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Spin tunnel and Spin Polarisation

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Summary

I- Introduction to Tunnel Effect

II-Magnetic Tunnel Effect

III-Bias Voltage and Temp Dependence

IV-Spin Polarisation

V- Half Metals







I- Introduction to Tunnel Effect





Tunnel Effect has a Quantum Mechanics Origin



A classical electron with energy $E < E_0$ cannot enter the barrier zone However a quantum electron obeys the Schrödinger equation !

(1D model)

$$-\frac{\hbar^2}{2m}\frac{d^2}{dx^2}|\psi\rangle + V(x)|\psi\rangle = E|\psi\rangle$$

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Off the barrier

$$-\frac{\hbar^2}{2m}\frac{d^2}{dx^2}|\psi\rangle = E|\psi\rangle$$

Plane waves

$$|\psi\rangle = e^{i(kr - \omega t)}$$
 and $k = \pm \sqrt{\frac{2mE}{\hbar^2}}$

In the barrier

$$-\frac{\hbar^2}{2m}\frac{d}{dx^2}|\psi\rangle = (E - E_0)|\psi\rangle \quad \text{and} \quad E - E_0 < 0$$

Evanescent waves

$$\left|\psi\right\rangle_{b} = e^{qr - i\omega t}$$
 and $q = \pm \sqrt{\frac{2m\Delta E}{\hbar^{2}}}$





$$q = \pm \sqrt{\frac{2m\Delta E}{\hbar^2}}$$

$$\Delta E = 1eV$$

 $m = free \quad electron \Rightarrow \frac{1}{q} = 0.2 \ nm$

Tunnel barriers must be very thin insulating layers Width = w < 10 nm





For a more general barrier shape



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What is neglected ?

$$\psi(x) = e^{\pm \int_{0}^{x} k(u) du}$$
$$\frac{d\psi(x)}{dx} = \pm k(x)\psi(x)$$

$$\frac{d^2\psi(x)}{dx^2} = \pm \frac{dk(x)}{dx}\psi(x) \pm k(x)\frac{d\psi(x)}{dx}$$
$$= \pm \frac{dk(x)}{dx}\psi(x) + k^2(x)\psi(x)$$

 $k(x) = \sqrt{\frac{2m}{\hbar^2}} (V(x) - E)$ The barrier potential should vary smoothly





We are dealing with transport. What about the current ?

To pass a current, we must apply a bias voltage across the barrier.



the barrier has a transmission coefficient T







Simmons

The probability to find the electron (energy E) on the other side of the barrier is :

$$P(E) = \left\langle \psi \left| \psi \right\rangle = \psi^{2} = e^{-2 \int_{0}^{n} \sqrt{\frac{2m(V(u) - E)}{\hbar^{2}}} du}$$

Electrons coming from the left to the right

$$dN = \int_{0}^{\infty} V_{x} n(V_{x}) P(E_{x}) dV_{x} dt$$

$$\frac{dN}{dt} = \frac{4\pi m^2}{h^3} \int_{0}^{E} P(E_x) dE_x \int_{0}^{\infty} f(E) dE_{//}$$



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No bias voltage

Current $1 \rightarrow 2 = \text{Current } 2 \rightarrow 1$

J(V=0)=0







$$J = e(\frac{dN_1}{dt} - \frac{dN_2}{dt}) = \frac{4e\pi m^2}{h^3} \int_0^E P(E_x) dE_x \int_0^\infty [f(E) - f(E + eV)] dE_{//}$$

Simmons (1963) has calculated approximate recipes

 $J = 3.16 \ 10^{10} \sqrt{E_0} \frac{V}{S} e^{-1.02S\sqrt{E_0}}$

Linear J(V) at low bias







$$J = \alpha (V + \beta V^3)$$

Simmons ' parabolic fit

$$\frac{dJ}{dV} = \alpha (1 + 3\beta V^2)$$

 β contains the barrier height and the barrier width





Why
$$\frac{dJ}{dV}$$
 ?

From the experimental point of view :

Using a voltage source : $V(t) = V_0 + v \cos(\omega t)$ $v \ll V_0$

$$J(V) = J(V_0 + v\cos(\omega t)) = J(V_0) + v\cos(\omega t)\frac{dJ}{dV}(V_0)$$

Measuring the ω component of the signal with a lock-in amplifier gives directly the differential conductance Filter all the constant voltage and non- ω noise







J increases rapidly but in fact such a bias voltage corresponds to the electrical breakdown regime









I-V non linear curves (ferromagnetic/insulator/ferromagnetic)



No temperature dependence of tunnel effect (1st order)

Thèse E. Favre-Nicolin (Grenoble 2003)





II-Magnetic Tunnel Effect





In 1972 Gittleman et al. measured the resistance and MR of Ni grains in a SiO₂ matrix Longitudinal MR is <0 contrary to the sign of the bulk Ni AMR $\rho_{//} > \rho_{perp}$







Gittleman et al. 1972 :

than the total resistivity of nickel. Accordingly we conclude that the electronic tunneling probability t and ρ_t are magnetic-field dependent. Such a field dependence of the tunneling probability can arise from the fact that as an electron tunnels into a neighboring grain its spin must be rotated whenever the moments of the grains are not aligned. This

$$\sigma(T,H) = \sigma_0(T) + \sigma_1(T) \left\langle \frac{\vec{m}_1 \cdot \vec{m}_2}{m^2} \right\rangle$$





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8 September 1975

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TUNNELING BETWEEN FERROMAGNETIC FILMS

M. JULLIERE

Institut National des Sciences Appliquées. 35031 Rennes Cedex, France

Received 25 June 1975

Fe-Ge-Co junctions conductance G(V) is studied when mean magnetizations of the two ferromagnetic film are parallel or antiparallel. Conductance measurement, in these two cases, is related to the spin polarizations of the conduction electrons.

Development of film deposition techniques Trilayer : better characterisation of electrodes and control of magnetisation

R changes by 14% at low temperature depending on the magnetic configuration





Large Magnetoresistance at Room Temperature in Ferromagnetic Thin Film Tunnel Junctions

J. S. Moodera, Lisa R. Kinder, Terrilyn M. Wong, and R. Meservey

Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139



FIG. 2. Resistance of CoFe/Al₂O₃/Co junction plotted as a function of H in the film plane, at 295 K. Also shown is the variation in the CoFe and Co film resistance. The arrows indicate the direction of M in the two films (see text).



 $200 \mu m \ge 300 \mu m$

11.8% at 300 K 24% at 24 K φ=1.9 eV and t=1.6 nm

V_{50%}=200mV

And also Miyazaki, Tezuka JMMM 139 (1995) L231





Magnetic Tunnel Junction



Same technical solutions as GMR structures to get 2 different coercive fields i.e. well defined parallel and antiparallel states.

Hard - Soft materials (Co - NiFe)Different shape anisotropies for both electrodesPinning to AF layer (MnFe) or Artificial AF layer (Co/Ru/Co)





Jullière 's model (1975)

Not magnetisation BUT Polarisation of electrodes is the parameter

$$P = \frac{N_{\uparrow}(E_F) - N_{\downarrow}(E_F)}{N_{\uparrow}(E_F) + N_{\downarrow}(E_F)}$$

$$N_{i\uparrow} = \frac{N_i(1+P_i)}{2} \qquad N_{i\downarrow} = \frac{N_i(1-P_i)}{2}$$

Assume : No spin-flip transition across the barrier at low voltage
2 parallel channels (spin up and spin down)
Conductance is the sum of spin up and down conductances
Conductance is proportional to the density of state (d.o.s.) 1 and d.o.s. 2

$$G_{spini} = G_0 N_{spinielectrode1} (E_F) N_{spinielectrode2} (E_F)$$

$$G_{\uparrow\uparrow} = G_0 N_{1\uparrow}(E_F) N_{2\uparrow}(E_F) + G_0 N_{1\downarrow}(E_F) N_{2\downarrow}(E_F)$$

 $G_{\uparrow\downarrow} = G_0 N_{1\uparrow}(E_F) N_{2\downarrow}(E_F) + G_0 N_{1\downarrow}(E_F) N_{2\uparrow}(E_F)$





Jullière 's model (M. Jullière, Phys. Lett. 54 A, 225 (1975))

TMR ratio : $G_{\uparrow\downarrow} + G_{\uparrow\uparrow} = G_0 N_1 N_2$ $G_{\uparrow\uparrow} - G_{\uparrow\downarrow} = G_0 N_{1\uparrow} N_2 P_2 - G_0 N_{1\downarrow} N_2 P_2 = G_0 N_1 P_1 N_2 P_2$ or $\frac{G_{\uparrow\uparrow} - G_{\uparrow\downarrow}}{G_{\uparrow\uparrow}} = \frac{2P_1P_2}{1 + P_1P_2}$ $\frac{G_{\uparrow\uparrow} - G_{\uparrow\downarrow}}{G_{\uparrow\uparrow} + G_{\uparrow\downarrow}} = P_1 P_2$ $\frac{R_{\uparrow\uparrow} - R_{\uparrow\downarrow}}{R_{\uparrow\uparrow}} = \frac{-2P_1P_2}{1 - P_1P_2}$ (pick your definition) Does depend on P_i Does not depend on the barrier (height, width) because of assumption about G_0 i.e. no spin dependence of transmission





Exp: TMR=14% Theor: $P_{Co}34\% + P_{Fe}44\%$ TMR 26%



CoFe TMR junction (Tohoku 2000)





Slonczewski 's model (1989)



Solve Schrödinger for both channels, calculate conductances

$$P = \frac{q^2 - k_{F\uparrow} k_{F\downarrow}}{q^2 + k_{F\uparrow} k_{F\downarrow}} \frac{k_{F\uparrow} - k_{F\downarrow}}{k_{F\uparrow} + k_{F\downarrow}}$$





$$P = \frac{q^2 - k_{F\uparrow} k_{F\downarrow}}{q^2 + k_{F\uparrow} k_{F\downarrow}} \frac{k_{F\uparrow} - k_{F\downarrow}}{k_{F\uparrow} + k_{F\downarrow}} \qquad q = \pm \sqrt{\frac{2m\Delta E}{\hbar^2}}$$

High barrier
$$P = \frac{q^2}{q^2} \cdot \frac{k_{F\uparrow} - k_{F\downarrow}}{k_{F\uparrow} + k_{F\downarrow}} = \frac{k_{F\uparrow} - k_{F\downarrow}}{k_{F\uparrow} + k_{F\downarrow}}$$

$$DOS(E) = \frac{m}{\hbar^3 \pi^2} \sqrt{2mE} = \frac{mk}{\hbar^2 \pi^2} \propto k$$

$$P = \frac{N_{\uparrow}(E_F) - N_{\downarrow}(E_F)}{N_{\uparrow}(E_F) + N_{\downarrow}(E_F)}$$

Back to Jullière 's formula





Improved models :

Bratkovsky : Correction to Slonczewski's model (different effective mass in the barrier)

$$P = \frac{q^2 - m_b^2 \cdot k_{F\uparrow} k_{F\downarrow}}{q^2 + m_b^2 \cdot k_{F\uparrow} k_{F\downarrow}} \frac{k_{F\uparrow} - k_{F\downarrow}}{k_{F\uparrow} + k_{F\downarrow}} \qquad q = \pm \sqrt{\frac{2m_b \Delta E}{\hbar^2}}$$

 $m_b/m=0.4$ for Fe/Al₂O₃

Ab initio band structure calculations :

to get the band structure close to the interface







III-Bias and Temp Dependence







Miyazaki group (Tohoku) APL 2000

TMR temperature dependence

Resistance Temperature dependence





Temperature dependence of resistance





Temperature dependence of the barrier transmission Going from 0 Kelvin to 300 Kelvin



Wavevector in the barrier (evanescent wave)



Conductance \propto Transmission Transmission = $T \propto e^{-2qw}$







Transmission =
$$T \propto e^{-2qw}$$

$$\frac{dT}{T} = d(-2qw) = -2w \cdot dq = -wq \frac{d\Delta E}{\Delta E}$$

$$q = 1 \text{ A} , w = 1 \text{ nm},$$

$$\Delta E = 2eV, d\Delta E = kT = 25 \text{ meV}$$

$$\frac{\Delta T}{T} = \frac{\Delta G}{G} = \frac{\Delta R}{R} = 12.5\%$$





Temperature dependence of TMR





Different contributions may rule this behaviour :

Polarisation is related to magnetisation

$$M(T) = M_{s}(T)(1 - \alpha T^{3/2})$$

 $P(T) = P_s(T)(1 - \alpha T^{3/2})$

Inelastic processes can appear

$$e^{\uparrow} \Rightarrow e^{\downarrow} + magnon$$
$$e^{\uparrow} + magnon \Rightarrow e^{\downarrow}$$

Opens a spin-flip conductance channel conductance increases TMR decreases

Surface magnetisation is less robust to thermal fluctuations

No general results, depends on the studied system (T_c , surface state ...)




Voltage dependence of TMR









50% MR at 3mV

Jullière, Phys. Lett. 1975







TMR - bias voltage dependence 50% decrease TMR for 400mV

R - bias voltage dependence





At large bias voltages, hot electrons are introduced in the second electrode : 0.1 V = 1200 Kelvin

Inelastic processes can be activated

 $e^{\uparrow} \Rightarrow e^{\downarrow} + magnon$ $e^{\uparrow} + magnon \Rightarrow e^{\downarrow}$

Opens a spin-flip conductance channel TMR decreases with V

The voltage decrease depends on experimental systems and years

 $\begin{array}{l} 1975:{\rm TMR}_{50\%}{=}2\;{\rm mV}\\ 1995:{\rm TMR}_{50\%}{=}200\;{\rm mV}\\ 2000:{\rm TMR}_{50\%}{=}450\;{\rm mV}\\ 2003:{\rm TMR}_{50\%}{>}1000\;{\rm mV} \end{array}$

May be due to non perfect samples, which improve with time







Epitaxial NiFe electrode : 50 % decrease of TMR at 750 mV

Yu et al. APL 2003





Tunnel junction with « perfect barrier » : STM with ferromagnetic electrodes in vacuum

Magnetic amorphous tip



Cobalt (0001)



FIG. 1. Tunnel magnetoresistance δ and its error of a clean Co(0001) surface vs bias voltage U, obtained with a magnetic tip stabilized at 1 V, 1 nA (a) and at 100 mV, 1 nA (b).

Ding et al. (M.P.I. Halle) PRL 2003

Voltage dependence of TMR not related to magnon excitations or surface magnetisation but more likely to defects in the barrier to be confirmed ...





A few words about

The insulating barrier





Making the barrier

Good recipe #1

Aluminium Film (0.7-2 nm) + thermal oxidation in oxygen atmosphere or air

Good recipe #2

Aluminium Film + oxygen plasma

Bad recipe #1

Deposition of Alumina (Al_2O_3) produces a less dense barrier with poor electrical properties















The barrier should be uniform

- flat
- 0 roughness
- fully oxidised, homogeneously
- no oxidation of the ferromagnetic metals



1 Volt across a 1 nm barrier is E = 1 GigaV/m

This is the order of magnitude of the electrical field necessary to ionise an atom

A 0.1 nm fluctuation means 100 mV decrease of the breakdown voltage





Formation enthalpy ΔH (298K) kJ/mol metallic atom

Ta2O5	-1023	HfO2	-1144
Nb2O5	-949	ZrO2	-1100
		CeO2	-1088
Y2O3	-952	TiO2	-944
Gd2O3	-909	SiO2	-910
Nd2O3	-903	NbO2	-796
La2O3	-896	CrO2	-598
Al2O3	-837	MnO2	-520
V2O3	-609		
Cr2O3	-570	MgO	-601
Ga2O3	-544	NbO	-405
Mn2O3	-479	MnO	-385
Fe2O3	-412	ZnO	-350
		FeO	-272
Mn3O4	-462	NiO	-238
Fe3O4	-372	CoO	-237
Co3O4	-297	CuO	-157
		SiO	-99
		Ga2O	-178
		Cu2O	-84

Al-O bond is stronger than 3d metal -O bonds

Better oxides exist : HfO₂, Rare earth-O

But

kinetics : (Al passivated « automatically » 1 nm thick),

local state : (amorphous, nanocrystallised, crystallised) when RT deposition





Crystallised barrier Fe Fe MgO HIO2 Co Co

Amorphous barrier



Smith et al. JAP,83,5154 (1998)

FIG. 3. High-resolution, cross-sectional electron micrograph of a [50 nm Co/10 nm HfO₂/50 nm Fe] junction showing abrupt, smooth interfaces and amorphous oxide structure.





Role of annealing a tunnel junction

NiFe(100 Å)/CoFe(20 Å)/Al2O3/CoFe(40 Å)/MnRh(170 Å)



FIG. 1. Tunneling magnetoresistance vs field for an as-deposited spin tunnel junction, and for the same junction after consecutive anneals up to $230 \,^{\circ}$ C. In the inset, the four-probe measuring scheme is illustrated using an optical microscope picture of the junction.

Barrier (Simmons' fit)

after anneal : thickness :0.87 to 0.77 nm wide barrier height :1.8 eV to 2.5 eV





RBS measurement of Al and O distributions



O from the CoFe electrodes goes back to Al_2O_3







Best present TMR junctions : 50% Room temperature (non exotic materials)



Ta (5 nm)/ Ni₇₉Fe₂₁ (3 nm)/ Cu (20 nm)/ Ni₇₉Fe₂₁(3 nm)/ Ir₂₂Mn₇₈(10nm)/ Co₇₅Fe₂₅(4 nm)/ Al (0.8 nm)-oxide/ $Co_{75}Fe_{25}(4 nm)/$ Ni₇₉Fe₂₁(20 nm)/ Ta (5nm)

Pinned layer

Free layer





IV-Spin polarisation





We have been using values for P.

Where do they come from ?

$$P = \frac{N_{\uparrow}(E_F) - N_{\downarrow}(E_F)}{N_{\uparrow}(E_F) + N_{\downarrow}(E_F)}$$

How to measure them ?





Density of States Band structure calculations (spin resolved)



Moroni et al. PRB56 (1997)15629







Nickel integrated density of states

Polarisation at Fermi level should be NEGATIVE







Be careful (M. B. Stearns JMMM, 5, 167 (1977))

Bands do not have the same effective mass at E_F

Electrons at Fermi level have different mobilities

d-like electrons are more localised (narrow bands) s-like electrons are less localised (wide bands) and more mobile





 $\frac{\hbar^2}{\partial^2 E}$ $\frac{\partial k^2}{\partial k^2}$

m







Be careful (M. B. Stearns JMMM,5 ,167 (1977)) Bands do not have the same effective mass at E_F

Electrons at Fermi level have different mobilities

d-like electrons are more localised (narrow bands) s-like electrons are less localised (wide bands) and more mobile

Ferromagnetism comes from the 3d bands but transport comes from s-electrons. s electrons are not supposed to be polarised !

Difficult to predict the polarisation of conduction electrons



 $\frac{\hbar^2}{\partial^2 E} \frac{\partial^2 E}{\partial k^2}$

 \boldsymbol{m}

When it is difficult to predict, let us measure !

Ferromagnetic / insulating / superconducting junctions M. I. T. speciality (Tedrow/Meservey)

$$J \propto \int_{0}^{\infty} |M|^{2} N_{1}(E) N_{2}(E) [f(E) - f(E + eV)] dE$$

$$|M|^2 \propto e^{-2qw}$$

N(E) non magnetic metal

 $N(E) \propto \text{constant}$











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Meservey et al; J.A.P. 50 (1979)

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Andreev reflexion : metal / superconductor clean interface





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But it only works at 1 Kelvin ! Andreev Reflexion does not give the sign of P

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To analyse the conduction electrons : spin resolved photoemission



Very clean surface, UHV prepared surface



Spin-resolved photoemission

Example : Epitaxial Fe on Ag



Jonker et al. PRL57 (1986)

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Polarisation at 0K for Ni, Co and Fe is known

Its temperature dependence is still unclear

Surface polarisation still a mystery

(enough to keep you busy after Brasov school)

What about the other magnetic metals ?





V-Half Metallic Ferromagnets







Ferromagnetic band structure







Predicted HMF







Compound	Curie temperature	Magnetisation $\mu_o M_s$	Crystallographic structure
$La_{0.7}Sr_{0.3}MnO_3$	350 K	0.74 T $3.7 \mu_{\text{B}}/\text{Mn}$	Perovskite
NiMnSb	728 K	$0.89 \text{ T} 4 \mu_{\text{B}}/\text{Mn}$	Semi Heusler
CrO ₂	396 K	0.81T 2 $\mu_{\rm B}/{\rm Cr}$	Rutile
Fe ₃ O ₄	860 K	0.63 T 4 $\mu_{\rm B}$ /F.U.	Inverse Spinel
PtMnSb	572 K	0.9 T 4 $\mu_{\rm B}/{ m Mn}$	Semi Heusler
Sr ₂ FeMoO ₆	415 K	0.73 T 4 $\mu_{\rm B}/{\rm F.U.}$	Double perovskite







Predicted Half metallic character of La_{0.7}Sr_{0.3}MnO₃



Zero K picture

Spin-polarized band structure of $La_{0:7}Sr_{0:3}MnO_3$. The majority bands are shown as solid lines, and the minority as dashed lines

Livesay et al. J. Phys.: Condens. Matter 11 (1999) L279–L285







HMF are a very interesting class of materials

Useful to study HMF state

Are they really 100% polarised ?
Up to T_c ?
Is the surface polarised ?

Useful to study other materials

Source of d-like electrons Highly spin polarised source







DeTeresa et al. PRL 82(1999)

Inverse TMR

Cobalt : positive P (previous studies) LaSrMnO : positive P (no down bands)

and negative TMR !

d electrons tunnel





Conclusions :

Tunnel is old but TMR is quite a recent field Spin-dependent transport properties are not fully understood HMF is still a mystery

A lot of material science is necessary to control the structures Only a few materials and structures have been controled

Bringing together magnetism and electronics is a fruitful playground ...









Improved structures : resonant junctions









Ivo Sturm

Manouk Rijpstra



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FIG. 2. Transport studies for a Fe(001)/MgO 20 Å(001)/FeCo(001) tunnel junction of diameter 10 μ m: (a) resistance vs temperature at V=10 mV and $H_{app}=5000$ Oe; (b) resistance and TMR vs magnetic field at V=10 mV and T=30 K.

Bowen et al. APL 2001

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Epitaxial junctions