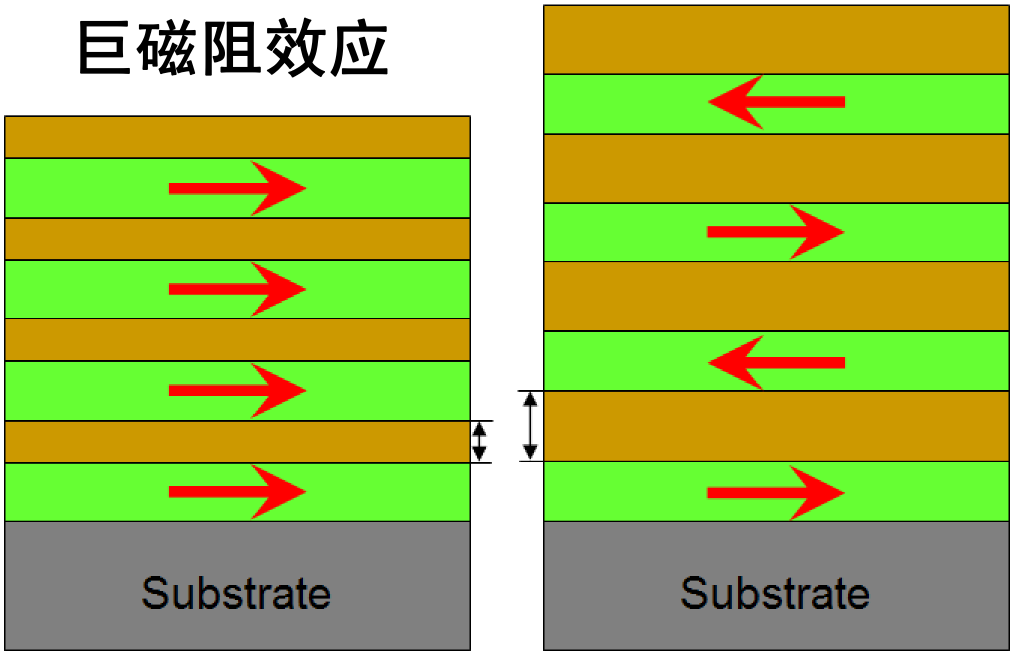
Tunable Magnetoresistance in Organic Spin Valves

Jian Shen

Department of Physics, Fudan University

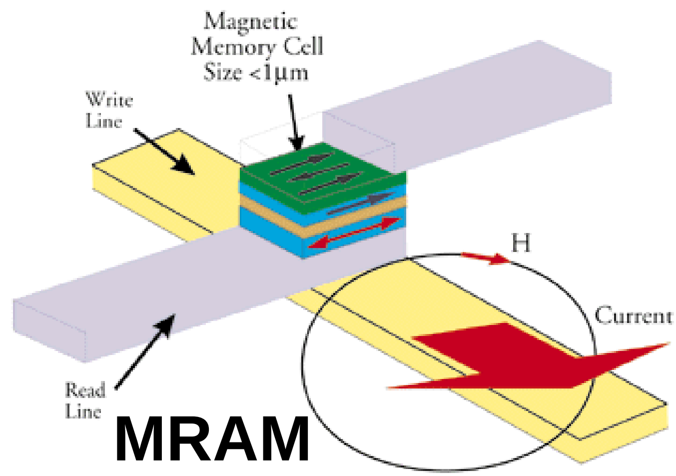
Inorganic Spin Valves and GMR

巨磁阻效应



PRL (1988)

硬盘读写(1997)

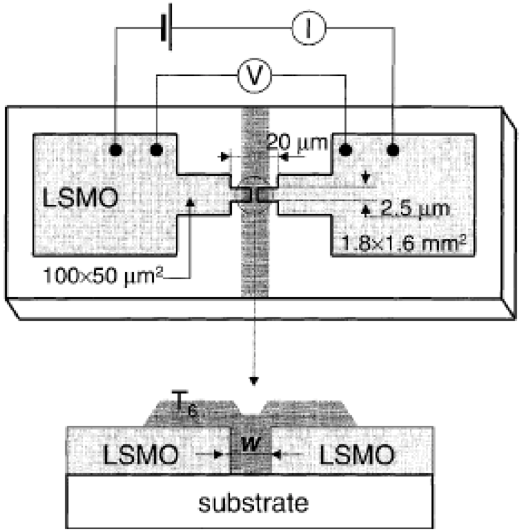


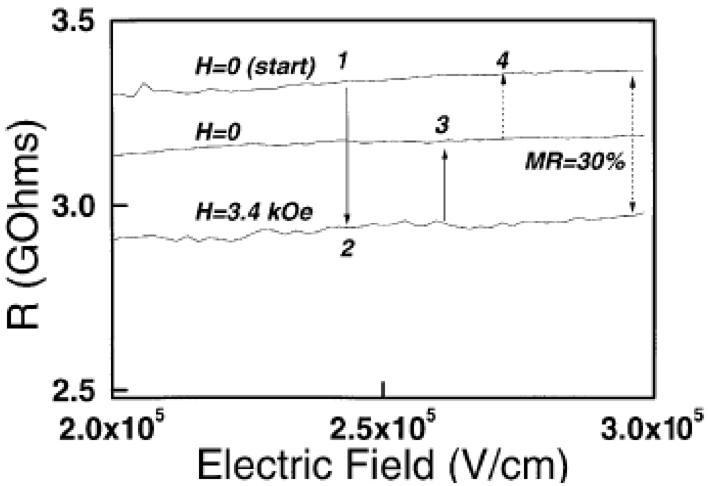
MRAM

The Nobel Prize in Physics 2007

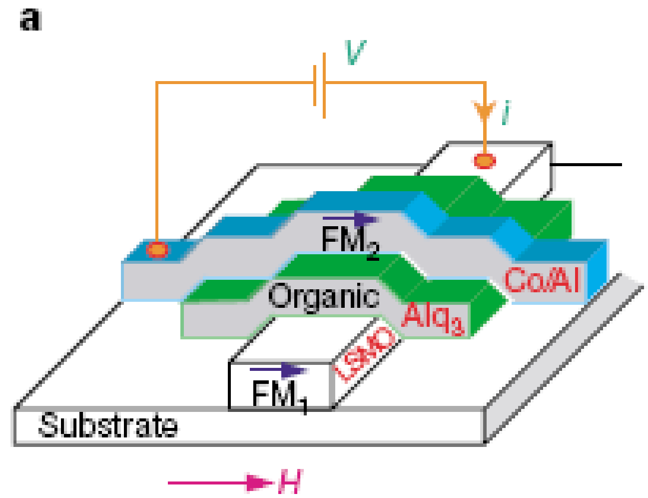
Albert Fert **Peter Grünberg**

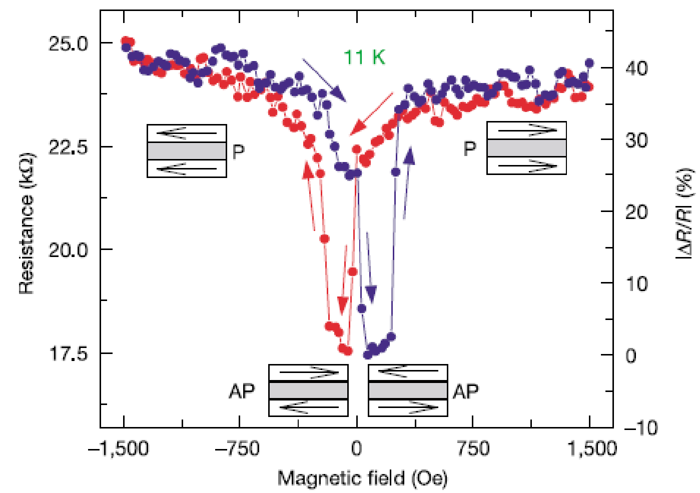
Organic Spin Valves





V. Dediu *et al.*, Solid State Comm 122, 181 (2002)

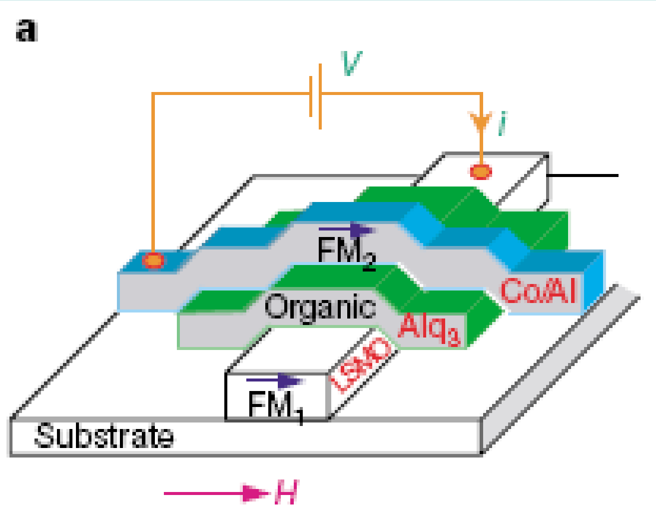




Z. H. Xiong *et al.*, Nature 427, 821 (2004)

Magnetoresistance in Organic Spin Valves

Spin Injection

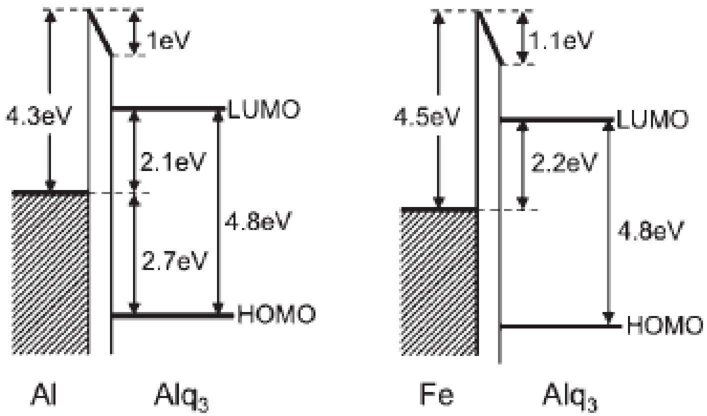


Z. H. Xiong *et al.*, Nature 427, 821 (2004)

- **~ 100nm organic layer**
- **Negative MR**
- **A few tens percent at low T**

Unclear issues

- Charge transport mechanism



- Spin-dependent transport mechanism
- Inconsistency with MR

MR is not large, and not tunable.

Origins of Controversies

PRL 102, 66104 (09); Nature Comm 5, 4528 (14)

Top electrode fabrication

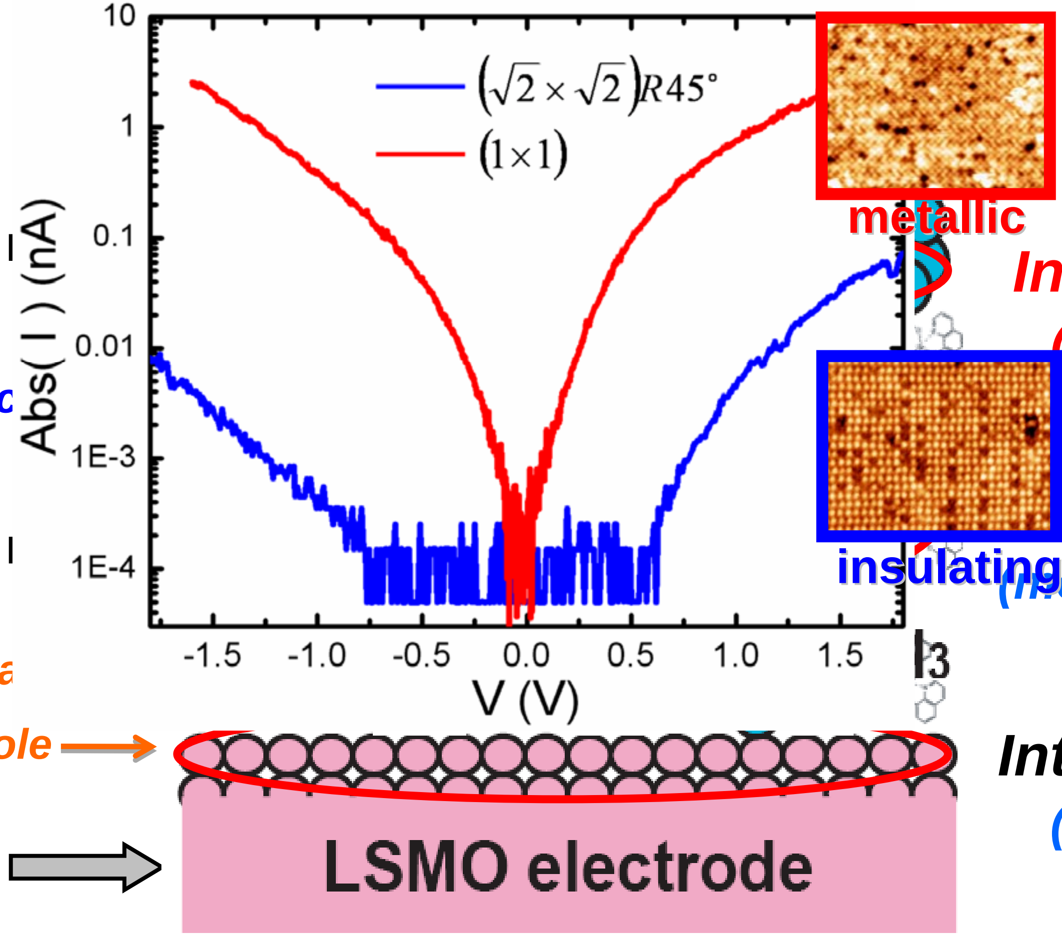
rough surfac

Organic material

Defects and tra

Interfacial dipole

Bottom electrode fabrication



Interface #2

(Interdiffusion)

ulk properties

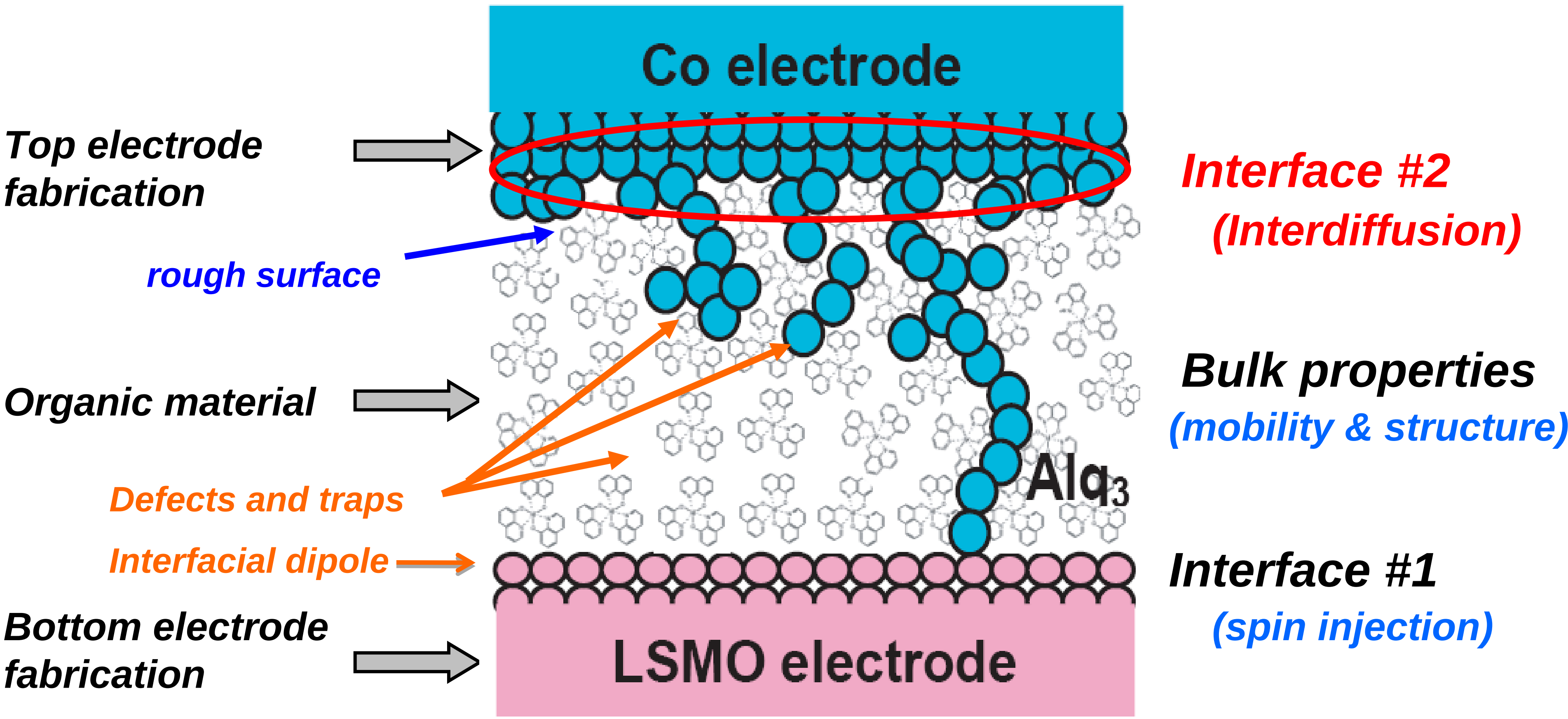
(mobility & structure)

Interface #1

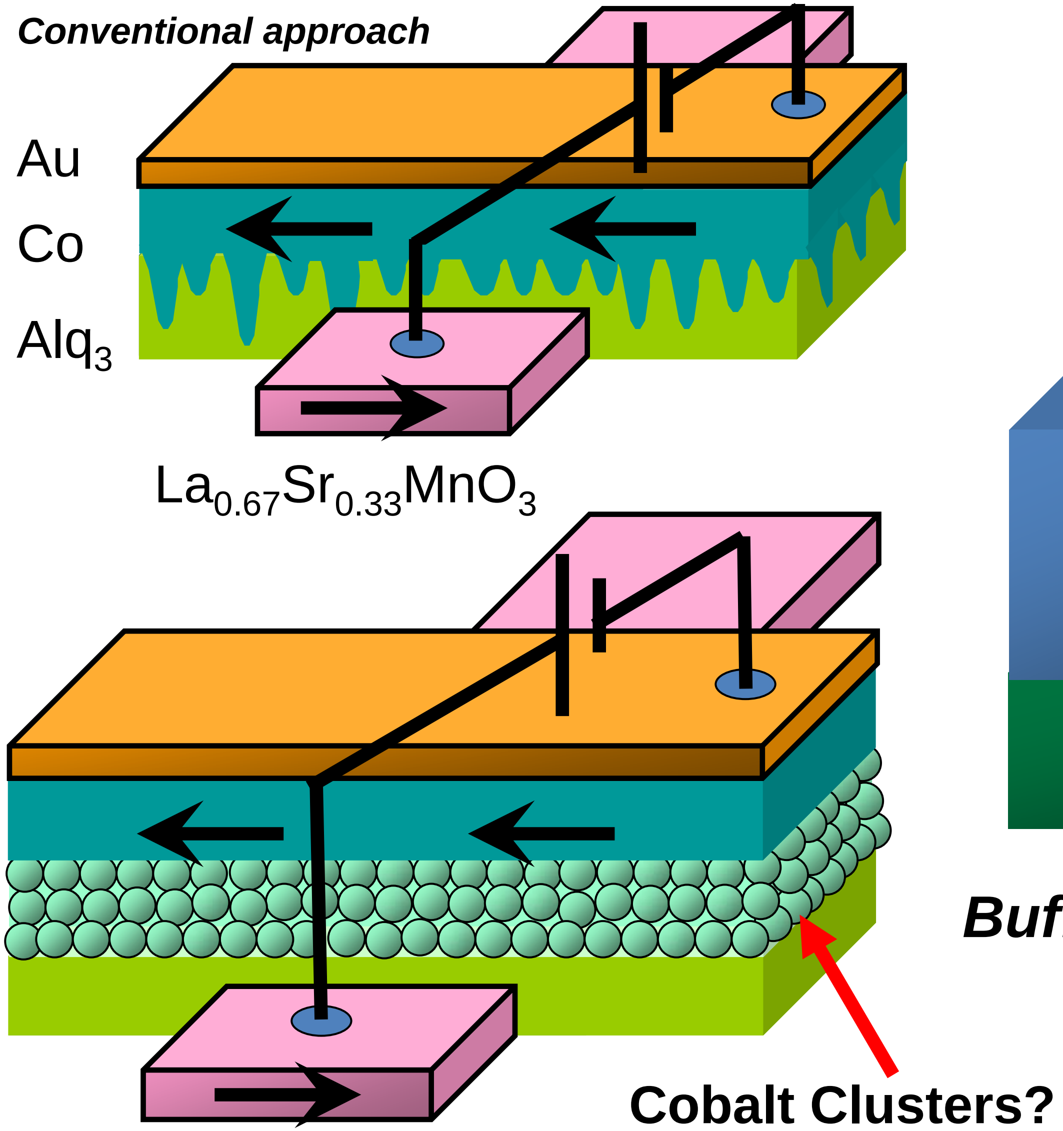
(spin injection)

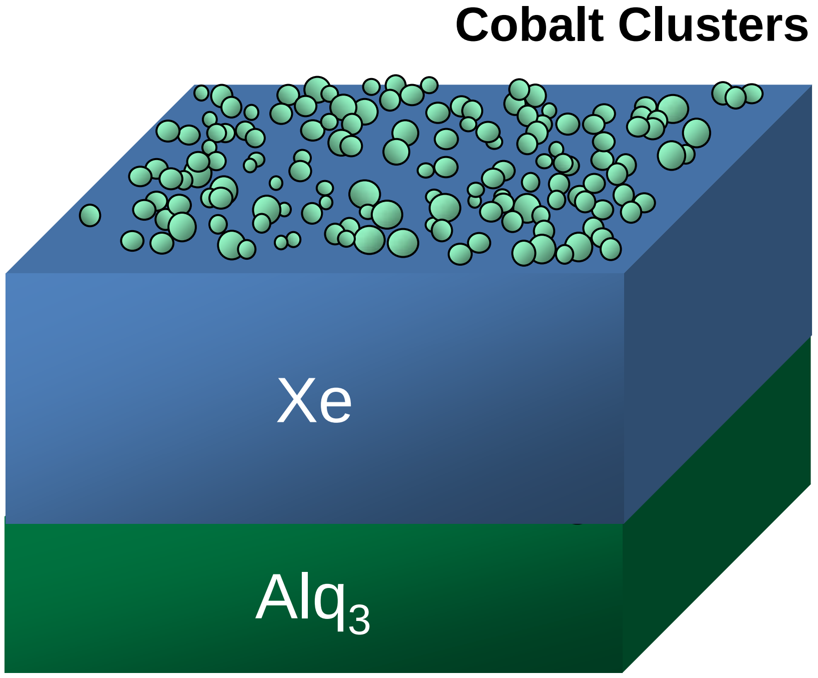
Controllable system is needed

Part I


Co electrode
Top electrode fabrication
rough surface
Organic material
Defects and traps
Interfacial dipole
Bottom electrode fabrication
LSMO electrode
Alq3
Interface #2
(Interdiffusion)
Bulk properties
(mobility & structure)
Interface #1
(spin injection)

Control Interdiffusion between Co and Alq_3

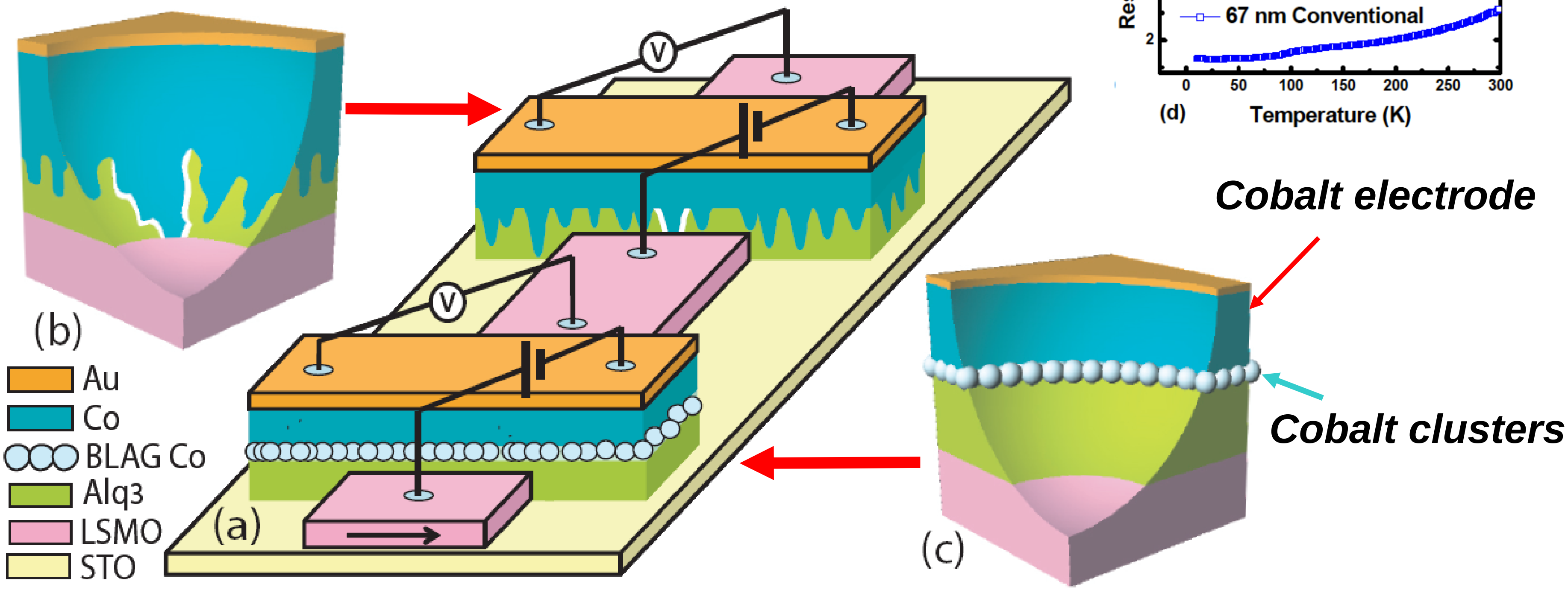


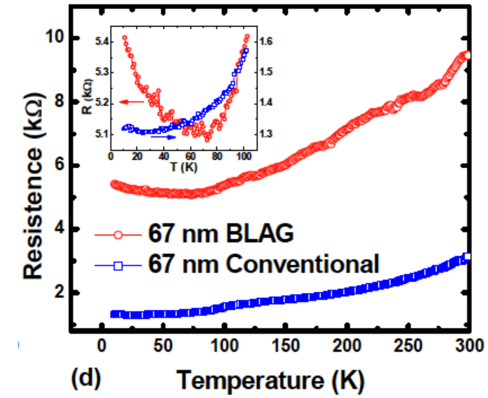


Buffer-layer-assisted growth

Comparing Conventional and BLAG Spin Valves

Conventional approach



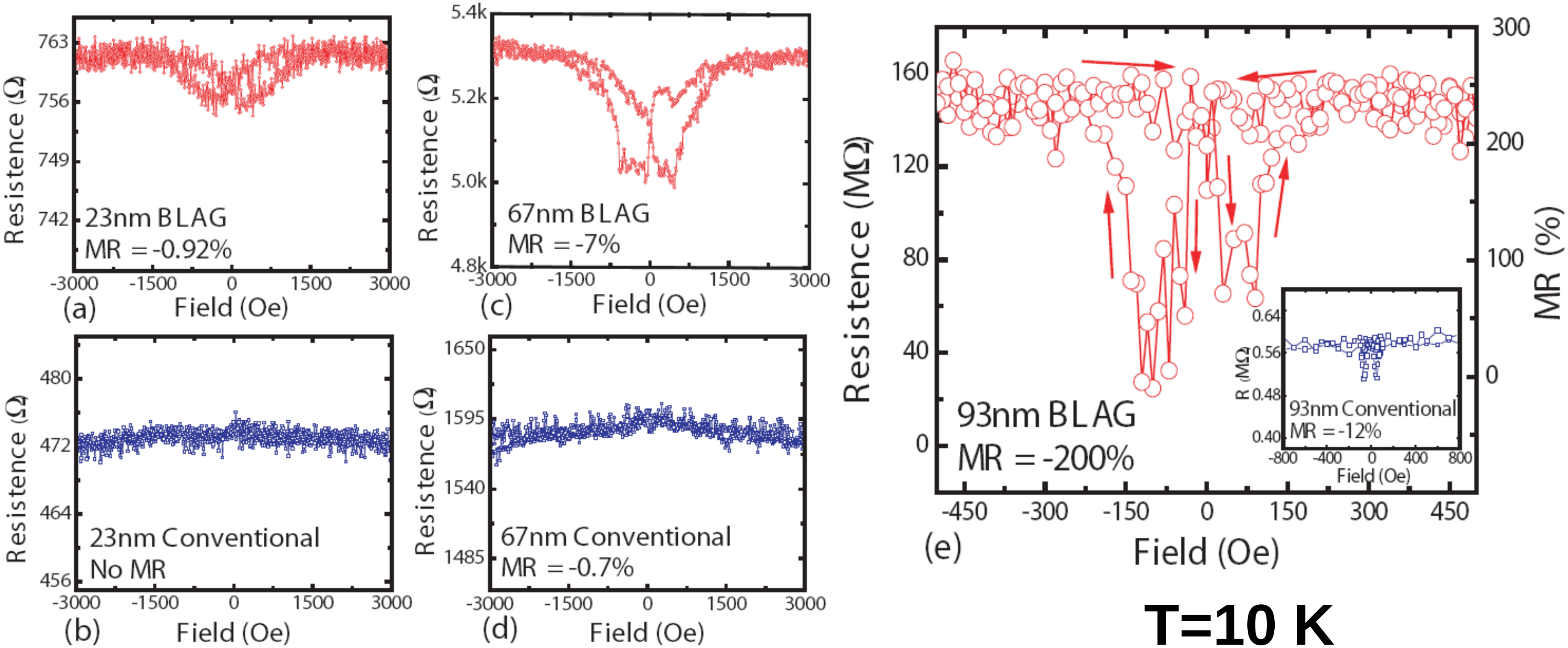


Active device area
~ 1 mm x 2 mm

Buffer-layer-assisted growth

Alq_3 Thickness Dependence of MR

At lower d_{eff} | At higher d_{eff}



T=10 K

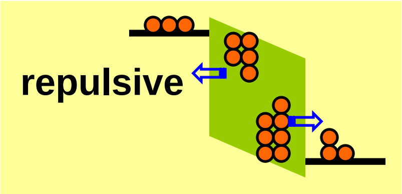
MR of BLAG spin valves is much larger than that of conventional ones.

Phys. Rev. Lett. 104, 236602 (2010)

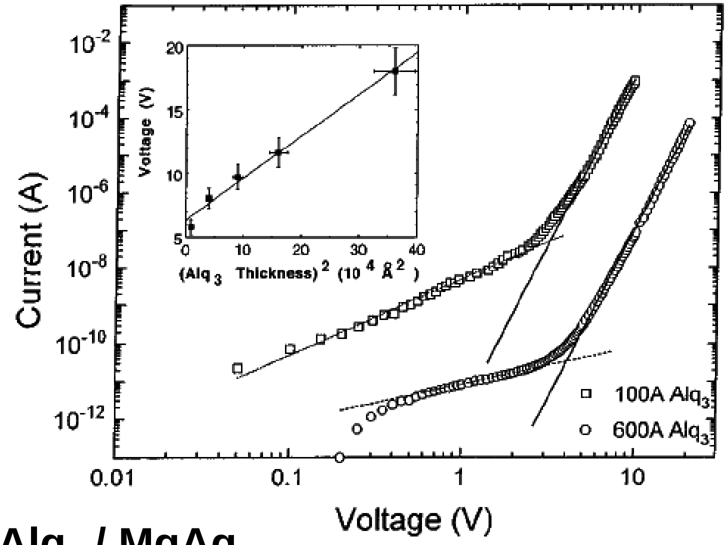
Transport Through an Organic Layer

Bulk-limited current

$$J \propto V^m$$



m = 1 low-mobility ohmic conduction
m = 2 *space-charge-limited (SCLC)*
m > 2 trapped-charge-limited (TCL)



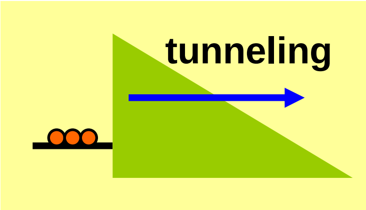
TPD / Alq_3 / MgAg

P. E. Burrows, *et al.*, JAP v79 p7991 (1996)

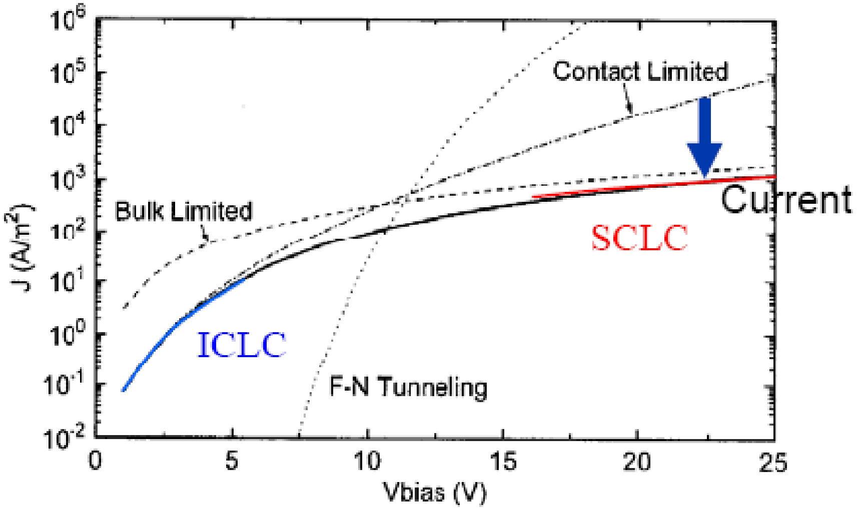
Injection-limited current

$$J(F) = BF^2 \exp\left[-\frac{4(2m_{eff})^{1/2} \cdot \Delta^{3/2}}{3\eta eF}\right]$$

$$B = \frac{e^3}{8\pi h \Delta}$$

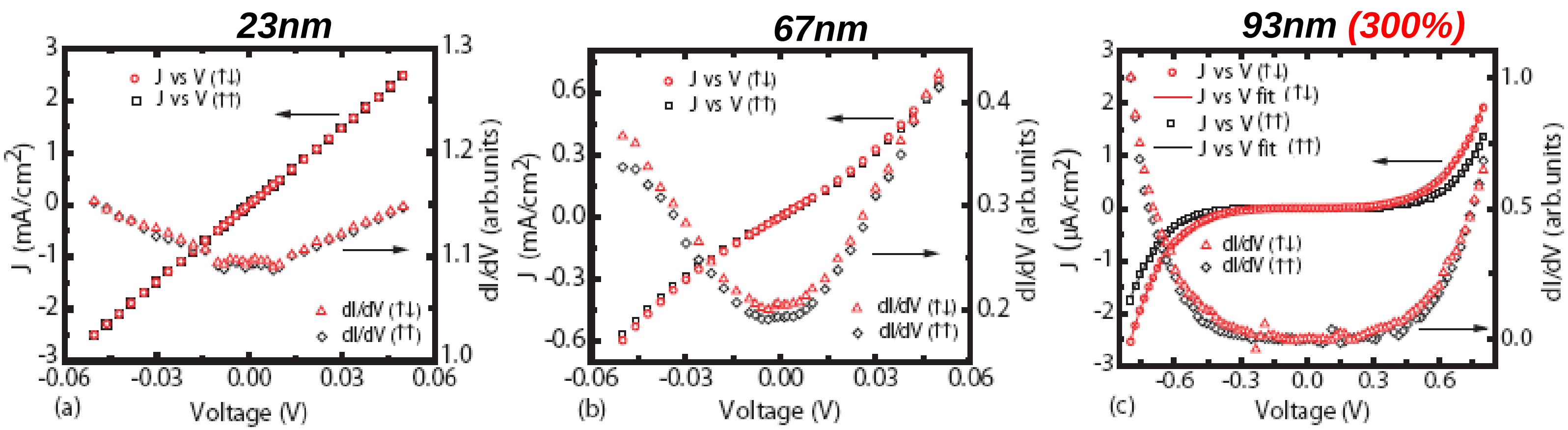


Fowler-Nordheim (FN) model for tunneling



V. I. Arkhipov *et al.*, JAP v84 p848 (1998)
M. A. Baldo *et al.*, PRB v64 p085201 (2001)

Model Fitting of J-V curves



$m \approx 1.0$ **Ohmic**

$m \approx 1.3$ **Ohmic+SCLC**

$m \approx 4.0$ ***TCL?***

Bias is still small!

Bulk limited current: $J \propto V^m$

Or SCLC (Mott-Gurney law) $J = \frac{9}{8}\mu\varepsilon\varepsilon_0 \frac{V^2}{L^3}\theta_0$

Simmons' tunneling (93nm): $\varphi_{\uparrow\uparrow} = 1.48eV, \varphi_{\uparrow\downarrow} = 1.47eV, d_{barrier} \approx 3nm$

Fowler-Nordheim tunneling: $J(F) = BF^2 \exp[-\frac{4(2m_{eff})^{1/2} \cdot \Delta^{3/2}}{3\eta eF}]$

$$B = \frac{e^3}{8\pi h\Delta}$$

SCLC with screened Frenkel effect

If the potential of the ion trap centers is ***screened***

$$V(x) = -\frac{e^2}{4\pi\varepsilon\varepsilon_0|x|}(-\frac{|x|}{\lambda}) - Ex;$$

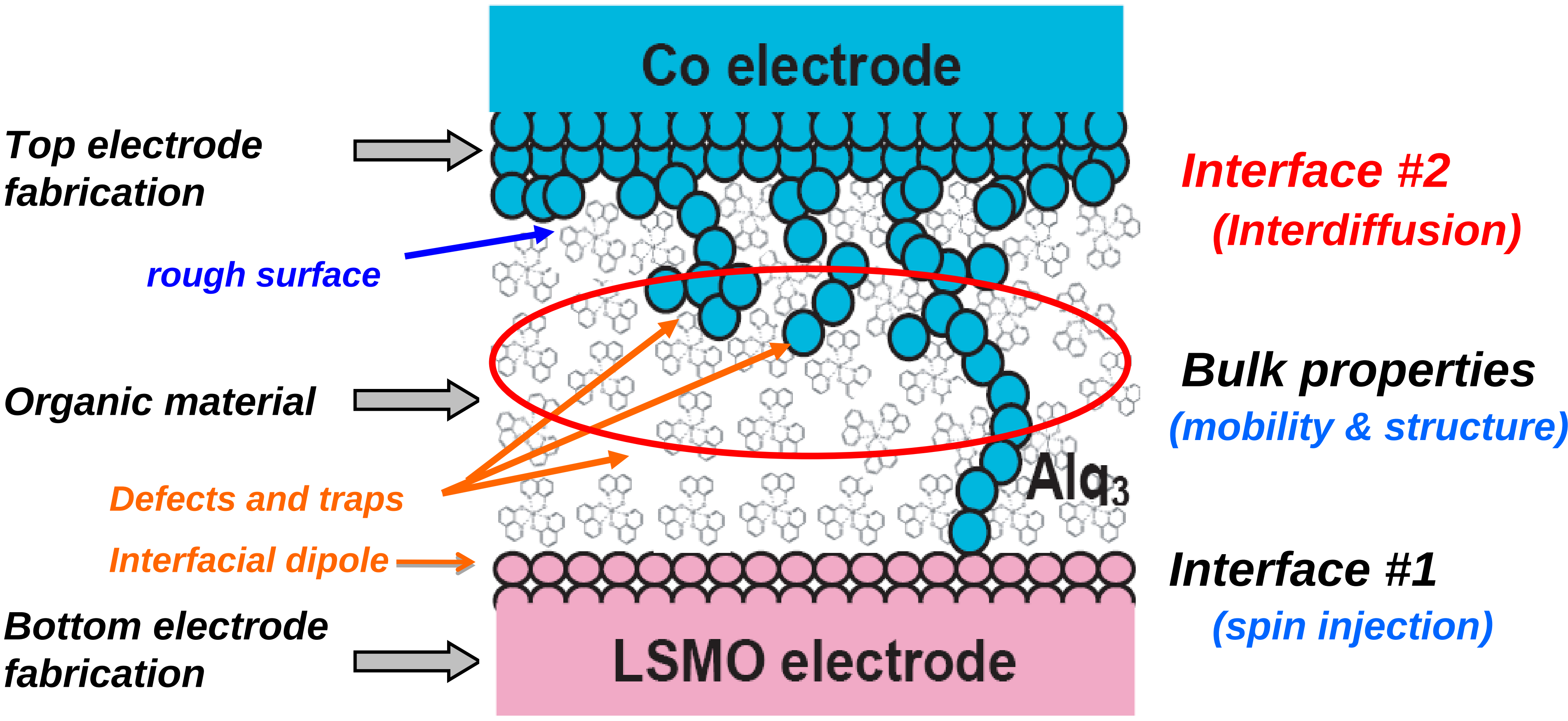
$$J_{\uparrow\uparrow(\uparrow\downarrow)} = D_{\uparrow\uparrow(\uparrow\downarrow)}\varepsilon\varepsilon_0\frac{V^2}{d^3}\exp\left(\frac{3.74}{kTd}\frac{eV\lambda_{\uparrow\uparrow(\uparrow\downarrow)}}{\left(1+4.2\lambda_{\uparrow\uparrow(\uparrow\downarrow)}\sqrt{\pi\varepsilon\varepsilon_0 V/ed}\right)}\right)$$

Only two fitting parameters, spin-dependent carrier injection density D, and screening length λ

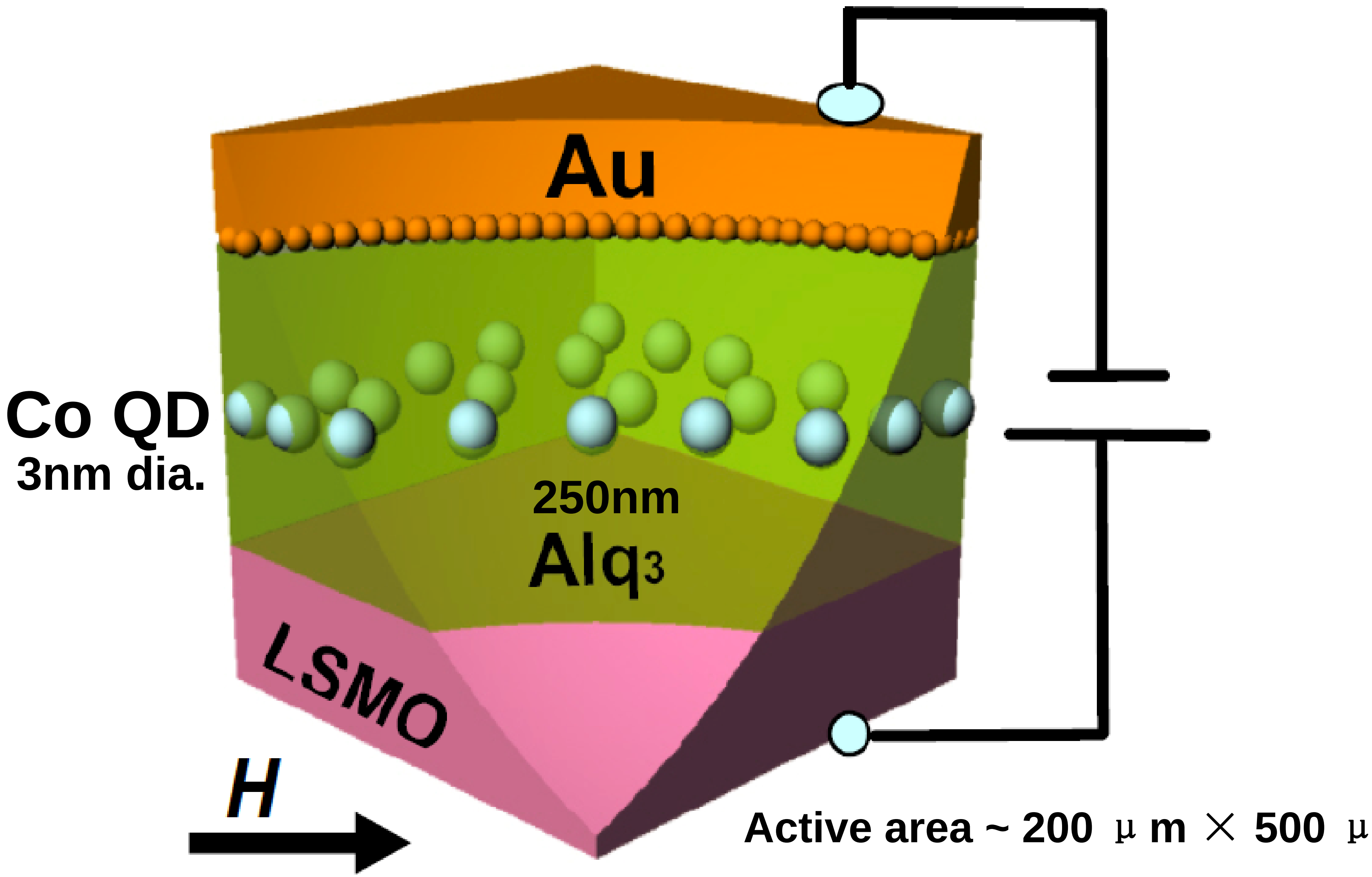
d (nm)	$D_{\uparrow\uparrow}$	$N_{tot(\uparrow\uparrow)}$ (cm^{-3})	$D_{\uparrow\downarrow}$	$N_{tot(\uparrow\downarrow)}$ (cm^{-3})	$D_{\uparrow\downarrow}/D_{\uparrow\uparrow}$	*MR*
93 (BLAG)	9.1688E-19	1.45379E19	3.9472E-18	2.77563E19	4.31 →	300
93 (conv)	3.8752E-16	3.41038E19	8.3410E-16	5.90169E19	2.15 →	35
135 (BLAG)	2.0580E-15	4.25549E19	2.2035E-15	4.68603E19	1.07 →	13
135 (conv)	1.4661E-15	4.33485E19	1.4790E-15	4.37145E19	1.01 →	4

Spin-dependent carrier injection density correlates with MR

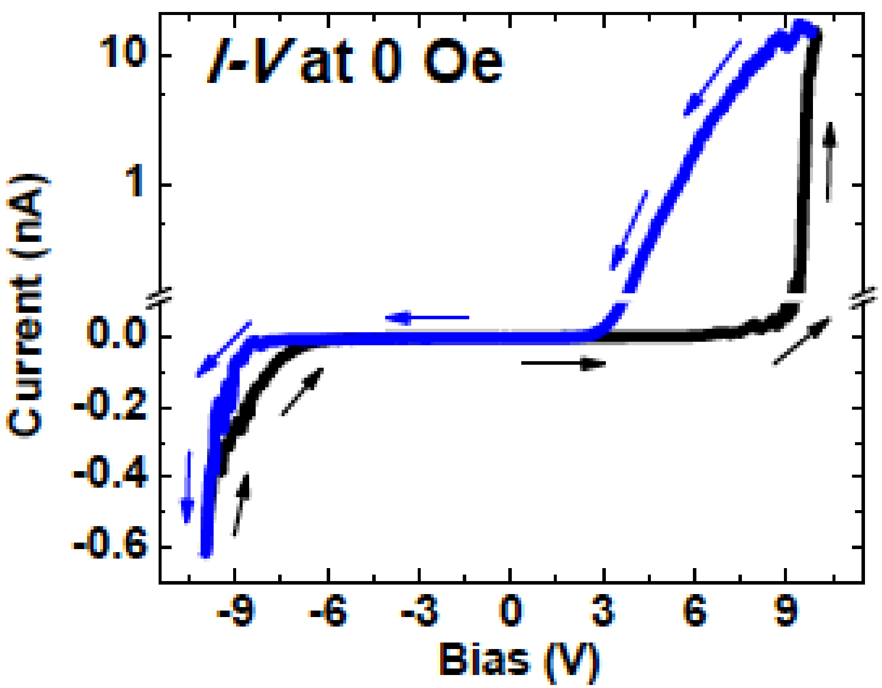
Part II

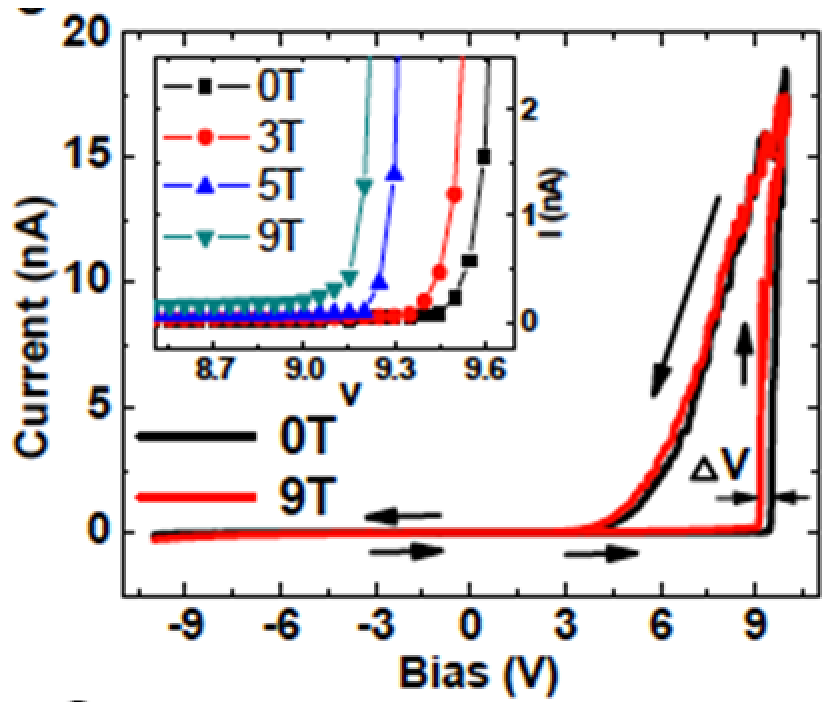

Co electrode
Top electrode fabrication
Interface #2
(Interdiffusion)
rough surface
Bulk properties
(mobility & structure)
Organic material
Defects and traps
Alq3
Interfacial dipole
Interface #1
(spin injection)
Bottom electrode fabrication
LSMO electrode

A New Avenue towards Colossal MR



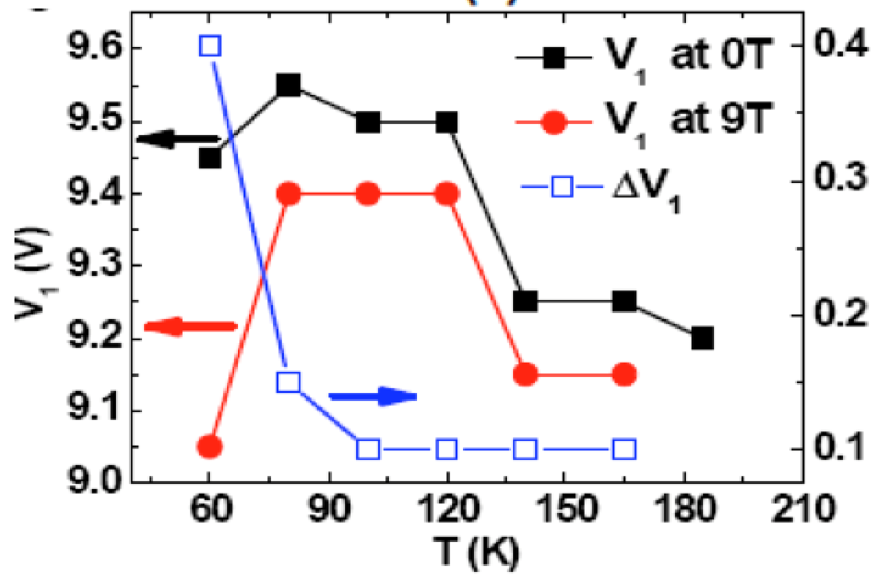
Transport Measurements





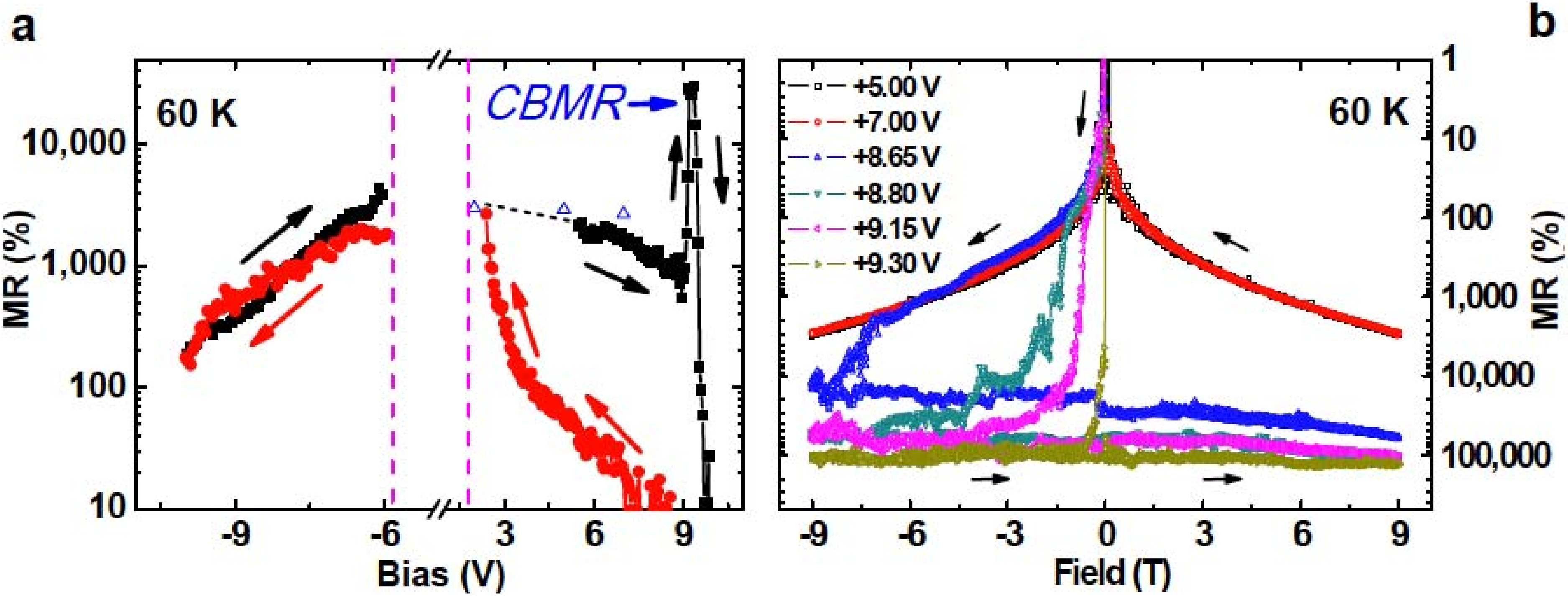
$$V_1 = eNL / 2\varepsilon\varepsilon_0$$

$$N = 3 \times 10^{10} / m^2, \ \varepsilon = 3, \ L = 100nm$$

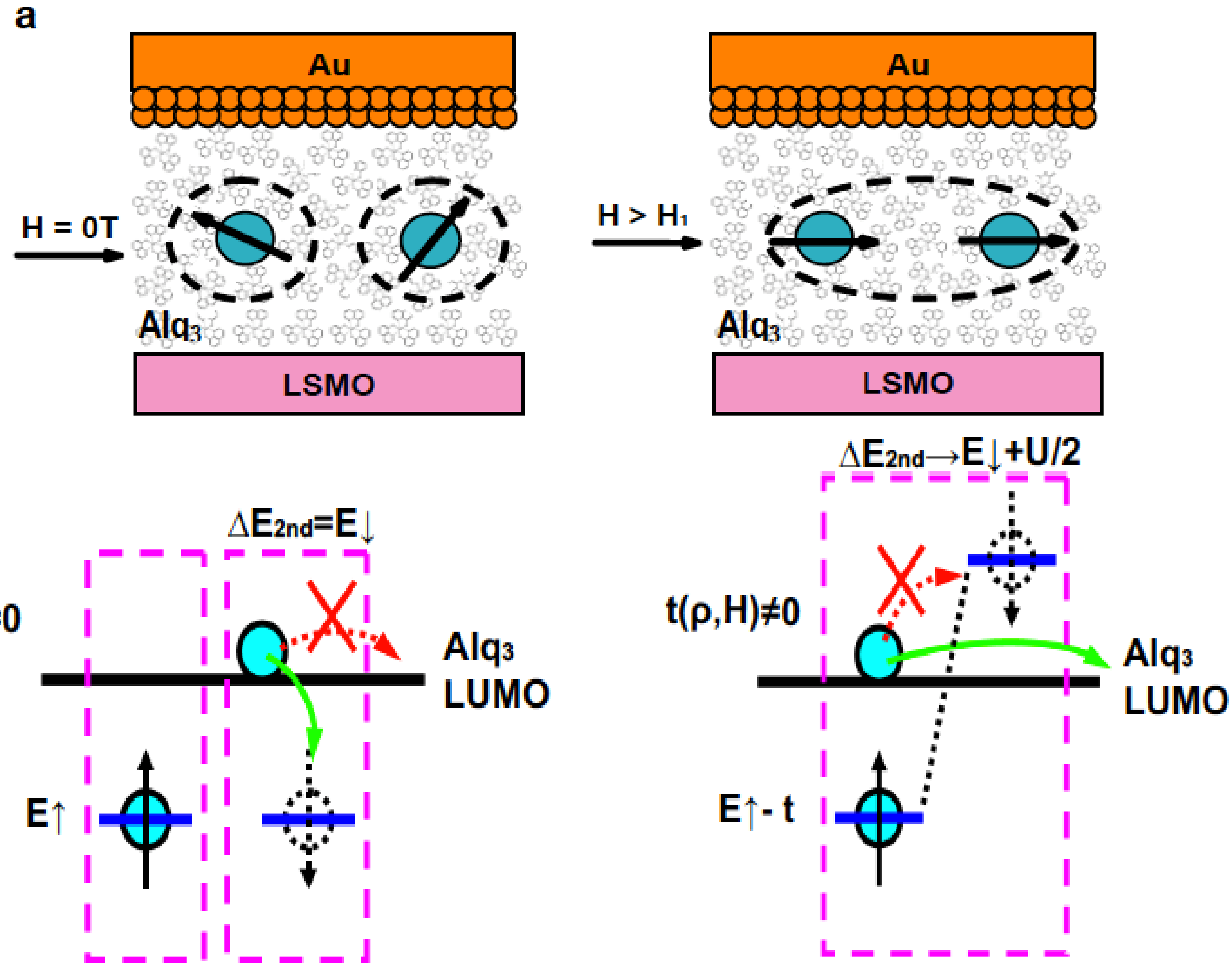


Colossal MR is expected

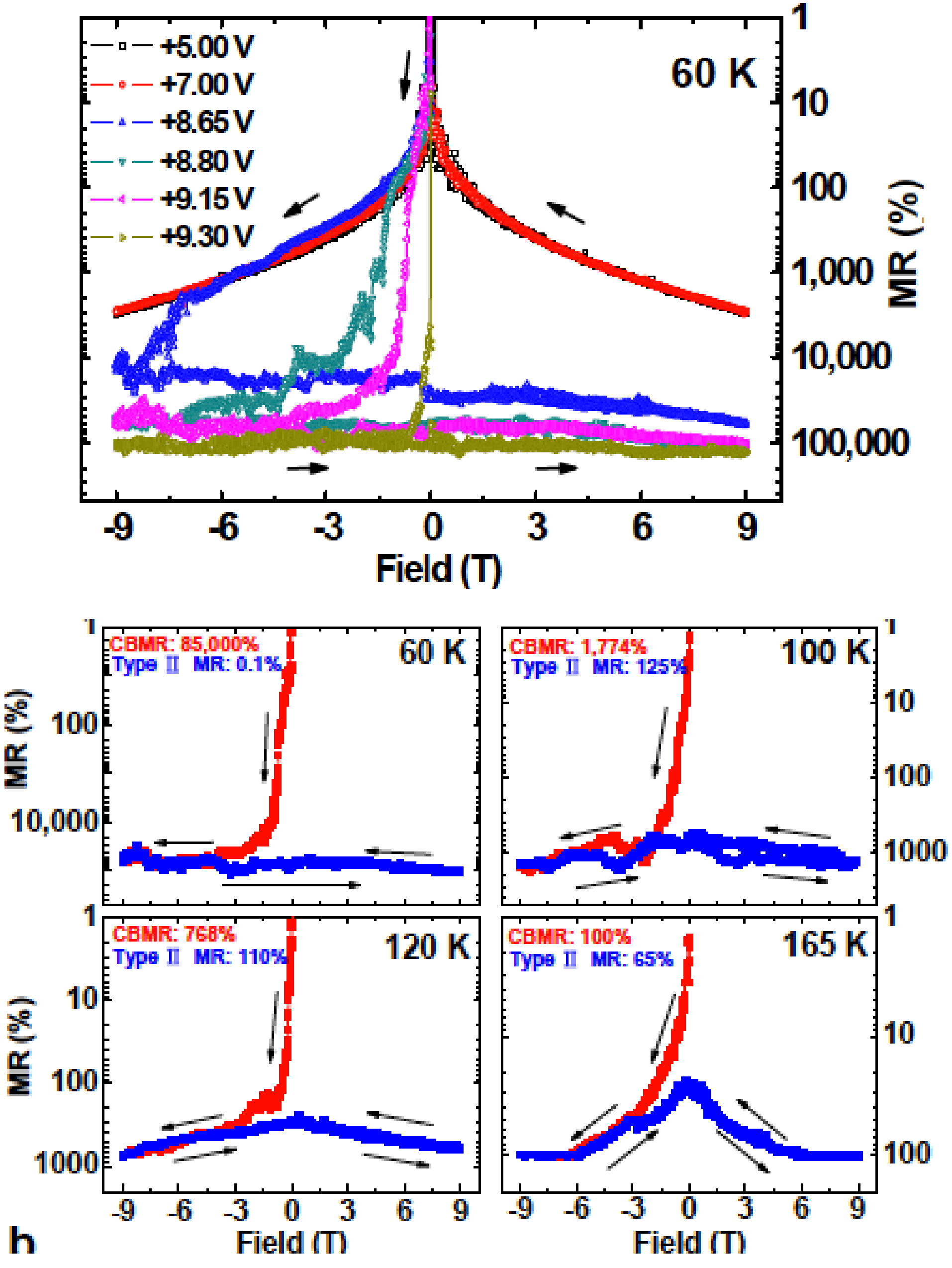
Nanodots induced Colossal MR
(~ 85000%)

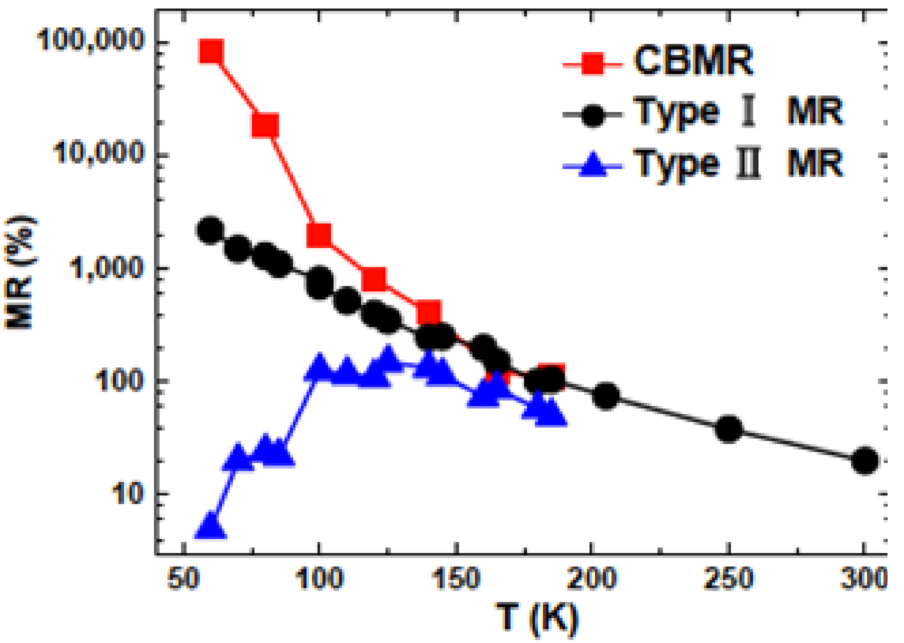


MR=(R(H)-R(0))/R(H)


a
Au
H = 0T
Alq3
LSMO
Au
H > H1
Alq3
LSMO
ΔE2nd→E↓+U/2
ΔE2nd=E↓
t=0
Alq3
LUMO
E↑
t(ρ,H)≠0
Alq3
LUMO
E↑- t

Three Types of MR





Part III

Top electrode fabrication

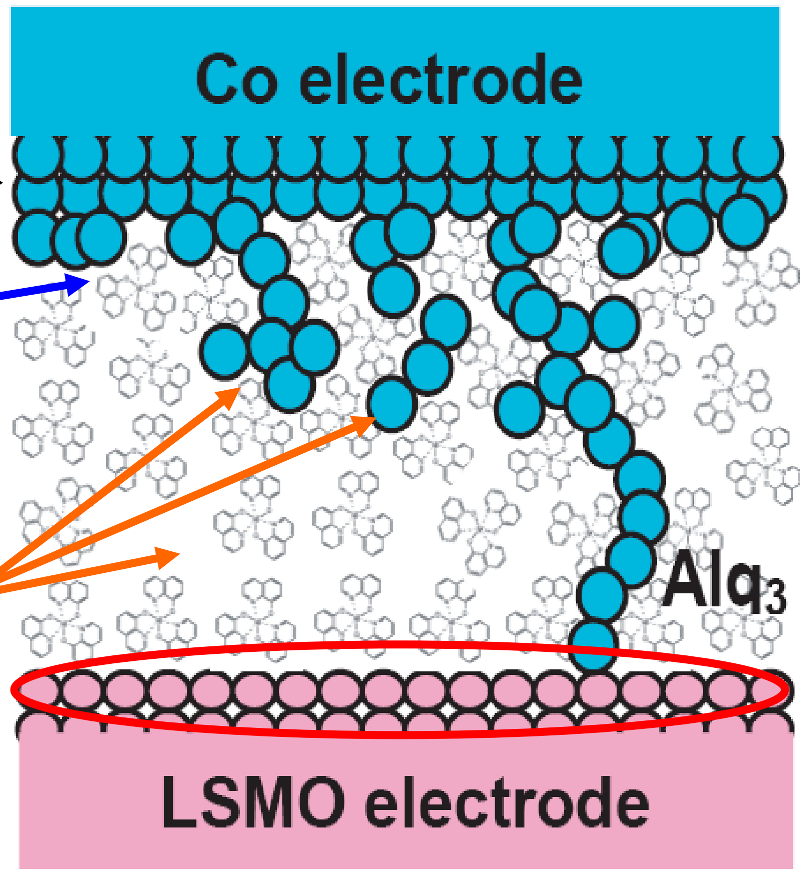
rough surface

Organic material

Defects and traps

Interfacial dipole

Bottom electrode fabrication

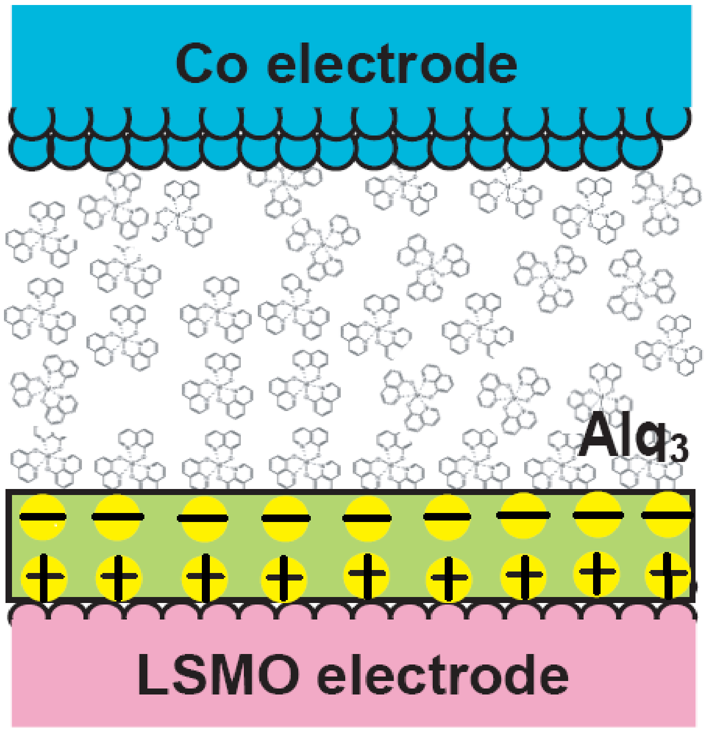


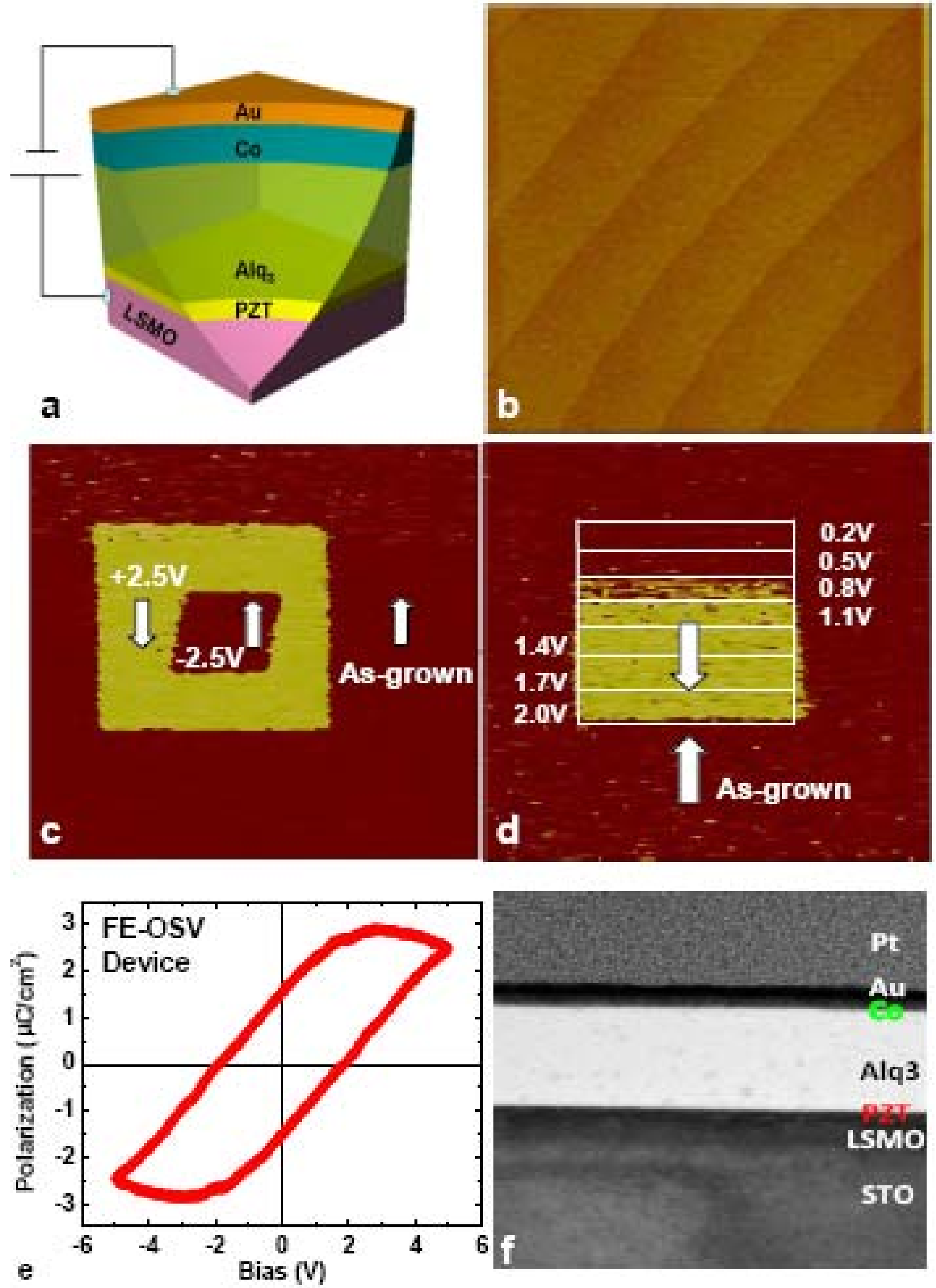
Interface #2
(Interdiffusion)

Bulk properties
(mobility & structure)

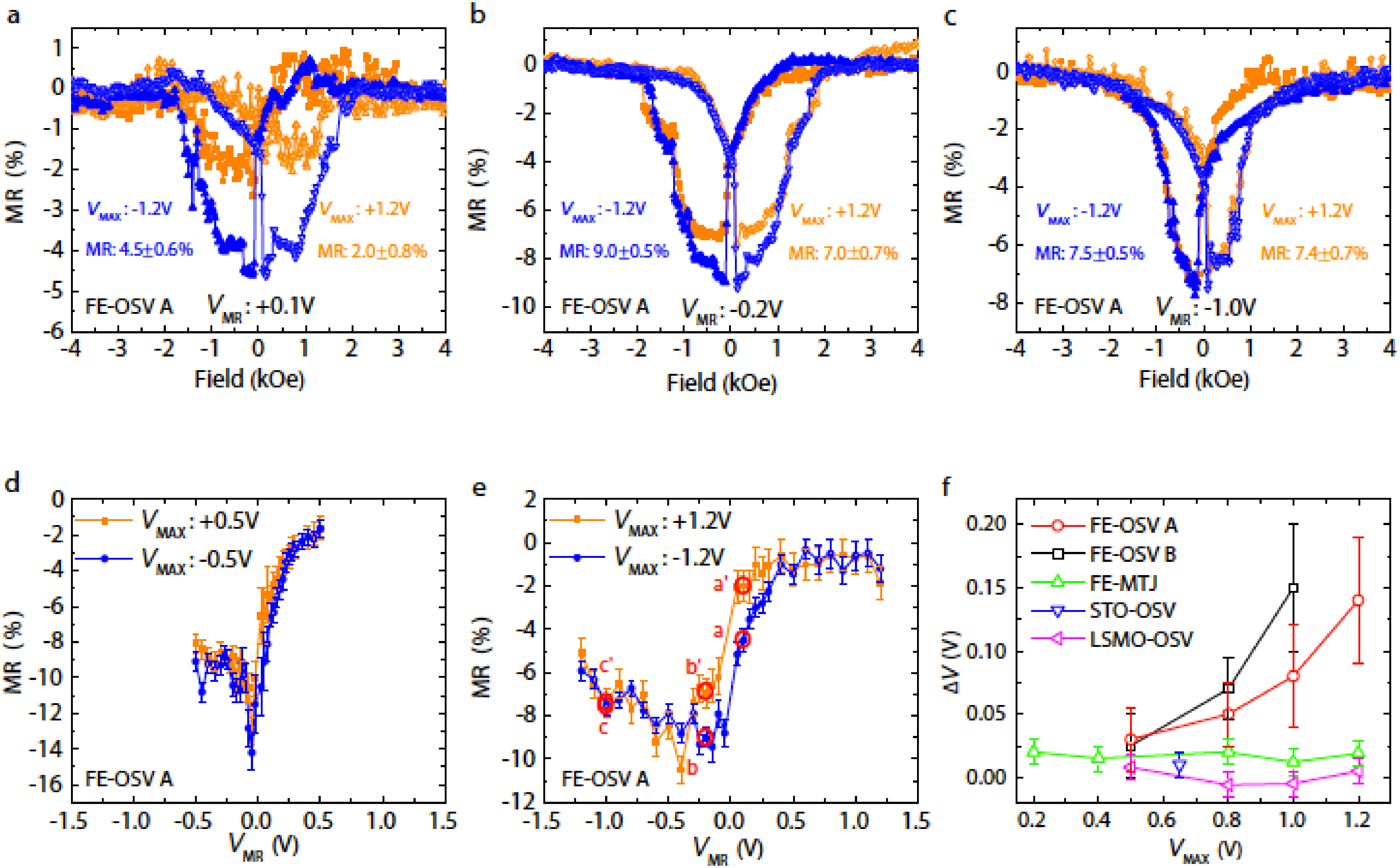
Interface #1
(spin injection)

Interfacial Control by Ferroelectricity

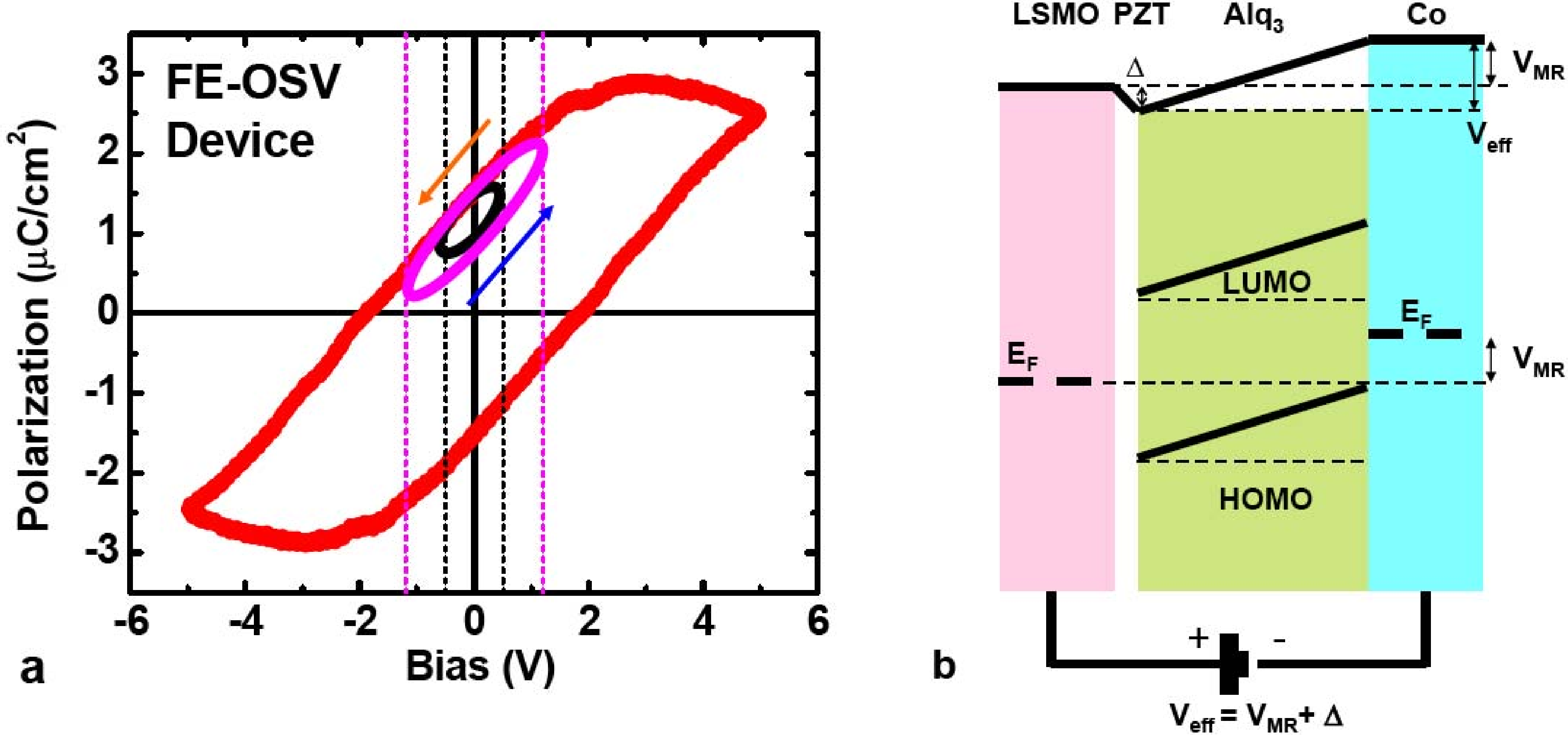




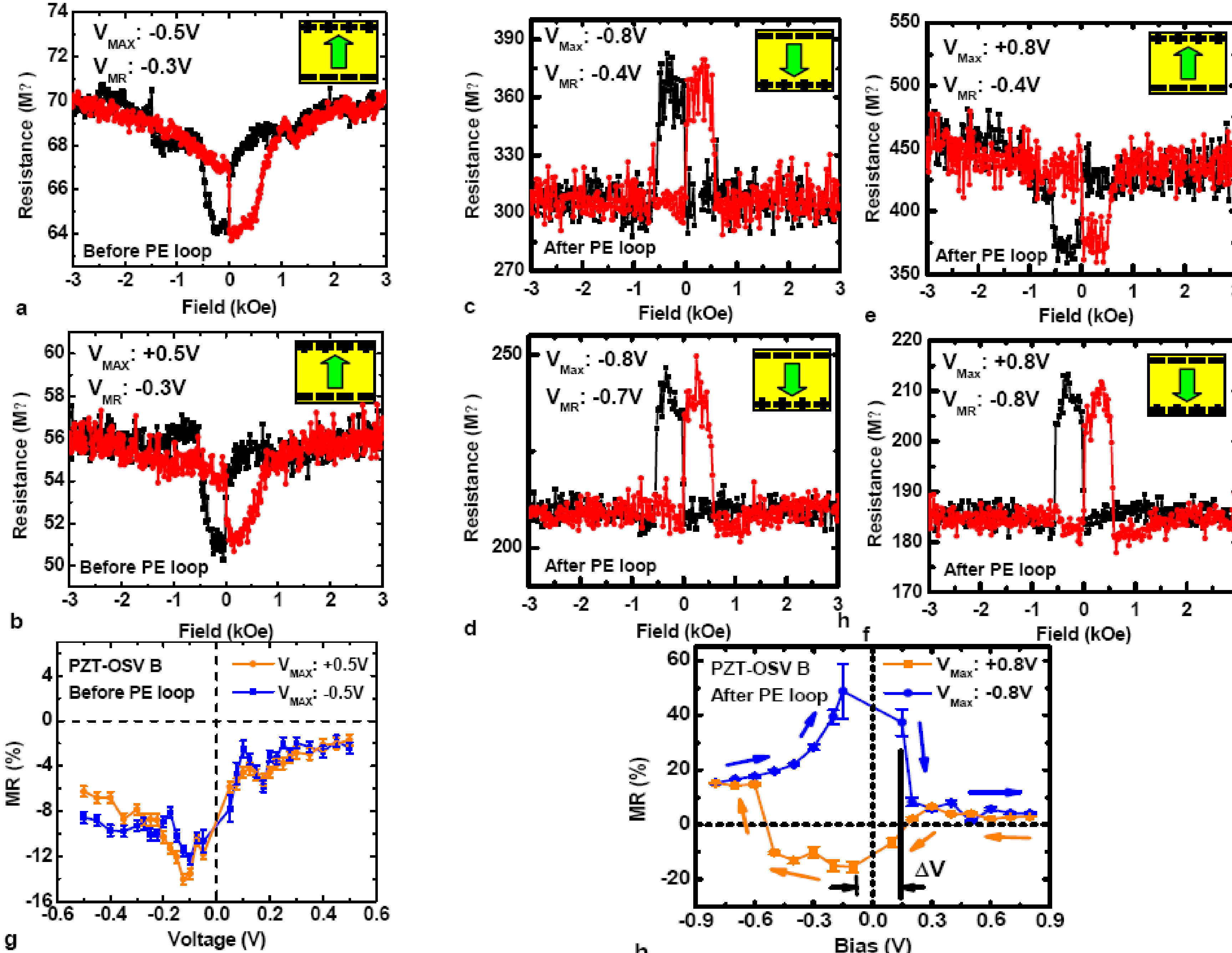
Hysteretic Behavior of MR

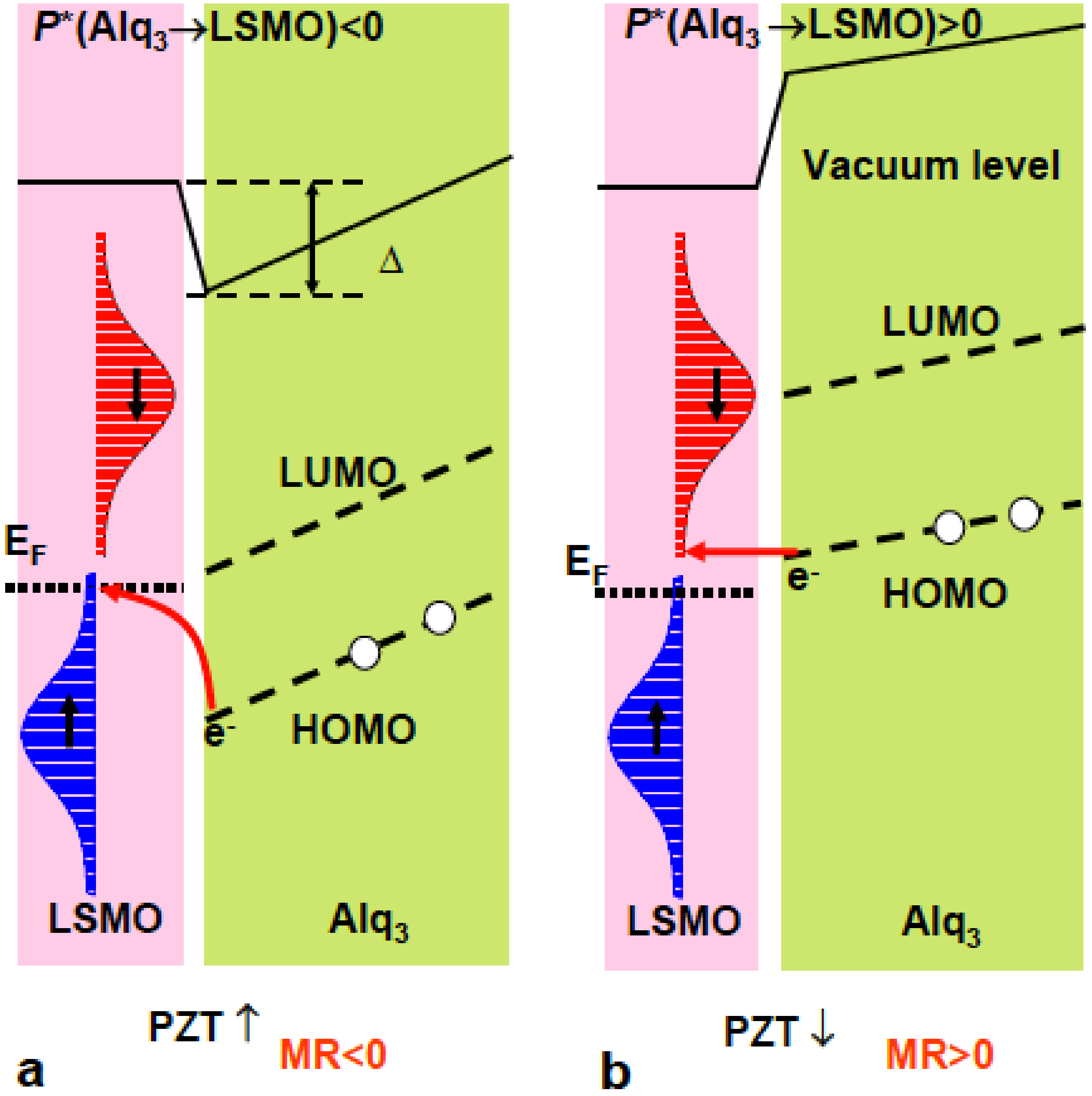


MR-V Hysteresis Originates from the FE Minor Loop



MR Sign Reversal After PE Loop





Nature Comm 4, 4396 (2014)

Summary

For vertical organic spin valves:

• On top of the organic film, magnetic nanodots can serve as top electrodes to minimize interdiffusion and lead to giant MR

• Inside the organic film, magnetic nanodots can be used to create spin-dependent resonant tunneling effect and lead to colossal magnetoresistance

• Underneath the organic film, inserting a ferroelectric layer can control the sign of MR

Acknowledgement

Da-li Sun **Mei Fang** **Xiaoguang Zhang**

Lifeng Yin (Fudan Univ)
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Wenting Yang (Fudan Univ)