

From Half Metal To Spintronics (從半金屬到自旋電子學)

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Chapter One Half Metal Chapter Two Colossal Magnetoresistance Chapter Three Experiments for Spintronics



Chapter One

Half Metal



- 1. Definition of half metal
- 2. Material classification
- 3. Polarization measurement
- 4. Applications

New Class of Materials: Half-Metallic Ferromagnets

R. A. de Groot and F. M. Mueller

Research Institute for Materials, Faculty of Science, Toernooiveld, 6525 ED Nijmegen, The Netherlands

and

P. G. van Engen and K. H. J. Buschow Philips Research Laboratories, 5600 JA Eindhoven, The Netherlands (Received 21 March 1983)

The band structure of Mn-based Heusler alloys of the $C1_b$ crystal structure (MgAgAs type) has been calculated with the augmented-spherical-wave method. Some of these magnetic compounds show unusual electronic properties. The majority-spin electrons are metallic, whereas the minority-spin electrons are semiconducting.

$N(E)^{\dagger}$	N(E)	n 30 ^{Mn} t	n_{3d} ^{Mn} \downarrow	μ_{tot}^{cacl}	μ_{tot}^{exp}
9,90	0	4.51	0.87	4.00	3.85
10.05	0	4.57	0.79	4.00	3.97
9.04	2.97	4.58	0.71	4.05	3.95
9.78	19.31	4.40	0.78	3.60	3.42
	9.90 10.05 9.04 9.78	N(E) $N(E)$ 9.90 0 10.05 0 9.04 2.97 9.78 19.31	$N(E)$ t $N(E)$ t n_{3d} Mn t9.9004.5110.0504.579.042.974.589.7819.314.40	$N(E)$ t $N(E)$ t n_{3d} n_{3d} n_{3d} n_{3d} 9.9004.510.8710.0504.570.799.042.974.580.719.7819.314.400.78	$N(E)$ $N(E)$ n_{3d} m_{3d} m_{3d} μ_{tot} cacl9.9004.510.874.0010.0504.570.794.009.042.974.580.714.059.7819.314.400.783.60

Half metals are the extreme case of strong ferromagnet, where not only 3d electrons are fully polarized, but also other (sp) down-spin bands do not cross the Fermi level. (examples: NiMnSb, PtMnSb---- Hesuler phases.)

Augmented-spherical-wave method



Fig.1. Band structure for NiMnSb [1].

JOURNAL OF APPLIED PHYSICS VOLUME 91 (2002)

Half-metallic ferromagnetism: Example of CrO₂ (invited)

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A half metal is a solid with an unusual electronic structure. For electrons of one spin it is a metal with a Fermi surface, but for the opposite spin there is a gap in the spin-polarized density of states, like a semiconductor or insulator. This definition presupposes a magnetically ordered state to define the spin quantization axis. The responses of a half metal to electric and magnetic field at zero temperature are quite different. There is electric conductivity, but no high-field magnetic susceptibility.



FIG. 4. The rutile structure of CrO_2 . The local axis frame for the t_{2g} orbitals is shown.



FIG. 5. Spin polarization of the density of states of CrO_2 .

Author		$\Delta_{\downarrow}~(eV)$	$\Delta_{sf}\left(eV\right)$	$N_{\uparrow}~{ m eV^{-1}}~{ m f.u^{-1}}$
Schwarz	Ref. 26 LSDA–ASW	1.3	0.3	0.8
Lewis et al.	Ref. 2 LSDA-PWPP	1.4	0.3	0.69
Korotin et al.	Ref. 27 LSDA+U(3 eV)	2.4	1.7	0.4
Mazin et al.	Ref. 31 LSDA/GGA	1.3	0.2 - 0.7	0.95
Brener et al.	Ref. 32 LSDA-LCGO	1.3	0.2	1.16
Kuneš <i>et al</i> .	Ref. 28 GGA	1.8	0.7	0.3

TABLE III. Some electronic structure calculation on CrO2.

 Δ_{Sf} : spin-flip gap



 $\rho_o = 4 \times 10^{-8}$ ohm-m ~ 90 nm mean-free path

$$\rho = \rho_{o} T^{2} e^{-\Delta/T}$$

with $\Delta \sim 80 \mathrm{K}$

FIG. 6. Resistivity of CrO₂ thin films.



FIG. 7. Magnetoresistance of a CrO_2 – Cr_2O_3 pressed powder compact, with temperature dependence shown in the inset.

	Density		↑ electrons	↓ electrons
Туре	of states	Conductivity	at E_F	at E _F
IA	Half-metal	Metallic	Itinerant	None (CrO ₂ ,NiMnSb)
IB	Half-metal	Metallic	None	Itinerant (Sr2FeMoO6)
IIA	Half-metal	Nonmetallic	Localized	None
IIB	Half-metal	Nonmetallic	None	Localized
IIIA	Metal	Metallic	Itinerant	Localized (Magnetite)
IIIB	Metal	Metallic	Localized	Itinerant (La _{0.7} Sr _{0.3} MnC
IVA	Semimetal	Metallic	Itinerant	Localized
IVB	Semimetal	Metallic	Localized	Itinerant $(Tl_2Mn_2O_7)$
VA	Semiconductor	Semiconducting	Few, itinerant	None (Doped EuO & Eu
VB	Semiconductor	Semiconducting	None	Few, itinerant (GaAsMn

TABLE I. Summary of the classification of half-metals.



FIG. 1. Schematic density of states for a half metal, (a) Type I_A with only \uparrow electrons at E_F and (b) Type I_B with only \downarrow electrons at E_F . In narrow *d* bands, the states at E_F may be localized (type II).



FIG. 2. Schematic density of states for (a) a type III_A half metal, where electrons of one spin direction are itinerant and the others are localized, (b) a semimetal, (c) a type IV_A half metal, and (d), (e) two types of ferromagnetic semiconductor.

Definition of polarization

$$P_{0} \!=\! (N^{\uparrow} \!-\! N^{\downarrow})/(N^{\uparrow} \!+\! N^{\downarrow}), \label{eq:poly}$$

--- straightforward

$$P_n = (\langle N^{\uparrow} v^{\uparrow n} \rangle - \langle N^{\downarrow} v^{\downarrow n} \rangle) / (\langle N^{\uparrow} v^{\uparrow n} \rangle + \langle N^{\downarrow} v^{\downarrow n} \rangle).$$

--- real measurement

v: fermi velocity of electrons



3. Point Contact

5. Andreev



FIG. 3. Comparison of five methods of measuring *P*: Photoemission, tunnel junction, point contact, Tedrow–Meservey experiment, Andreev reflection.

	CrO ₂ (Ref. 2)	(La _{0.67} Ca _{0.33})MnO ₃ (Ref. 7)	Tl ₂ Mn ₂ O ₇ (Ref. 8)
N^{\uparrow} (eV ⁻¹ f.u ⁻¹)	0.69	0.58	1.25
N^{\downarrow} (eV ⁻¹ f.u ⁻¹)		0.27	0.24
$V_F^{\uparrow} (10^6 \text{ ms}^{-1})$	0.25	0.76	0.06
$V_F^{\downarrow} (10^6 \text{ ms}^{-1})$		0.22	0.33
$P_0 \%$	100	36	66
P ₁ %	100	76	-5
P ₂ %	100	92	-71

TABLE II. Calculated spin polarization in ferromagnetic oxides.

Applications of Half metals

- a. Magneto-optical effects Large Kerr rotation in PtMnSb.
- b. **Magneto-resistance applications** Spin-valve system --- pick-up head, MRAM
- c. **Spin electronics**--- Injection of polarized carriers
 - i) The spin injection in a normal metal can give information on the spin diffusion length in this metal.
 - ii) Spin injection may act as a pair-breaking agent in a superconductor.
 - iii) Half metals can also be used to build a spin transistor
 - iv) Another possible application is as polarized tips in STM, in order to visualize the orientation of magnetic domains.



Chapter Two

Colossal Magnetoresistance

Outline



Introduction
 Material structure
 Physical Properties & Mechanism

1. Introduction --- 何謂電阻(率)





Introduction ----物質的基本磁性





Introduction ---何謂磁阻













 $MR = [\rho(H)-\rho(0)]/\rho(H=6 \text{ tesla })$

S. Jin et al. Science 265 (1994)

MR ratio of spintronic materials



Туре	MR	Field	Temp.	Sample
OMR	0.01%	~Tesla	RT	Cu,Al
AMR	2 %	10 Oe	RT	Fe,Co,Ni
GMR	10 %	2 Oe	RT	Fe/Cr/Fe
TMR	40 %	10 Oe	RT	Co/AlO/Co
CMR	10 (99)%	~Tesla	$\mathbf{RT}(\mathbf{I},\mathbf{T})$	La-Sr-Mn-O

CIVIN 10(99)%~1551a

2. Material Perovskite layered structure LaMnO₃: Cubic (insulator, (La,Sr)MnO₃: orthorhombic Antiferromagnetic) (Metal, Ferromagnetic) a=5.471Å Min (0.70 Å) Mn⁺³ (1.32 Å) Ο (1.216 Å) La La,Sr c=7.77Å La, Sr b=5.5Å



X – ray diffraction pattern



Bragg condition:

 $n\lambda = 2dsin\theta$



Phase diagrams of R_{1-x}A_xMnO₃

degrees of freedom: charge, spin, orbital





3. Physical properties & mechanism

What's the new physics





- FM/AFM superexchange
- Charge/orbital ordering
- double exchange
- Lattice distortion
 (John-Teller effect)

- --- spin
- --- charge/orbital
- --- charge/spin
- --- lattice

Physical Review B, Volume 58, Number 17

1 November 1998-I

Phase diagram of manganese oxide

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$$H = H_K + H_{Hund} + H_{Onsite} + H_S$$

$$H_{K} = \sum_{\sigma \gamma \gamma'(ij)} t_{ij}^{\gamma \gamma'} d_{i\sigma \gamma}^{+} d_{j\sigma \gamma'} \text{ (Kinetic energy of } e_{g} \text{ electrons)}$$

$$H_{Hund} = -J_H \sum_{i} \vec{S}_{t_{2g}i} \cdot \vec{S}_{e_{g}i}$$

(Hund coupling between $e_g \& t_{2g}$ spins)

 $H_{\text{onsite}} = -\sum_{i} (\tilde{\beta}T_{i}^{2} + \tilde{\alpha}S_{e_{gi}}^{2}) \quad \text{(Coulomb interaction between } e_{g}\text{-electrons)}$

$$Hs = Js \sum_{(ij)} \vec{S}_{t_{2gi}} \cdot \vec{S}_{t_{2gj}}$$

(Super exchange t_{2g} spins)







(x=0.0) ; spin F



(x=0.3) ; spin F (0.3<x<0.8) ; spin F



Α

(x=0.0) ; spin A

(x=0.0) ; spin C



(0.1<x<0.45) ; spin A

(x≠0.0) ; spin C









(0.45<x<0.75); spin A



Charge Ordering $(Mn^{+4}/Mn^{+3} = n/8)$



Spin/orbital structure

a (b) (Pbnm) Mn³⁺ С -b(a)Mn⁴⁺

Neutron diffraction Magnetic Dichroism


Kajimoto et al., PRB 30, 9506 (1999)

Orbital switching in A-type spin state



Origin of CMR: Phase separation



Spin line up (metal)

M. Uehara, et all, Nature 399 (1999)



Chapter Three Experiments for Spintronics



III-1. Bulk

Phase separation --- fine tuning the MR value by Ionic radius size



Key parameters

hole concentration (charge/orbital/spin)
radius of A-site (lattice/ spin)







Physical Property measurement System (PPMS)



0 – 7 Tesla 1.4 – 400 K Hall effect resisitivity AC susceptibility



Para- to antiferromagnetic

Para- to ferromagnetic



 $M \sim C/T$, $C/(T+T_N)$, C/(T-Tc)





PHYSICAL REVIEW B, VOLUME 65, 024422 (2001) Thermal and magnetic instability near the percolation threshold of Nd_{0.5}Ca_{0.5-y}Sr_yMnO₃

C. W. Chang,* A. K. Debnath,† and J. G. Lin‡



PHYSICAL REVIEW B, VOLUME 65, 024422 (2001) Thermal and magnetic instability near the percolation threshold of Nd_{0.5}Ca_{0.5-y}Sr_yMnO₃

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 $\Delta \rho / \rho = A \ln t + const.$

J OF APPLIED PHYSICS VOLUME 90 (2001) Enhancement of magnetoresistance in the intermediate state

of Pr_{0.5}Sr_{0.3}Ca_{0.2}MnO₃

C. W. Chang and J. G. Lin



FIG. 2. I - V curves for Pr0.5Sr0.3Ca0.2MnO3 at 120, 100, and 80 K, respectively. The arrows denote the jumps of voltage. The inset shows the temperature dependence of the onset voltage of the jump with the forward scan (increasing) and the backward scan (decreasing).

J OF APPLIED PHYSICS VOLUME 90 (2001)

Enhancement of magnetoresistance in the intermediate state

of Pr_{0.5}Sr_{0.3}Ca_{0.2}MnO₃

C. W. Chang and J. G. Lin



FIG. 3. Current dependence of V and MR ratio at 95 K with different magnetic fields.

J OF APPLIED PHYSICS VOLUME 90 (2001) Enhancement of magnetoresistance in the intermediate state

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III-2 films & bilayers



Subject (1): Nanocrystalline LSMO films ---- High electroresistance & low field MR effects

Experiment Flow Chart



PPMS & SQUID



Target preparation

 $\bigstar \text{Nd}_{0.7}\text{Ca}_{0.3}\text{MnO}_3 \text{ (NCMO):}$

 Nd_2O_3 , CaCO₃, MnCO₃ Powder 100°C (3 hrs.) in air 1100°C (24 hrs.) in air

♦ YBa₂Cu₃O₇ (YBCO)

 Y_2O_3 , BaCO₃, CuO₂ Powder 100°C (3 hrs.) in air 1050°C (36 hrs.) in oxygen 400°C (12 hrs.) in oxygen

Powders



Solid state reaction





RF sputter system (Millton CVT, 13.6 MHz)



10⁻⁸ torr four guns 1000 °C





Reactive co-sputtering process

• Substrate \Rightarrow	Si (100) , LaAlO ₃ (100)		
• Target ⇒	YBCO, LSMO		
• RF power ⇒	80 Watt		
• Base pressure \Rightarrow	3 × 10 ⁻⁷ torr		
• Mixed gas \Rightarrow	Ar:0 ₂ =98:2		
 Sputtering pressure 	70 mtorr		
• Base temperature \Rightarrow	Room temperature		
• pre-sputtering \Rightarrow	3 minutes		
• Working distance \Rightarrow	10 cm		
 Annealing tempert. 	800 – 920 °C (700 °C)		
• Annealing time \Rightarrow	1 hrs		



Non-uniform strain in La_{0.67}Ca_{0.33}MnO₃ film



Lattice parameter a LCMO=3.86Å NGO(110)=3.86 Å (no strain) LAO(100)=3.79 Å (Compressive) STO(100)=3.91 Å (Tensile)

PRB 61, 9665 (2000)

Structures of LSMO layer

XRD – monoclinic

c-oriented



Magnetization for LSMO

H=500 Gauss ZFC(black line); FC (red line)









JOURNAL OF APPLIED PHYSICS 98, 114318 (2005)

Low-field magnetoresistance in nanocrystalline La_{0.7}Sr_{0.3}MnO₃ films

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(Received 28 March 2005; accepted 1 November 2005; published online 14 December 2005)

Nanocrystalline La_{0.7}Sr_{0.3}MnO₃ films with thickness t=10-60 nm were grown on LaAlO₃(100) substrates by radio-frequency magneton sputtering. Their electrical resistivity and low-field magnetoresistance (MR) were measured. Metal-insulator transitions occur above 275 K for films with t=20-60 nm, but the electron localization prevails in the 10 nm thick film. Furthermore, only the 10 nm thick film has an MR that depends on the inverse of temperature, consistent with the model of spin-polarized tunneling. This relationship may reflect a critical aspect of the structure of grain/grain-boundaries. Accordingly, the tunneling MR in this film is 27% at 75 K. © 2005 American Institute of Physics. [DOI: 10.1063/1.2140081]

PHYSICAL REVIEW B 67, 064412 (2003)

Current-induced giant electroresistance in La0.7Sr0.3MnO3 thin films

A. K. Debnath* and J. G. Lin[†]



PHYSICAL REVIEW B 67, 064412 (2003) Current-induced giant electroresistance in La0.7Sr0.3MnO3 thin films A. K. Debnath* and J. G. Lin[†]



PHYSICAL REVIEW B 67, 064412 (2003)

Current-induced giant electroresistance in La0.7Sr0.3MnO3 thin films

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Current in nanowire of LSMO induced magnetic field, and the thermal energy delocalized the electrons.

thickness (nm)	MR _H (%)	MR _I (%)	ratio
60	2.96	5.6 (I = 0.3)	1.9
80	2.85	6.4 (I = 0.3) 11.3 (I = 0.9)	2.3 4.0
100	3.85	3.5 (l = 0.3) 4.6 (l = 0.9)	0.9 1.2



Subject (2): Nanocrystalline YBCO/LSMO bilayers --- Proximity effect, spin injection & vortex pinning

Multilayer spintronic devices



Spin-injection I_c - depression

proximity effect T_c - depression FM/N/FM structure



Interesting topics on YBCO/LSMO



- 1) Proximity effects
- 2) Andreev reflection
- 3) Spin injection
- 4) Spin-accumulation
- 5) π –phase shift


1) Influence of magnetism on supercond.

Intermixing \Rightarrow effective exchange field $H_{effect} = H_{ex} [d_F/(d_s+d_F)], T_c oscillation$

2) Influence of supercond. on magnetism

Intermixing \Rightarrow reconstruction of magnetic order

T_{curie} oscillation



Spin-injection



- Normal electron injected to superconductor will be reflected as a hole;
- Polarized electron injected into superconductor will kill a Cooper pair.

SCIENCE VOL 282 2 OCTOBER 1998



Superconductor: $RBa_2Cu_3O_7$, R = Y or rare earth element



• YBa₂Cu₃O₇ $\xi_s \sim 3-10 \text{ nm}$ • Critical parameter T_c = 90 K, H_{c2} ~ 165 T • SQUID, Bolometer, Filter, Resonator...



a=3.89Å

Wu, Chu et al., PRL (1987)



Half metal: $(R,Ca)MnO_3$, R = La, Nd, Pr....





Jin et al, Science (1995)



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LCMO = 15 unit cell ~ 10 nm

Superconductivity depression in ultrathin $YBa_2Cu_3O_{7-\delta}$ layers in $La_{0.7}Ca_{0.3}MnO_3/YBa_2Cu_3O_{7-\delta}$ superlattices

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Superconducting and transport properties of YBa₂Cu₃O₇/La_{0.7}Sr_{0.3}MnO₃ bilayers

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Vortex pining device









I-V characteristic



Critical current I_C Normal state resistance RnEnergy gap Δ $Ic = I(V=1 \ \mu V)$ $I_C Rn = \pi \Delta / 2$

$$= 3.52 \pi K_{\rm B} T_{\rm C} / 4$$



I-V Characteristic (1.9 K)



Ic decreases with increasing thickness of LSMO



Hysteresis in YBCO/LSMO(30nm)





Available online at www.sciencedirect.com



Physica C 437-438 (2006) 187-189



www.elsevier.com/locate/physc

High field pinning-effects in YBa₂Cu₃O₇/La_{0.7}Sr_{0.3}MnO₃ nanocrystalline bilayers

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Available online 26 January 2006



Subject (3): Epitaxial YBCO/NCMO bilayers ---- How the superconductivity is affected by weak ferromagnetic insulator ?



Temperature (K)



E. Dagotto *et. al.*, Physics Reports 344, 1-153 (2001) C. N. R. Rao et al. J. Phys. Chem. Solids 59, 487 (1998) APPLIED PHYSICS LETTERS 88, 222507 (2006)

Low-current-induced electrical hysteresis in Nd_{0.7}Ca_{0.3}MnO₃

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- 1. Atomically flat
- 2. Chemically comparable
- 3. Well lattice mismatch

For YBCO, LaAlO₃ (100) is ~ 0.4%;

SrTiO₃(100) is 0.2% for *a*-parameter, 0.3%

for *b*-parameter;

 $NdGaO_3$ (110) is 0.7 % for *a*- and *b*-parameters



Nd_{0.7}Ca_{0.3}MnO₃ (001), orthorhombic

YBa₂Cu₃O₇ (001), orthorhombic

LaAIO₃ (100), rhombohedra







Pulse-Laser-Deposition system –Neocera 180

KrF Laser (248 nm)

Focus Len



Chamber 10⁻⁸ torr

Pump Flow meter Vacuum gauge

Pulse-Laser Deposition system (II) - Chamber

YBCO





NCMO







Both NCMO & YBCO are with c-axis perpendicular to the film surface









Basic characterization

(High resolution TEM & electron diffractions)



(Scanning EDX)





Magnetization(M) vs. Temperature (T) - by SQUID

T_c is 88 K for YBCO; T_N is ~ 75 K for NCMO; Both transition temperature do not change in NCMO(40nm)/YBCO(160nm).





4-Probe method

• YBCO : $T_c \sim 88 \text{ K} \& \Delta T < 2 \text{ K}$; NCMO: Insulating





Resistivity (ρ) vs. Temperature (T)

For I_a = 1 – 90 mA, normal state increases & T_c drops with a rate 0.1K/mA.

open a gap at 230 K ?





• For $I_a = 5 - 30$ mA, resistivity decreases.

5 mA 10⁴ 10 mA — 20 mA – 30 mA p (ohm-cm) 10⁻¹ 10³ p (ohm-cm) 10² 200 250 0.0001mA T(K) 0.001mA 10¹ 0.01mA 0.1mA 10⁰ 1mA 100 150 200 250

NCMO(200nm)

T(K)



Resistivity (ρ) vs. Temperature (T)





Resistivity (ρ) vs. Temperature (T)

For I_a = 1 – 40 mA, normal state decreases 30% & T_c drops with a rate of 1.4 K/mA.





Superconducting temperature vs. applying current





Proximity effect

 $H_{eff\text{-}ex} = H_{ex} \{ d_F / (dS + d_F) \}$

 $0.5H_{ex}$ for NCMO(200nm)/YBCO(200nm) $0.2H_{ex}$ for NCMO(40nm)/YBCO(160nm)

 $\Delta T_c \sim 34 K / 16 K$

• Current induces spin-injection effect

Observe a threshold of effective current I > 1 mALarge T_c-suppresion in a rate of 1.4 K/mA. JOURNAL OF APPLIED PHYSICS 101, 09G106 (2007)

Proximity effect of superconductivity and magnetism in the Nd_{0.7}Ca_{0.3}MnO₃/YBa₂Cu₃O₇ bilayer

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Current enhanced magnetic proximity in Nd_{0.7}Ca_{0.3}MnO₃/YBa₂Cu₃O₇ bilayer

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Thickness dependent spin-injection effects in Nd_{0.7}Ca_{0.3}MnO₃/YBa₂Cu₃O₇ bilayers

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Two Nd_{0.7}Ca_{0.3}MnO₃/YBa₂Cu₃O₇ (NCMO/YBCO) bilayers with different thickness ratios are fabricated and the spin-injection effects are investigated. The NCMO/YBCO samples have thicknesses of 100 nm/200 nm and 200 nm/200 nm, which are denoted as N/Y(1) and N/Y(2), respectively. It is shown that the current-induced suppression rate of superconducting transition temperature (dT_c/dI) in YBCO is enhanced by four to six times of magnitude in N/Y(1) and N/Y(2) compared with that in pure YBCO. Furthermore, dT_c/dI in N/Y(2) is larger than that in N/Y(1), which suggests that the thickness of NCMO has influence on the pair breaking in YBCO. © 2008 American Institute of Physics. [DOI: 10.1063/1.2833819]



Conclusion Remark

Half metal is the future material for various kinds of spintronic devices