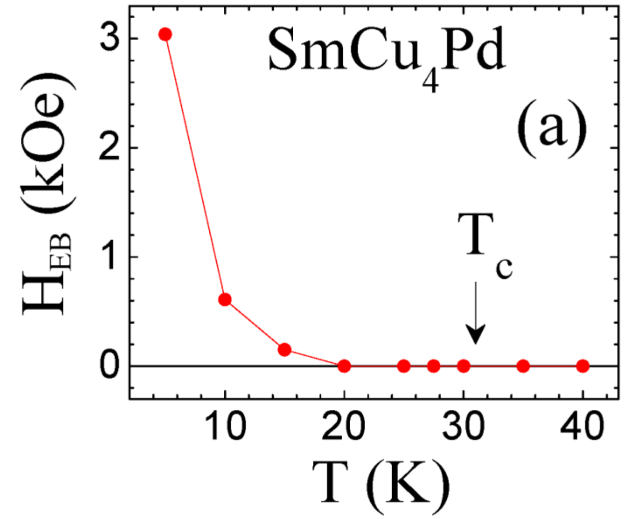
Fingerprint of self-magnetic compensation

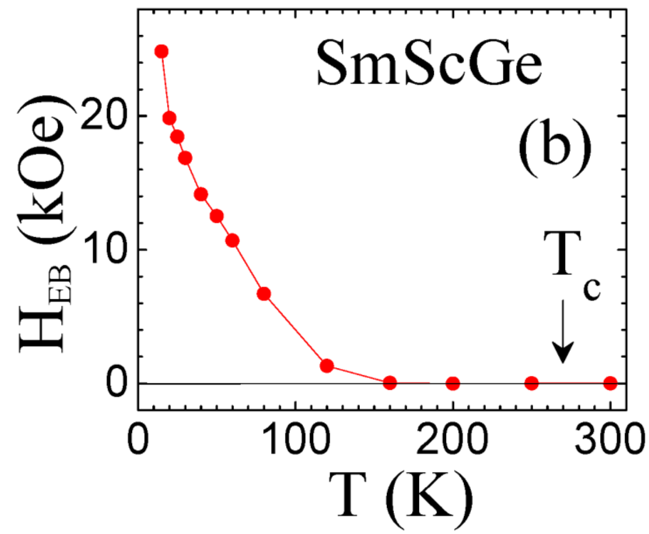


$$H_{EB} = -(H_- + H_+)/2$$

P. D. Kulkarni *et al.*, PRB 82, 144411 (2010)

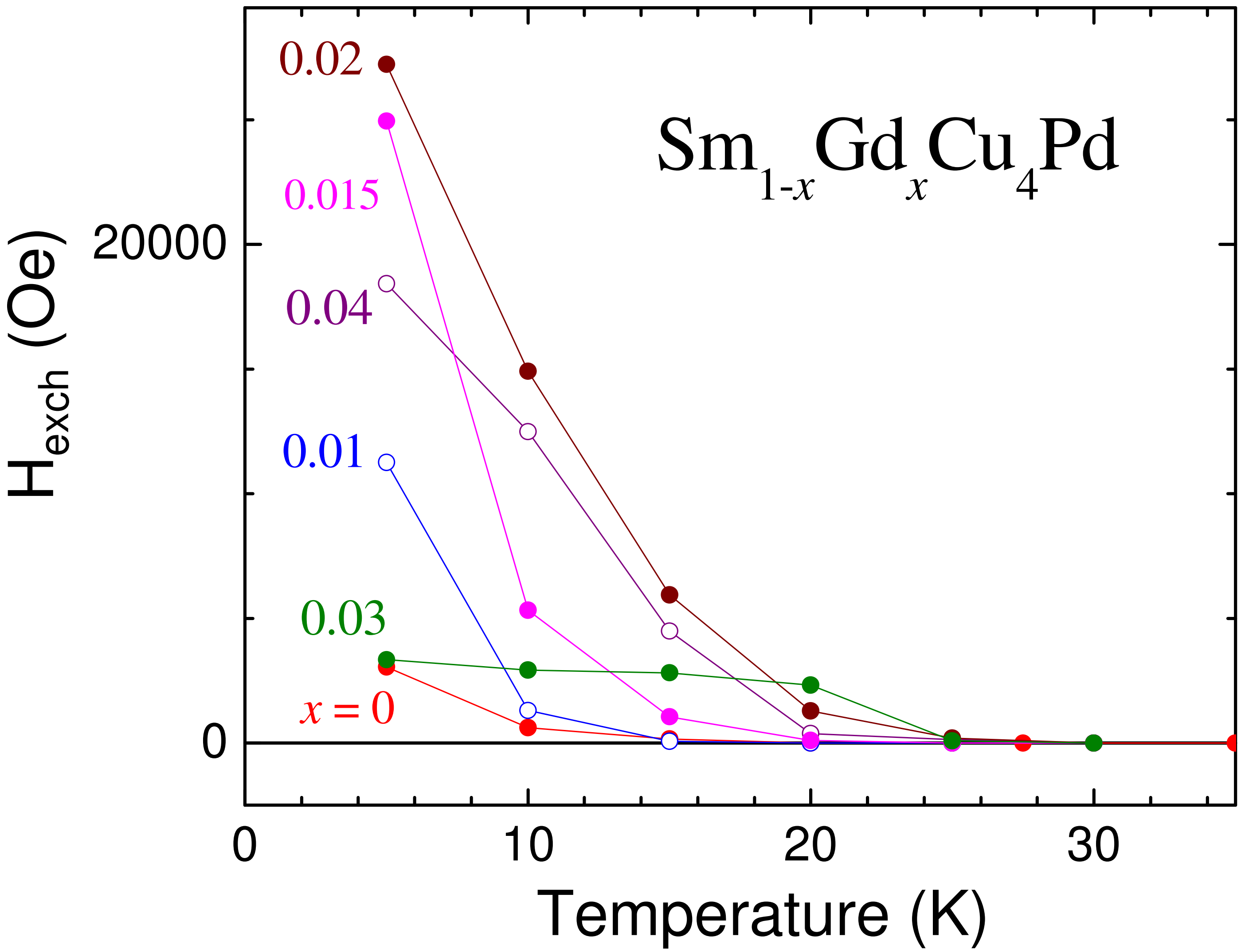
Temperature dependences of exchange bias in two Sm-ferromagnets





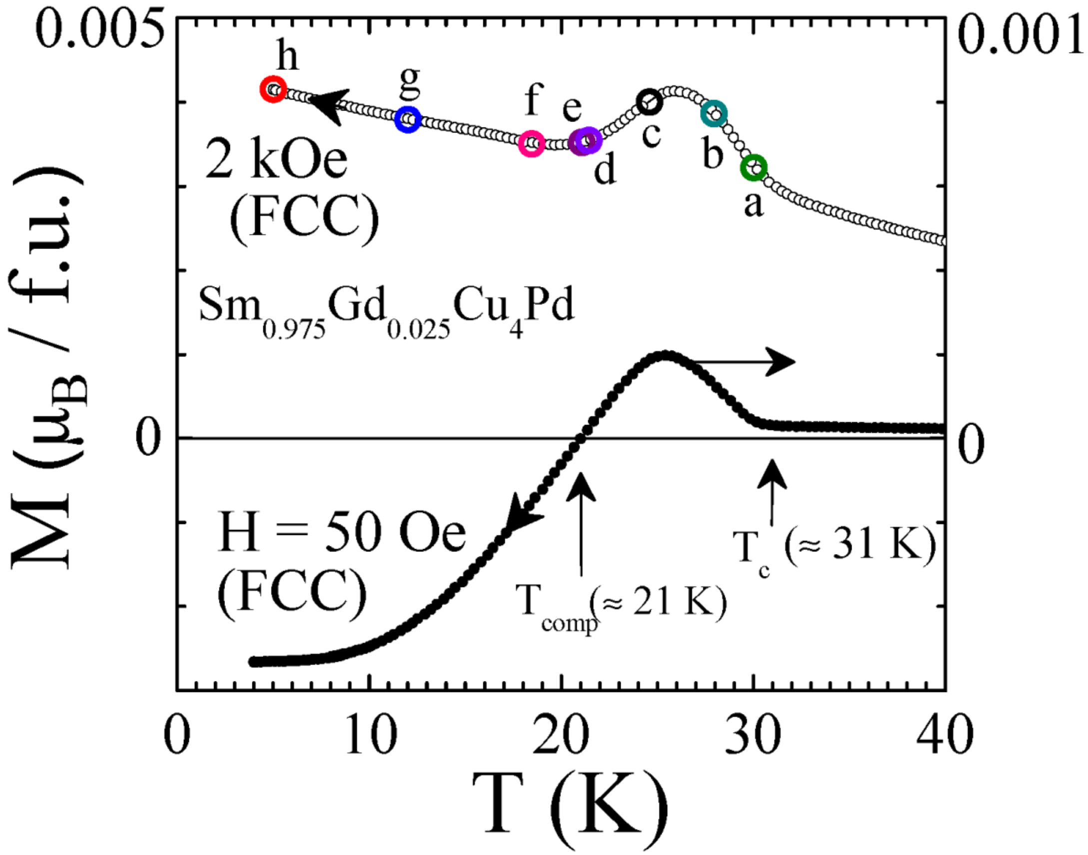
P. D. Kulkarni *et al.*, PRB 82, 144410, 2010

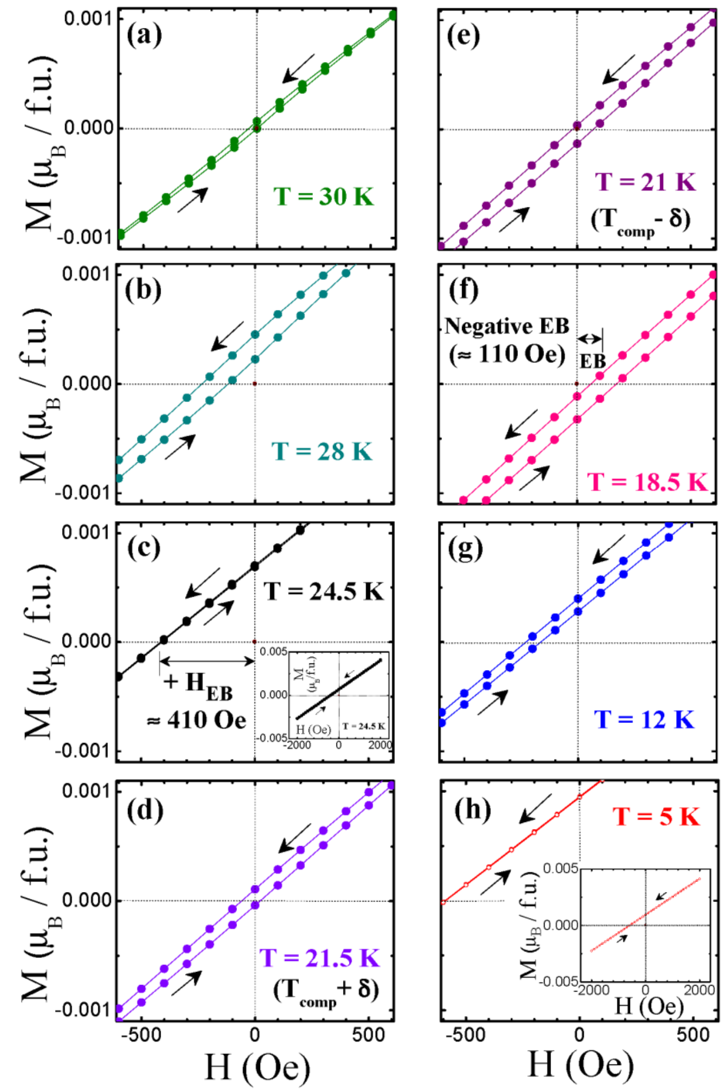
Tuning of the exchange bias field in $Sm_{1-x}Gd_xCu_4Pd$



P D Kulkarni *et al.*, unpublished

Compensated *pseudo–Ferrimagnet:* $Sm_{0.975}Gd_{0.025}Cu_4Pd$



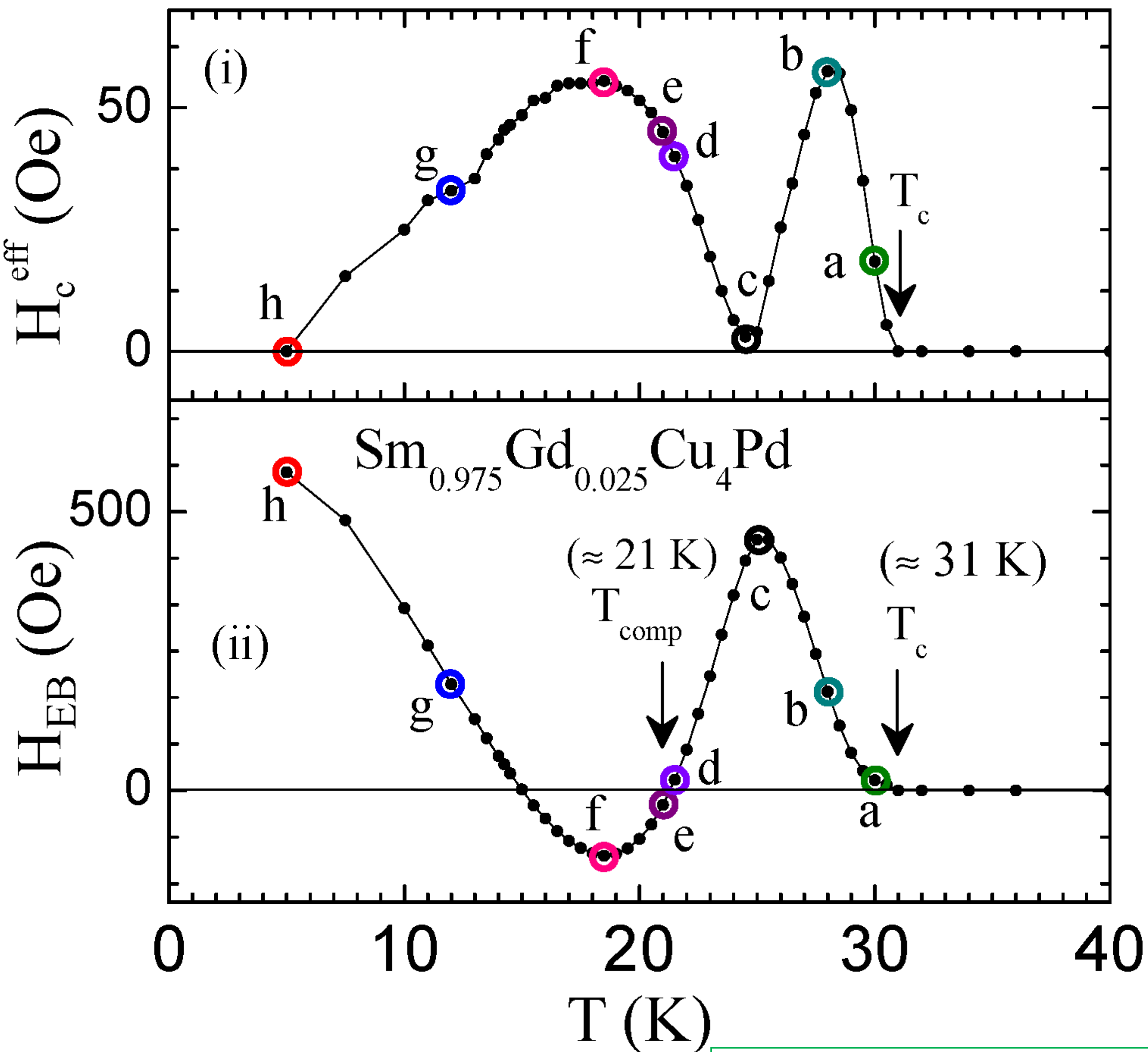


SHIFTED ANTI-FERROMAGNETIC LIKE M-H RESPONSE AT T << T_{comp}

(see curve at 5 K in panel (h))

P. D. Kulkarni *et al.*, PRB 82, 144410, 2010

EXCHANGE BIAS ITS PHASE REVERSAL IN Gd DOPED $SmCu_4Pd$



P. D. Kulkarni *et al*., PRB 82, 144410, 2010

Explorations needed to find RE based materials to perform the function of a pinned ferromagnetic layer in Spin Valves at Room temperature

- **They should be magnetically ordered at room temperature**
- **Exchange bias should exist over large temperature range including RT**

New Samarium and Neodymium based admixed ferromagnets with near-zero net magnetization and tunable exchange bias field. *(near ambient temperatures)*

P. D. Kulkarni *et al*., J. Phys. D: Appl. Phys. 42 (2009) 082001 (*Fast Track Publication*)

Solid State Communications 151 (2011) 985–987

Published May 13, 2011

Contents lists available at ScienceDirect

Solid State Communications

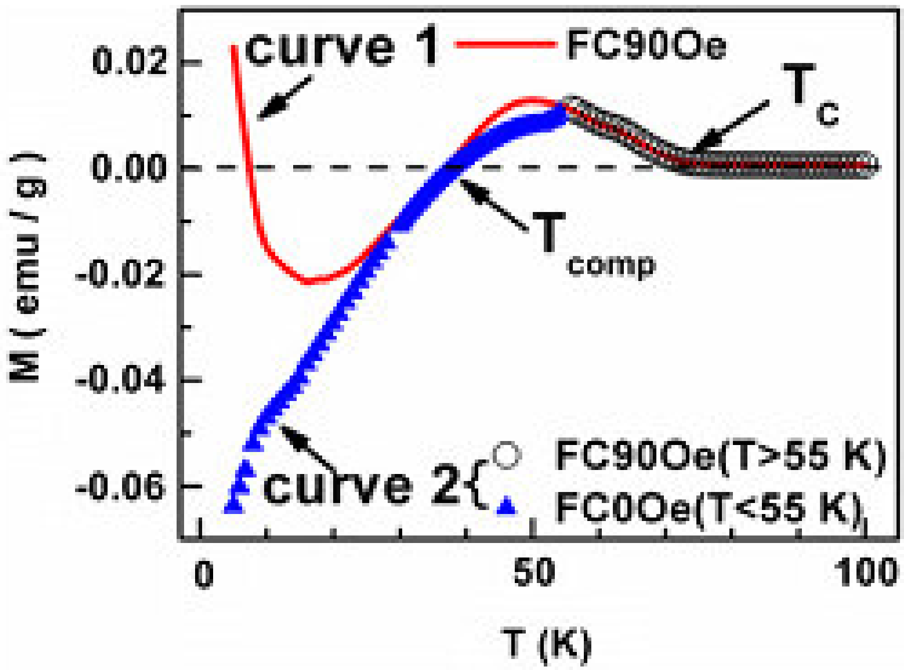
journal homepage: www.elsevier.com/locate/ssc

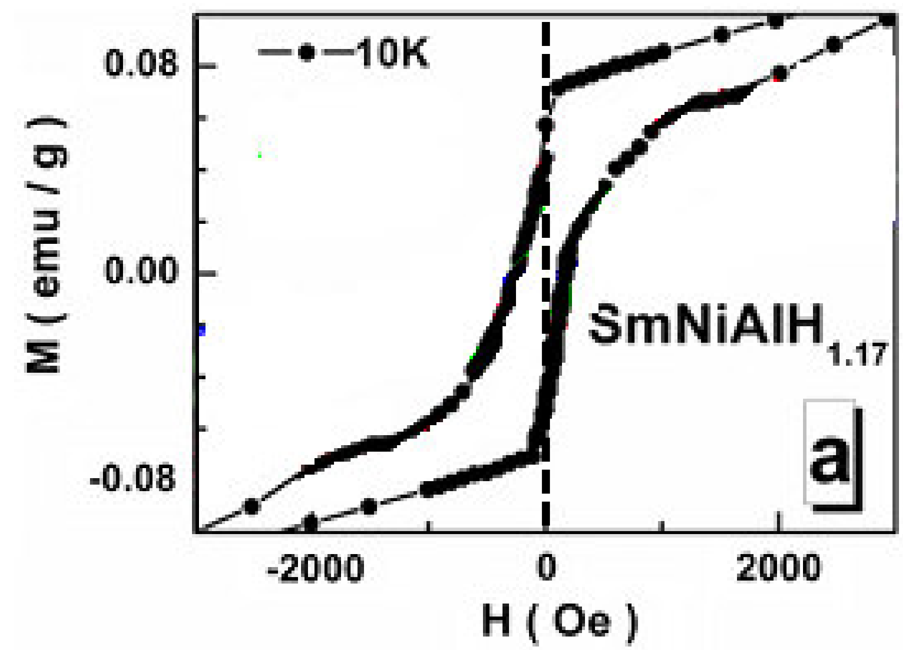
Zero-magnetization ferromagnet induced by hydrogenation

Lin He *

FIRST EXAMPLE OF ZERO CROSSOVER

IN M vs. T CURVE IN A PRISTINE Sm FERROMAGNET





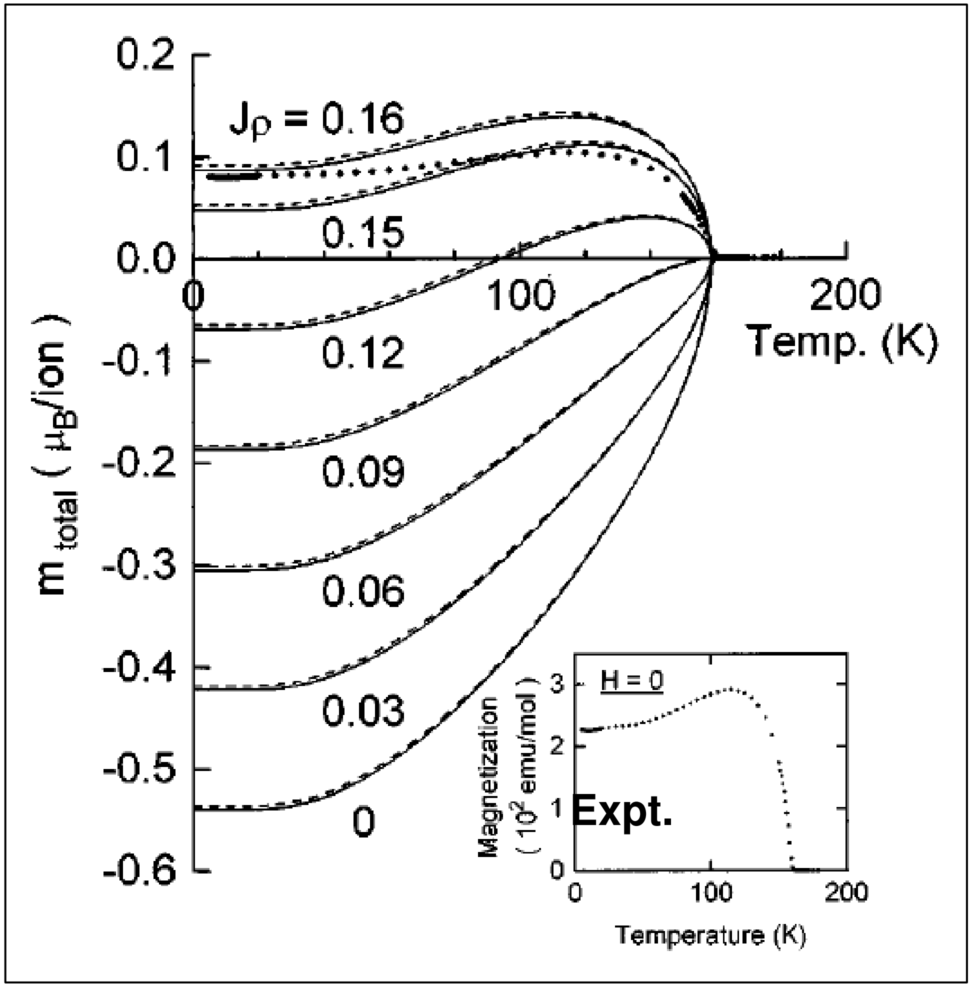
Lin He's manuscipt from Peking University,

Received , accepted and published in three weeks

Mom. /Sm ion ~0.005 μ_B

Calculations for Pure Sm metal

Zero crossover possible in Pristine Sm Ferromagnets



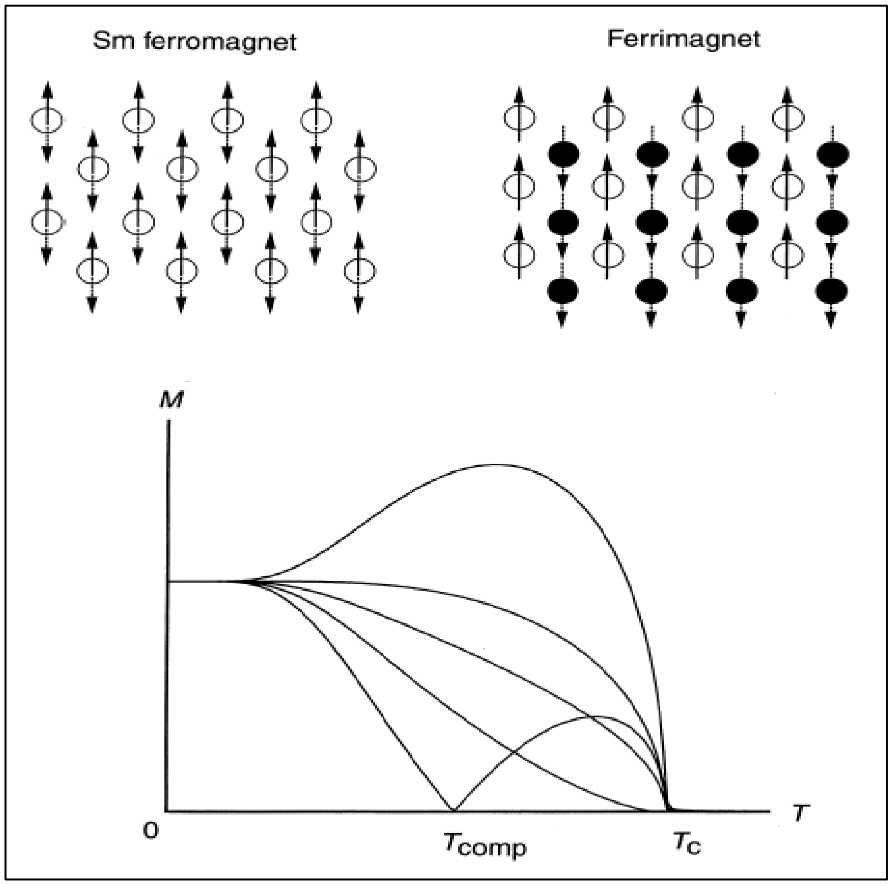
$$\mathcal{H} = \lambda \mathbf{L}\cdot\mathbf{S} + \mathcal{H}_{\text{cryst}} + \mu_B \mathbf{H}\cdot(\mathbf{L}+2\mathbf{S}) + 2\mu_B J\rho \mathbf{H}\cdot\mathbf{S} - 2J_{ff}\langle\mathbf{S}\rangle\cdot\mathbf{S},$$

H. Adachi *et al*, *Phys. Rev. B* **56,** 349 (1997)

Paper included in their patent

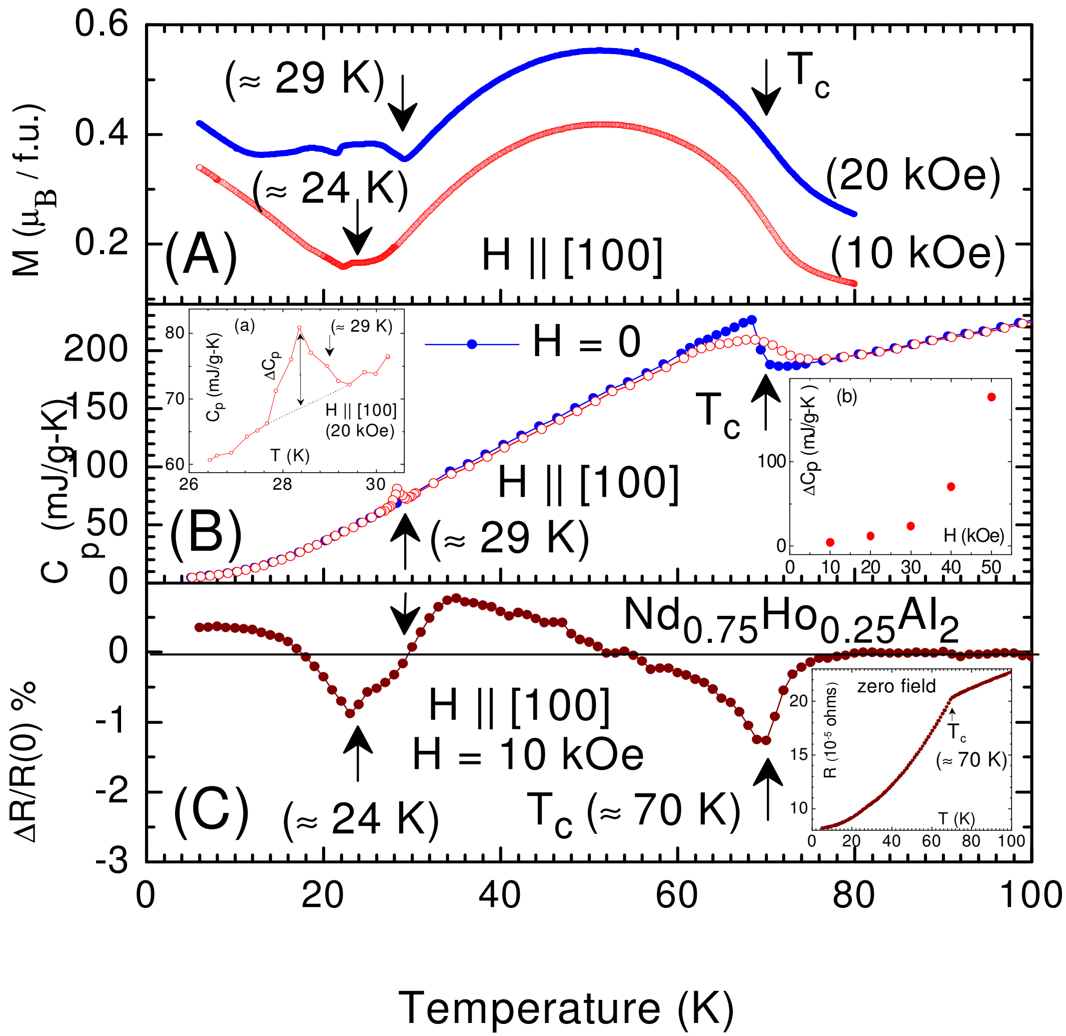
"A ferromagnet having no magnetic moment"
H. Adachi, H. Ino *Nature*, **401**, 148 (1999)

S K Malik's Local AF



More interesting Physics results
in Single crystal of $Nd_{0.75}Ho_{0.25}Al_2$

Magnetic compensation in a single crystal of $Nd_{0.75}Ho_{0.25}Al_2$

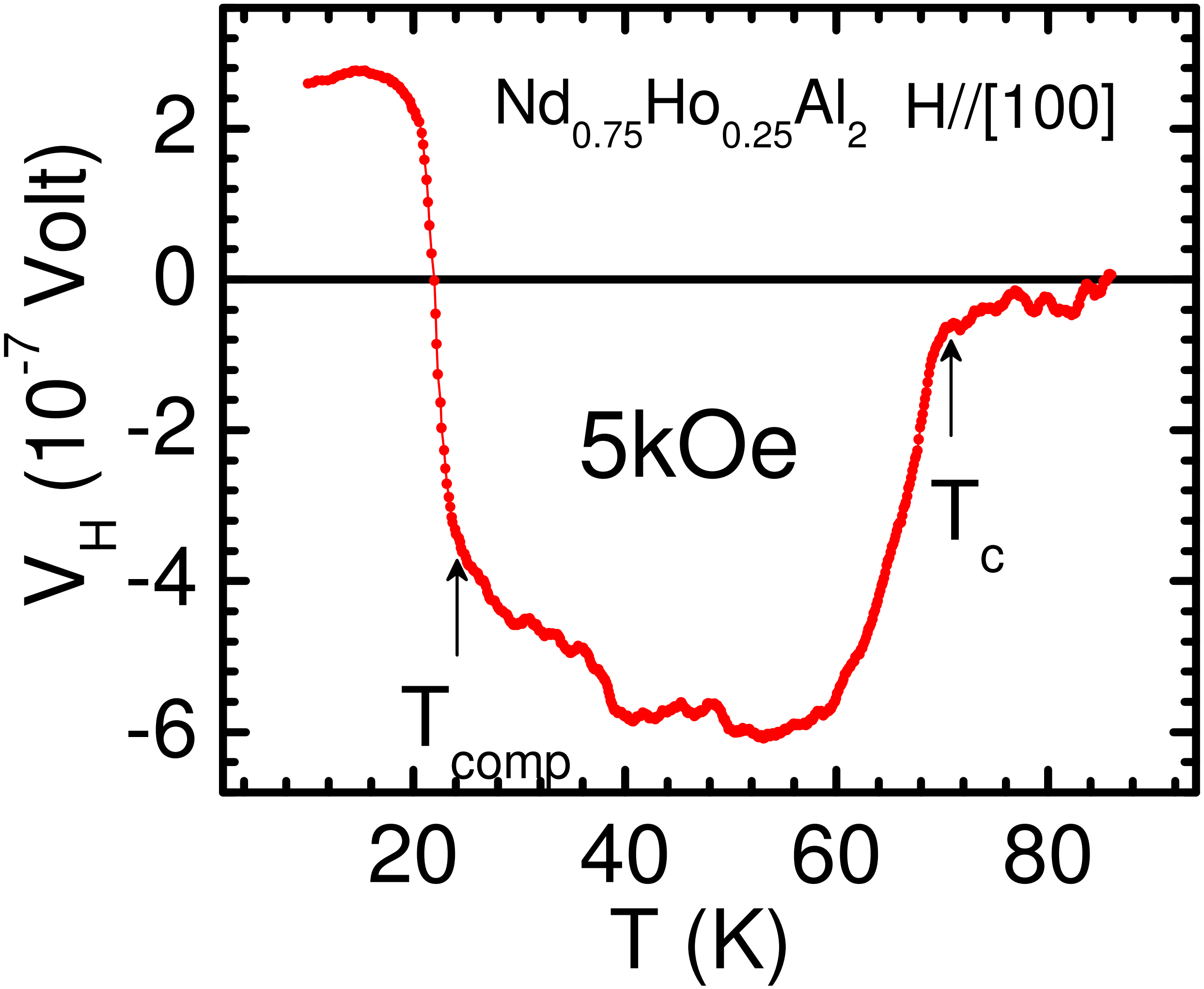


➢ **Field induced reversal in magnetic moments of Ho/Nd imprints as a peak in the specific heat data**

➢ **Oscillatory response in the magneto-resistance**

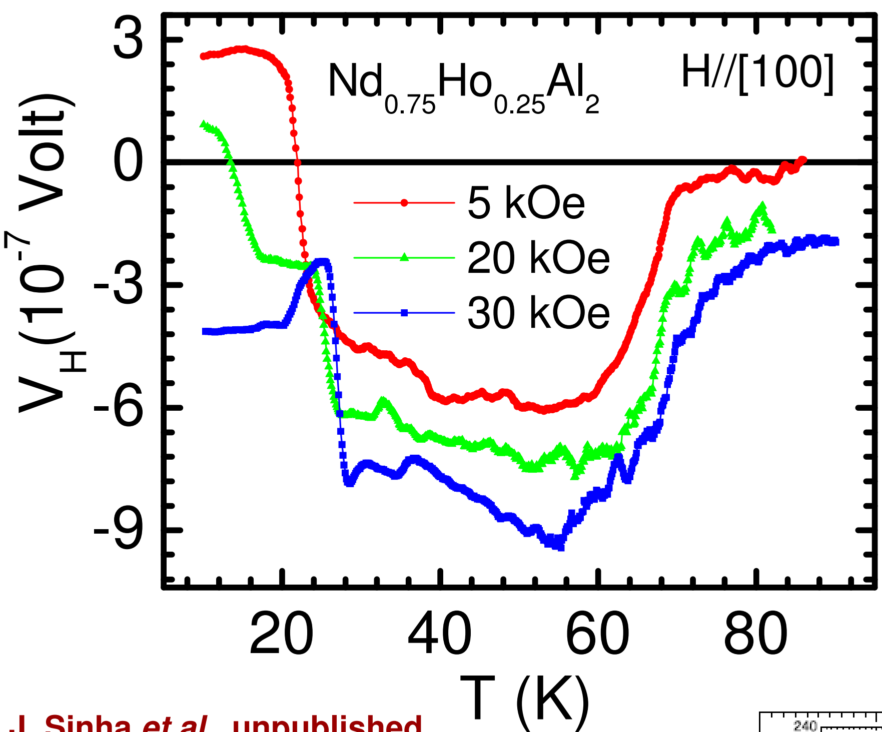
P. D. Kulkarni *et al., Europhysics Letters 86,47003 (2009)*

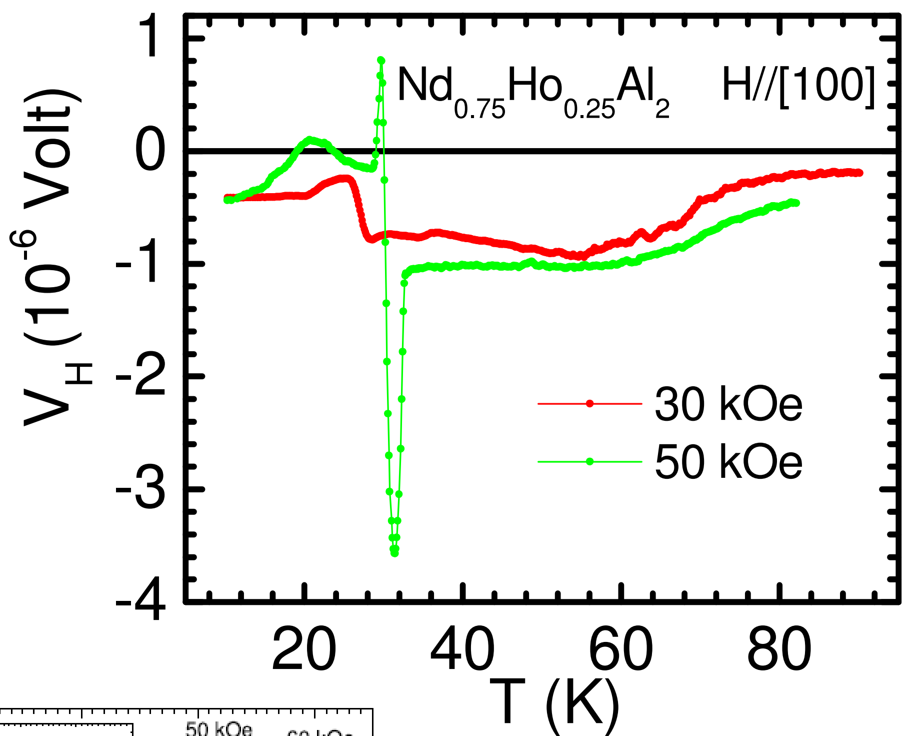
Unexpected sign reversal in Hall-voltage across T_{comp} in a single crystal of $Nd_{0.75}Ho_{0.25}Al_2$



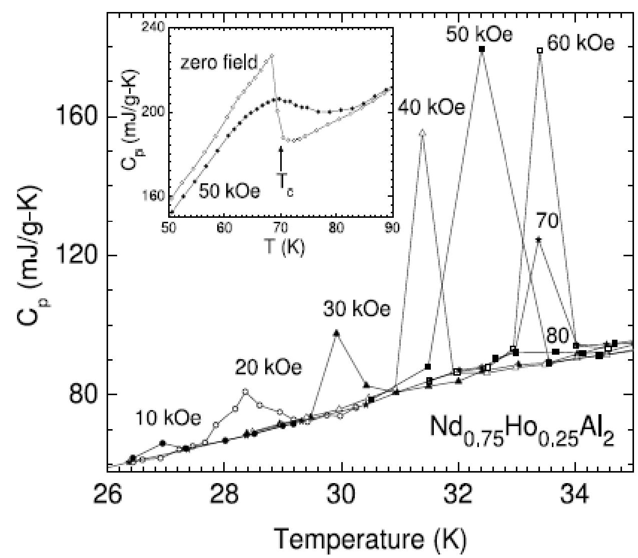
First noted in 1 & 2 % Gd doped $SmAl_2$ by Paul Chu's group in 2005 and attributed to characteristics of Sm

Further Hall-voltage & Sp. Heat data in crystal of $Nd_{0.75}Ho_{0.25}Al_2$





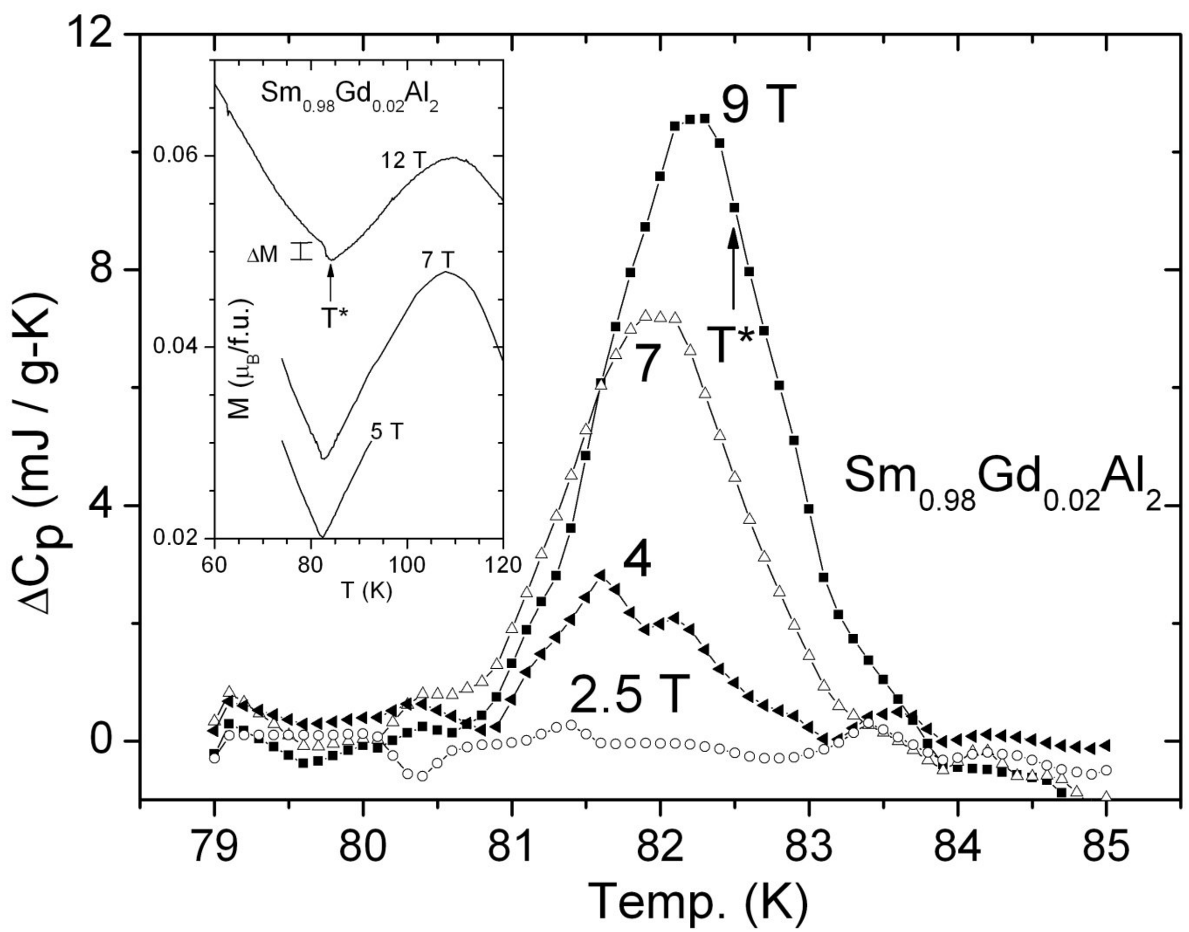
J. Sinha *et.al.*, unpublished



Sign of Hall Voltage imprints occurrence of reorientations of local moments

P. D. Kulkarni *et.a*l, Journal of Phys. Conference Series, 150, 042045 (2009)

Field induced step change in Magnetization and peak in Specific heat data at T_{comp} in $Sm_{0.98}Gd_{0.02}Al_2$



Fingerprints of field-induced phase transition at T_{comp}

S Sumithra *et al.*, unpublished

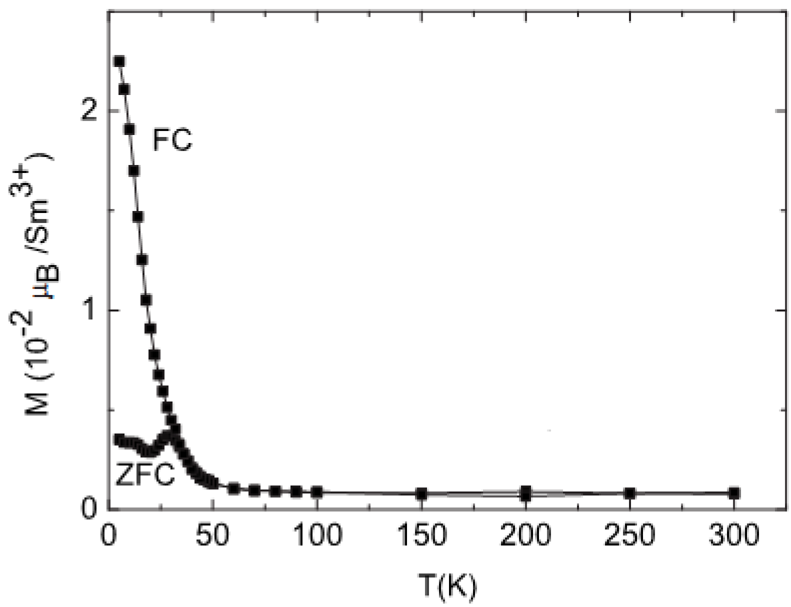
PHYSICAL REVIEW B **78**, 174406 (2008)

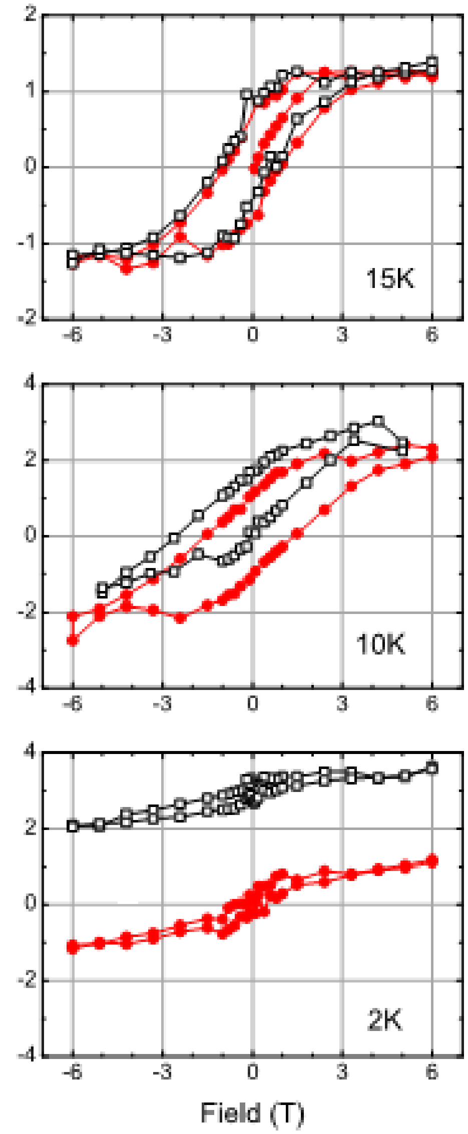
Near-zero-moment ferromagnetism in the semiconductor SmN

C. Meyer,[1,2] B. J. Ruck,[1,*] J. Zhong,[1] S. Granville,[1] A. R. H. Preston,[1] G. V. M. Williams,[3] and H. J. Trodahl[1,4]

SmN: Indirect Band Semiconductor

Band Gap ~ 1eV





No conduction Band

& No visible Exchange Bias in red curves

Moment / Sm ion at 2 K ~ 0.035 μ_B

XMCD STUDIES

X-ray Magnetic Circular Dichroism

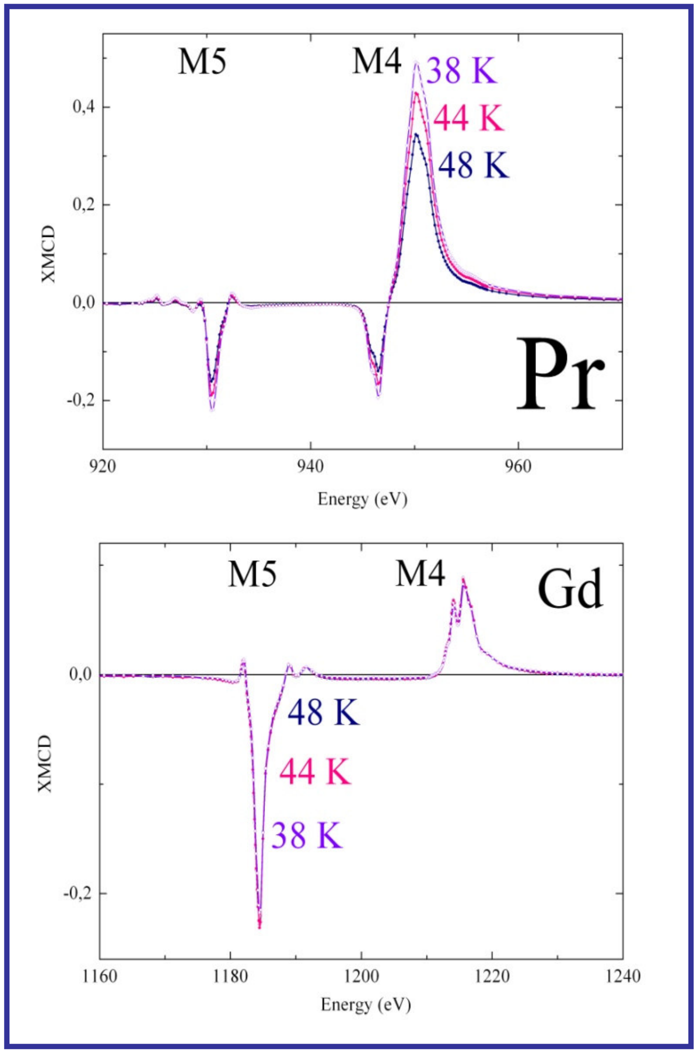
provides information on spin and orbital contribution to magnetization at individual

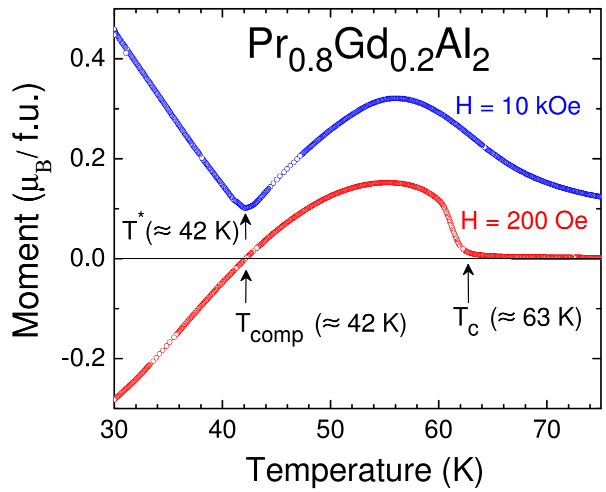
Rare Earth ion level

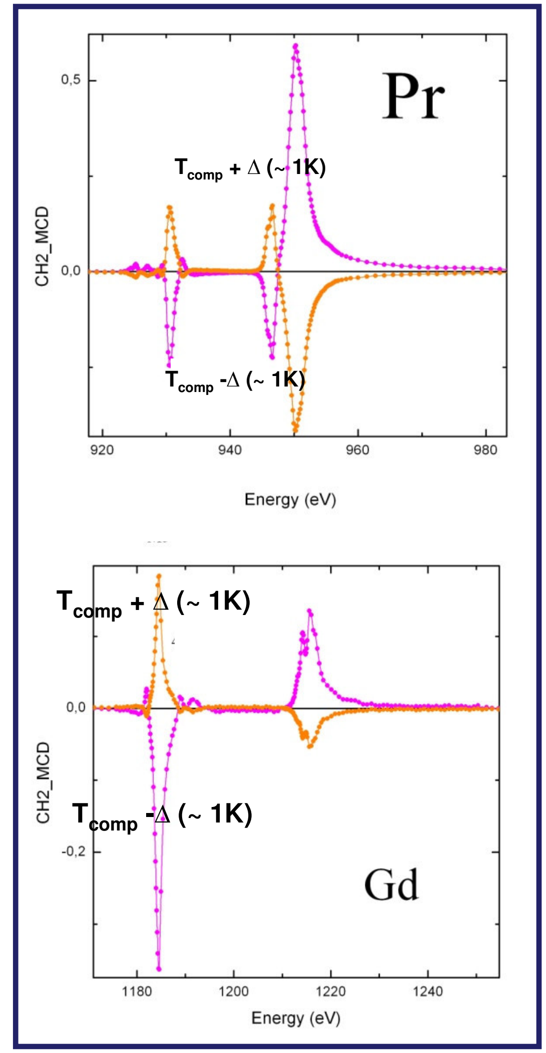
(XMCD Expts. at SPRING8, Japan)

P. D. Kulkarni, A. Thamizhavel *et al.*

XMCD data in a single crystal of $Pr_{0.8}Gd_{0.2}Al_2$







Low Field (~ 200 Oe)

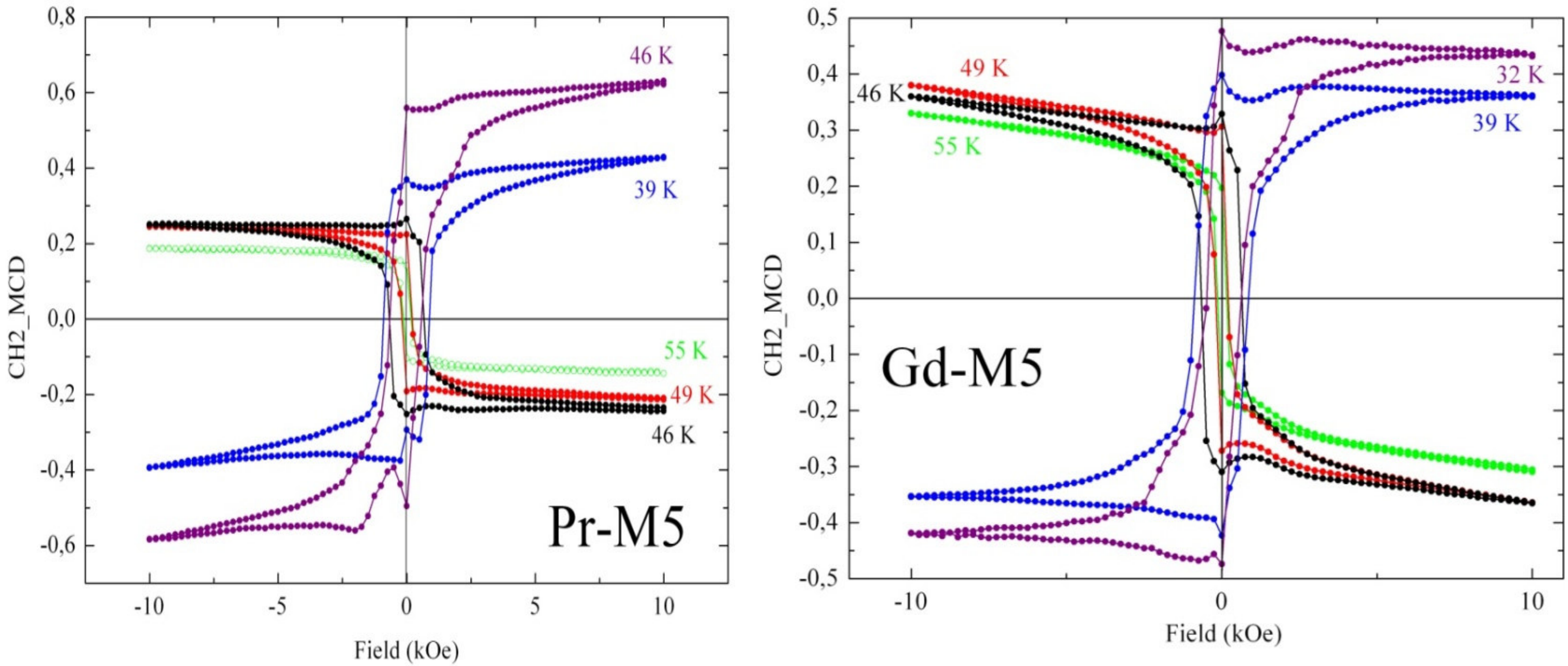
No moment reorientation at T_{comp}

P. D. Kulkarni *et al.*, Proc. DAE SSPS 2010, AIP Conf. Proc. 1349, 1197, 2011

High Field (~ 5000 Oe)

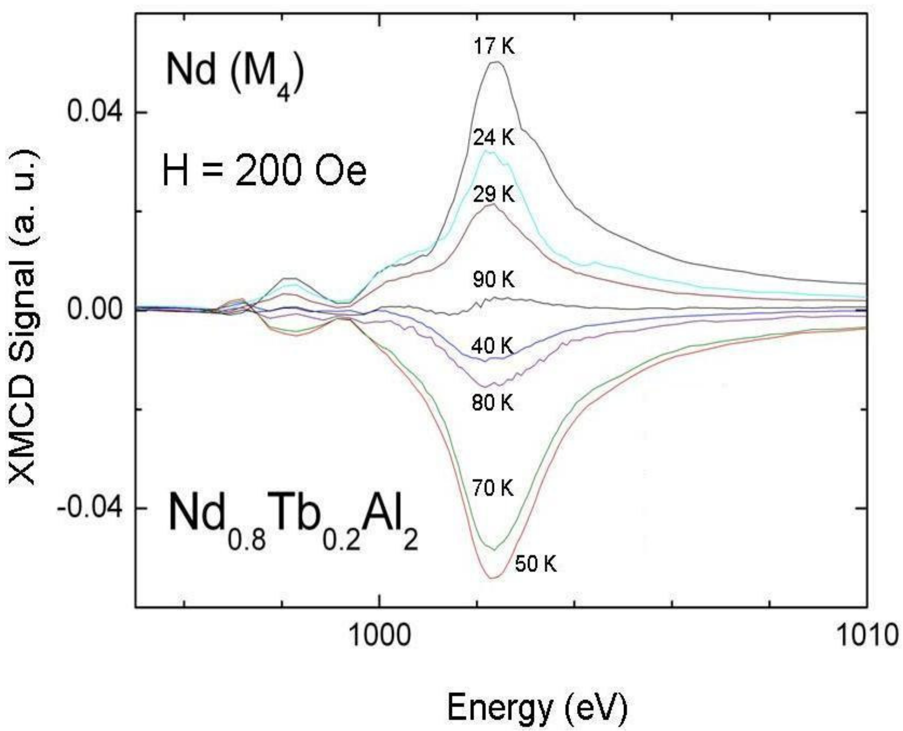
Reversal in orientation across T_{comp}

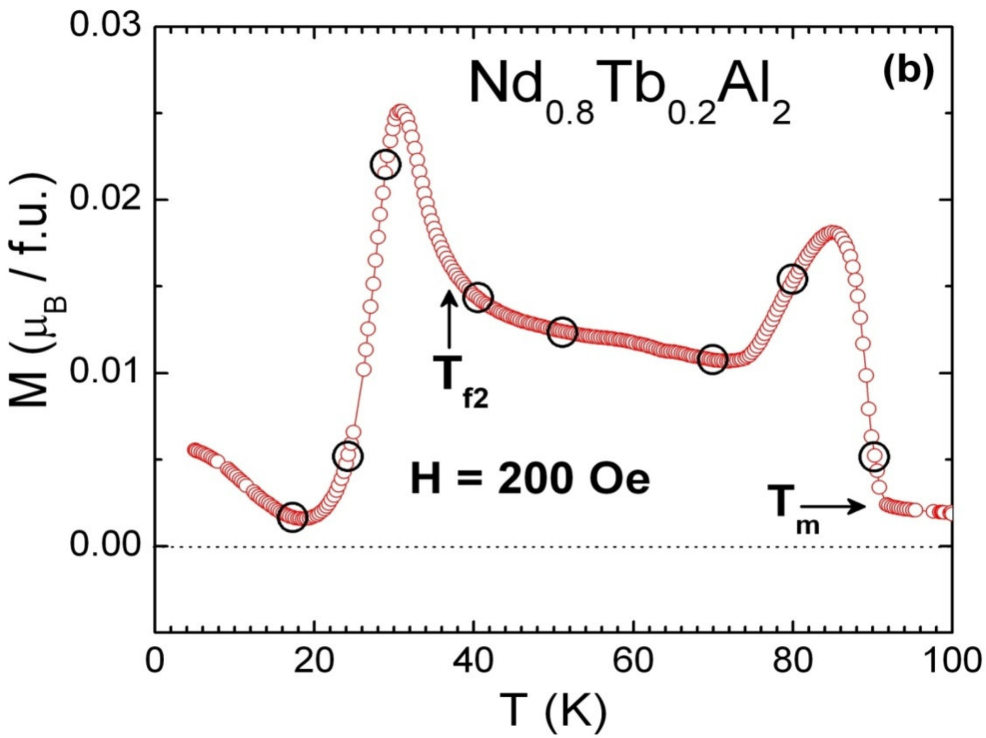
Hysteresis Loops at individual rare earth ions in single crystal of $Pr_{0.8}Gd_{0.2}Al_2$



P D Kulkarni, A Thamizhavel *et al*., unpublished

XMCD data in $Nd_{0.8}Tb_{0.2}Al_2$ elucidating repeated magnetic compensation





Spontaneous reversal in the orientationsof both the coupled local moments at the onset of second transition at T_{f2} via XMCD study at SPring 8, Japan

P. D. Kulkarni *et al.*, Proc. DAE SSPS 2010, AIP Conf. Proc. 1349, 1217, 2011

Summary

Described some of the new findings in fundamental science of magnetic materials which have potential in spintronics and magnetic STM imaging.

*** Exemplification of the existence of an exchange bias field on approaching T_{comp} and its phase reversal across T_{comp} in admixed RE intermetallics.**

*** Identified alloys undergoing magnetic orderings close to the ambient temperatures and possessing large CEP, but, near-zero bulk magnetization, and permitting easy tuning of the exchange bias field for novel applications.**

*** Self-compensation of local magnetization in pristine Sm Ferromagnets.**

*** Step change in high field magnetization and its correlation with the fingerprint of field-induced entropic change in specific heat data.**

*** Sign reversal in Hall voltage across T_{comp} and its unexpected correlation with sign change in orientation of conduction electron polarization.**

*** Oscillatory character in the magneto-resistance response, including a change in its sign at T_{comp}** (not described) .

*** Repeated magnetic compensation behavior in some specific admixed alloys near zero-magnetization stoichiometry (not described).**

Publications

1: Journal of Magnetism & Magnetic Materials, 310, 1761-1763 (2007)
U. V. Vaidya, V.C. Rakecha, S. Sumithra, S. Ramakrishnan and A. K. Grover

2: Physical Review B, 78, 064426-1 { 064426-6 (2008)
P. D. Kulkarni, U.V. Vaidya, V.C. Rakhecha, A. Thamizhavel, A. K. Nigam,
S. Ramakrishnan and A. K. Grover

3: IEEE Transactions on Magnetics, 45 2902-2906 (2009)
P. D. Kulkarni, S. Venkatesh, A. Thamizhavel, V.C. Rakhecha, S. Ramakrishnan
and A. K. Grover

4: J. Phys. D: Appl. Phys., 42 082001 (5 pages) (2009)
P. D. Kulkarni, U.V. Vaidya, S.K. Dhar, P. Manfrinetti and A. K. Grover

5: Europhysics Letters, 86, 47003-p1 { 47003-p6 (2009) *
P. D. Kulkarni, A. Thamizhavel, V.C. Rakhecha, A. K. Nigam, P.L. Paulose,
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Thank You