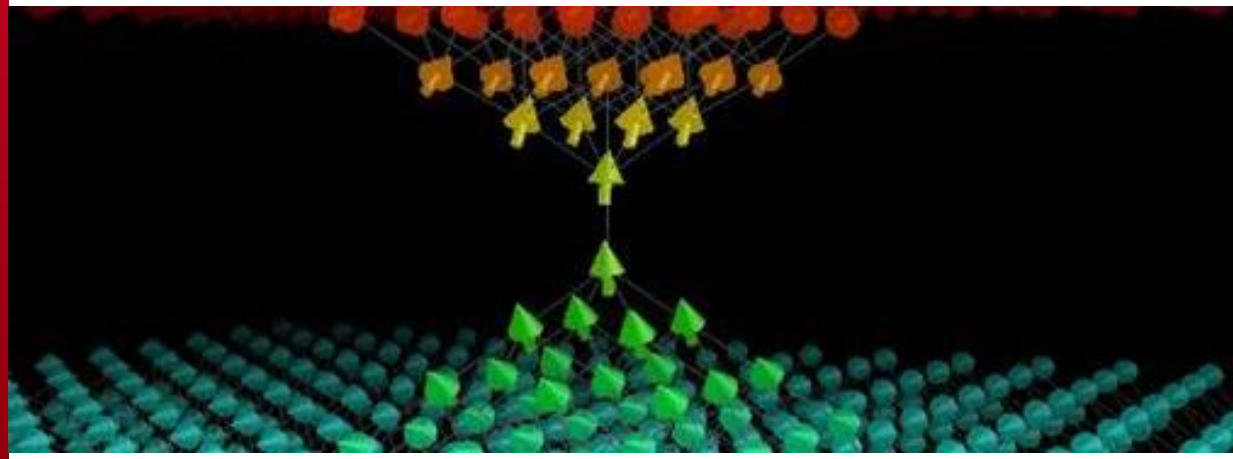
DE LA RECHERCHE À L'INDUSTRIE

SPIN-POLARIZED TRANSPORT AT THE NANOSCALE



Cyrille Barreteau

DTU Nantotech, Lyngby 24 May 2013

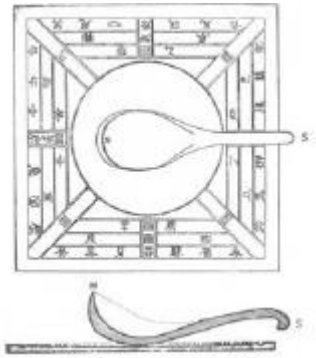
www.cea.fr

A BRIEF HISTORY

Magnetism is a very old science

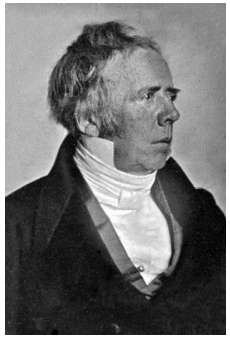
- The first magnetic devices

Chinese
South pointer (1080)



Portuguese Compass
15th century

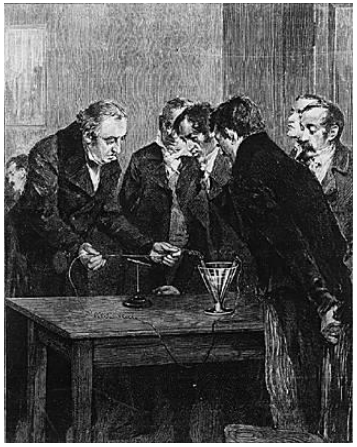
- Connection between magnetism and electricity (19th century)

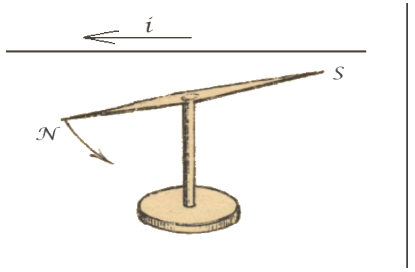


Oersted

Ampère

Faraday





A BRIEF HISTORY

- Maxwell put it in equations

$$\nabla \cdot \boldsymbol{B} = 0,$$
$$\epsilon_0 \nabla \cdot \boldsymbol{E} = \rho,$$
$$(1/\mu_0)\nabla \times \boldsymbol{B} = \boldsymbol{j} + \epsilon_0 \partial \boldsymbol{E}/\partial t,$$
$$\nabla \times \boldsymbol{E} = -\partial \boldsymbol{B}/\partial t.$$

- Magnetism and quantum mechanics

Intrinsic spin of the electron:	Uhlenbeck Goudmsith 1925
Ferromagnetim:	Heisenberg
Antiferromagnetism:	Néel

REVIVAL OF MAGNETISM

Magnetism is still a very active science & technology

- Permanent magnets

Rare earth + transition metal

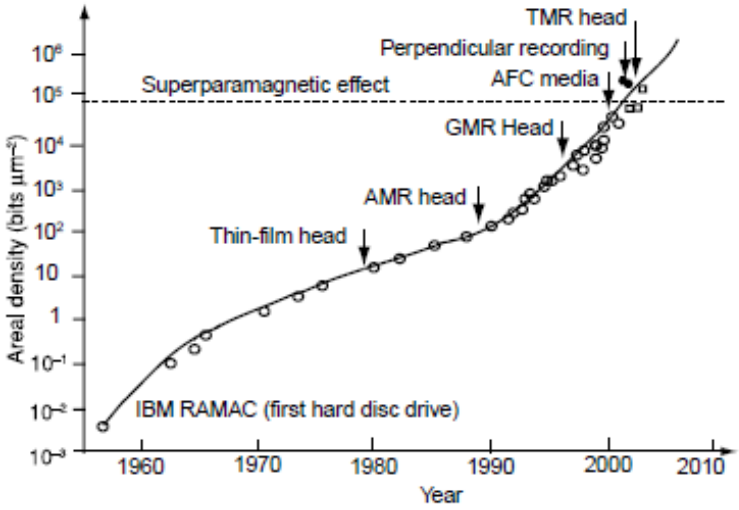
Nd-Fe-B Sm-Co

- Sprintronics & magnetic recording

wires and needles, small particles,
Nanostructured Bulk materials

Nanophysics + Material science

Thin films (GMR)
nanocolumns
Organic and molecular spintronics



CONTENTS

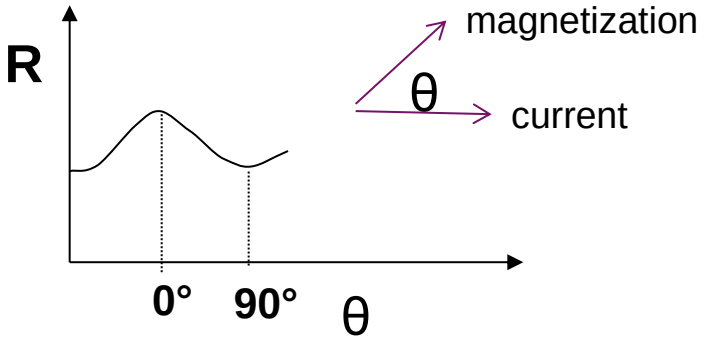
ANISOTROPIC MAGNETO RESISTANCE IN NANOCONTACTS

WHAT IS AMR?

An OLD Story

Anisotropic Magneto Resistance of bulk Iron (Thomson 1857!)

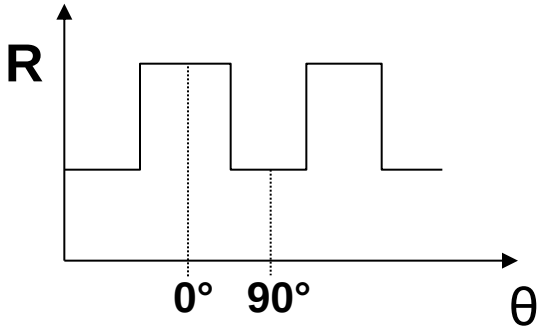
- $\rho_{\parallel} \neq \rho_{\perp}$ $\quad AMR = \dfrac{\rho_{\parallel} - \rho_{\perp}}{\rho_{av}}$
- AMR≈1%
- Origin: Spin-Orbit Coupling (1960)



Revisited by Nanophysics

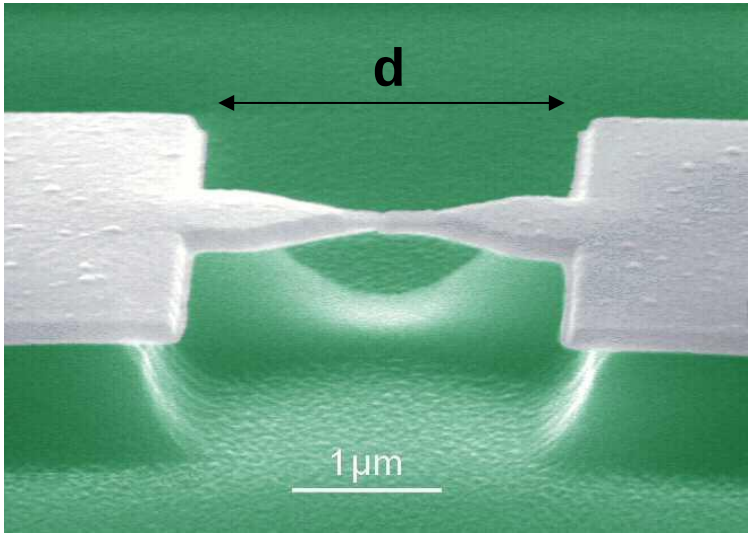
AMR in atomic contacts (M. Viret 2006!)

- AMR≈20-30%
- Two-levels curve

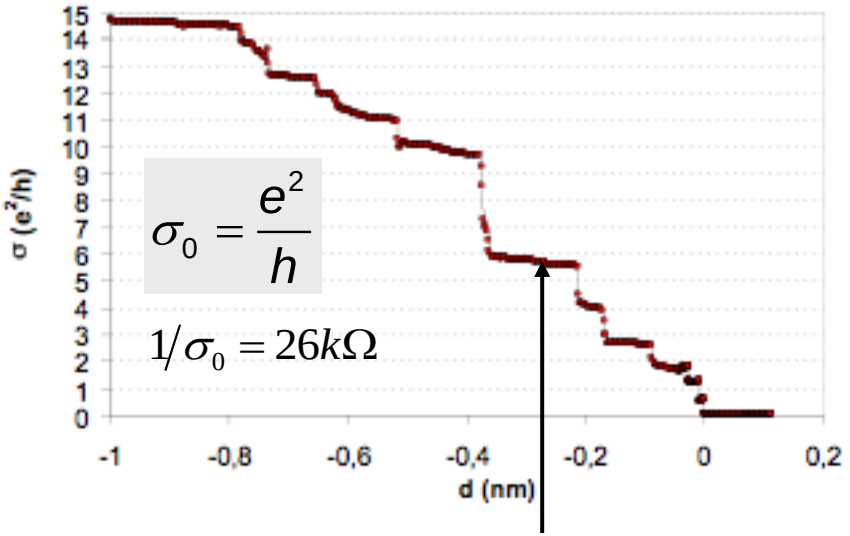


EXPERIMENTAL SET-UP

Break junction technique



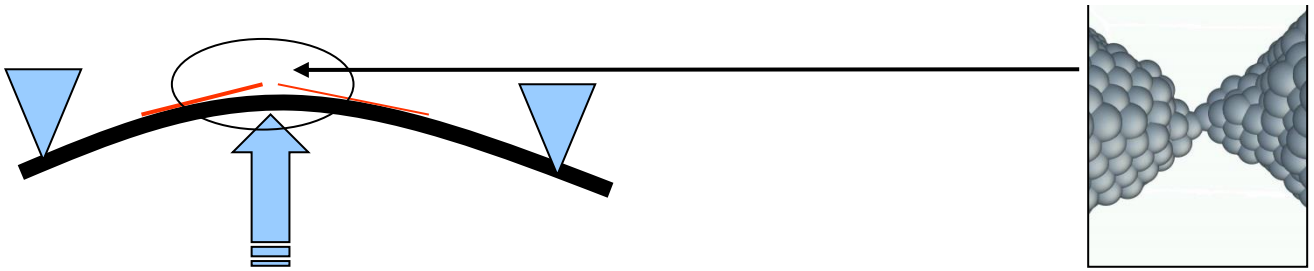
Conductance σ(d)



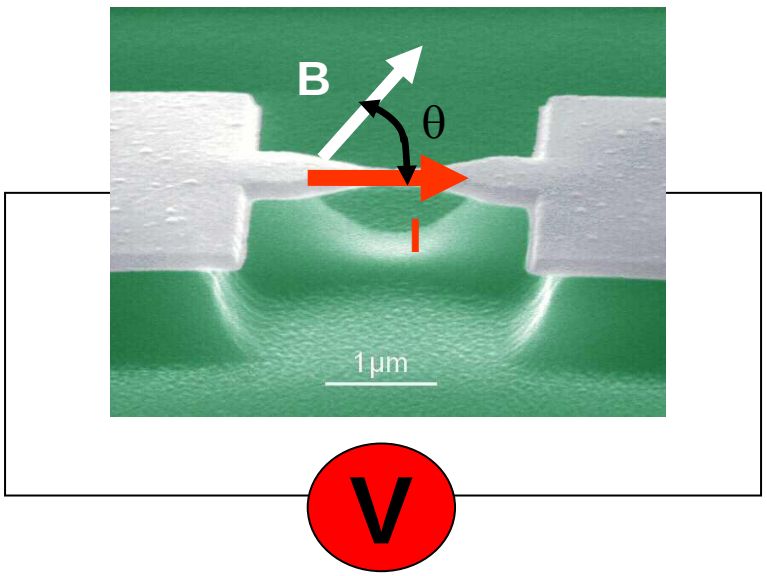
$$\sigma_0 = \frac{e^2}{h}$$

$$1/\sigma_0 = 26k\Omega$$

Atomic contact

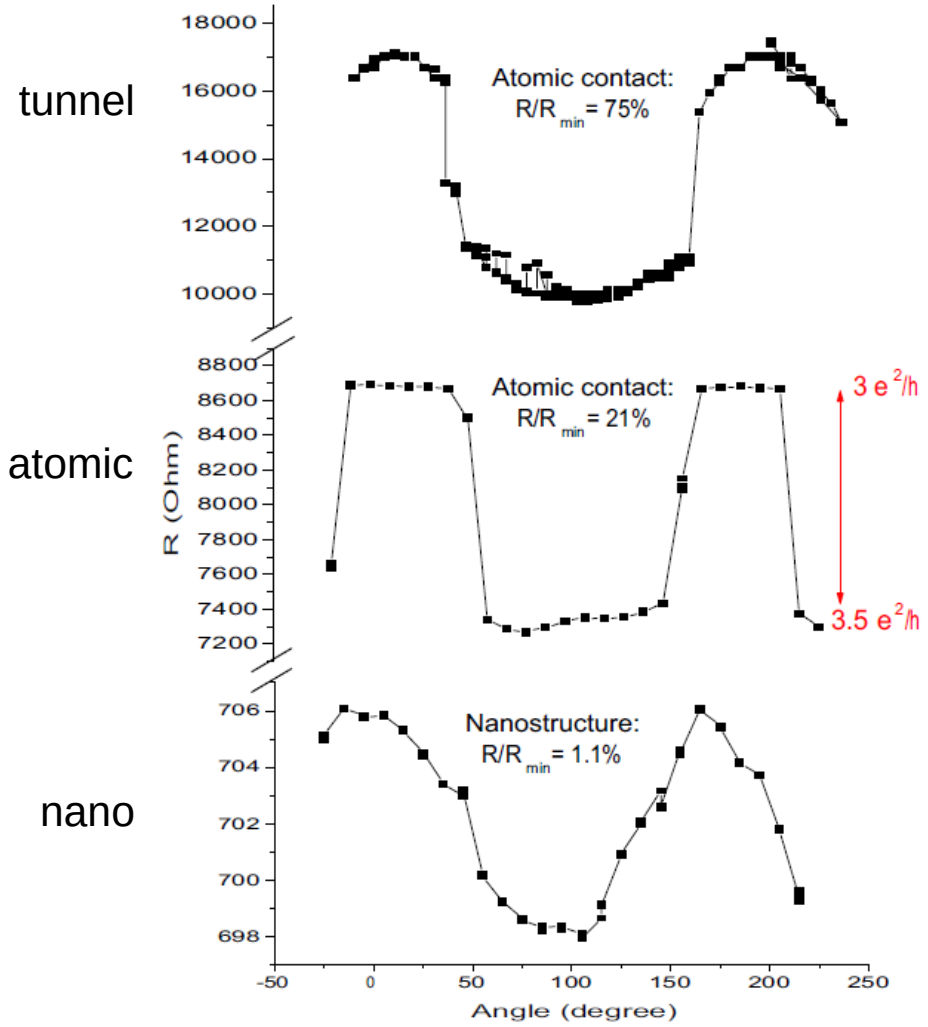


EXPERIMENTAL FACTS



Giant Anisotropic Magnetoresistance

(AMR)



*EPJB **51**,1 (2006)*

MODELLING: MAIN INGREDIENTS

Electronic and Magnetic structure

TB Hamiltonian in a minimal atomic-like basis set

Spin polarized calculation

Spin-orbit coupling

Magnetic anisotropy

ElectronicTransport

Green function formalism in a local basis set

MAGNETIC TB MODEL

$$H = H_0 + H_{LCN} + H_{mag} + H_{SOC}$$

- H_0: Non-magnetic
- H_{LCN}: Local charge neutrality (LCN)
- H_{mag}: Stoner-like or beyond
- H_{SOC}: Spin-Orbit Coupling

$$E_{tot} = \underbrace{E_{band}}_{\text{band energy}} - \underbrace{E_{dc}}_{\text{double counting}}$$

TB_0: MEHL AND PAPACONSTANOPOULOS

$$H_0 = \sum_{ij\lambda\mu} |i\lambda\rangle\langle i\lambda|H|j\mu\rangle\langle j\mu|$$

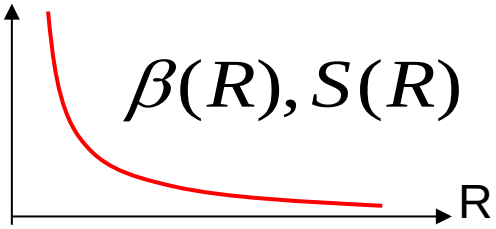
i, j : atoms
λ, μ : orbitals

$\lambda =$ s p_x p_y p_z d_{xy} d_{xz} d_{xz} $d_{x^2-y^2}$ $d_{3z^2-r^2}$

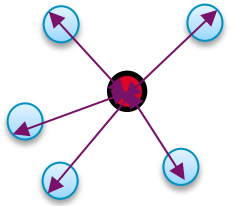
Hopping integrals | overlap integrals | on-site elements

Two-center SK formulation

$$\beta_{i\lambda,j\mu} = \langle i\lambda|H|j\mu\rangle \qquad S_{i\lambda,j\mu} = \langle i\lambda|j\mu\rangle \qquad \varepsilon_{i\lambda} = \langle i\lambda|H|i\lambda\rangle$$



$$\varepsilon_{i\lambda} = f(\rho_i)$$



Total energy: the MP trick

$$E_{tot} = \sum_{\alpha} f_\alpha \varepsilon_\alpha$$

PRB **54**, 4519 (1996)

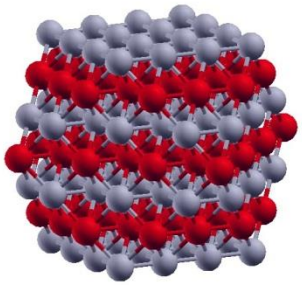
PRB **58**, 9721 (1998)

LOCAL CHARGE NEUTRALITY

$$H_{LCN}^{i\lambda,i\lambda} = U_i \Delta n_i = U_i (n_i - n_{i,0})$$

$$H_{LCN}^{i\lambda,j\mu} = \frac{1}{2}\left[U_i (n_i - n_{i,0}) + U_j (n_j - n_{j,0})\right] S_{i\lambda,j\mu}$$

Avoid charge transfers between inequivalent atoms
(very good for metals)



Double counting

$$E_{tot} = \sum_{\alpha} f_\alpha \varepsilon_\alpha - \tfrac{1}{2} \sum_i U_i (n_i^2 - n_{i,0}^2)$$

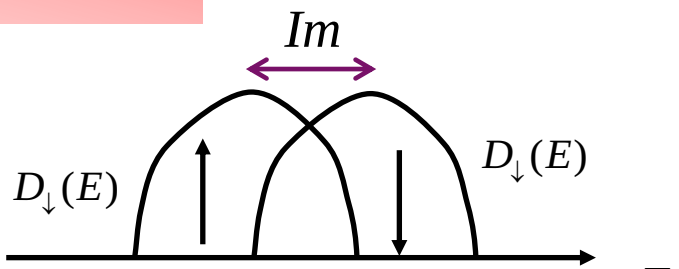
TB_{MAG}: STONER MODEL

Exchange splitting

$$H_{mag} = -\tfrac{1}{2}\sum_{i,\lambda} I_{i,\lambda}\,\vec{m}_{i\lambda}.\vec{\sigma}$$

$$\varepsilon_{i,\lambda,\sigma} \rightarrow \varepsilon_{i,\lambda,0} - \tfrac{1}{2} I_{i,\lambda} m_{i,\lambda}\,\sigma \qquad \sigma = \pm 1$$

$$I_s = I_p = \tfrac{1}{10} I_d$$



$$D_\sigma(E) = D_0(E + \tfrac{1}{2}\sigma IM)$$

Stoner criterion

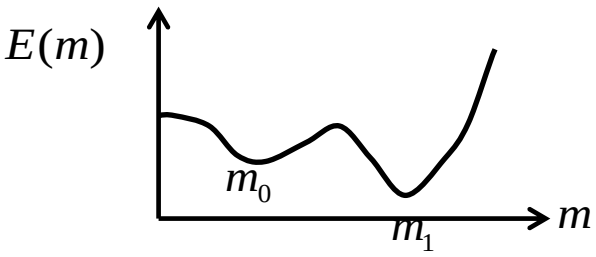
Onset of magnetism when: $ID_0(E_F) \geq 1$

Double counting

$$E_{tot} = \sum_{\alpha_{occ}} \varepsilon_\alpha + \tfrac{1}{4}\sum_{i,\lambda} I_i m_{i,\lambda}^2 = \int^{E_F} ED(E)dE + \tfrac{1}{4}\sum_{i,\lambda} I_i m_{i,\lambda}^2$$

Fixed spin moment calculation

$$E_{tot} = \int^{E_F^{\uparrow}} ED_{\downarrow}(E)dE + \int^{E_F^{\downarrow}} ED_{\downarrow}(E)dE + \tfrac{1}{4}\mathrm{Im}^2 \qquad D_{\sigma}(E) = D_0(E + \tfrac{1}{2}\sigma IM)$$



$$E_{tot}(M) = \int^{E_F^{+}} ED_0(E)dE + \int^{E_F^{-}} ED_0(E)dE - \tfrac{1}{4}\mathrm{Im}^2$$

$$M = \int^{E_F^{+}} ED_0(E)dE - \int^{E_F^{-}} ED_0(E)dE \qquad E_F^{+} = E_F^{\uparrow} + \tfrac{1}{2}Im$$

$$N = \int^{E_F^{+}} ED_0(E)dE + \int^{E_F^{-}} ED_0(E)dE \qquad E_F^{-} = E_F^{\uparrow} - \tfrac{1}{2}Im$$

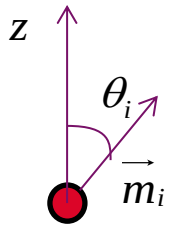
Penalization of local magnetization

$$E_{tot} = \sum_{\alpha occ}\sum_{i,j} c_i^{\alpha} c_j^{\alpha} H_{ij} + E_{\mathrm{pen}}$$

$$E_{\mathrm{pen}} = \sum_i \lambda_i (m_i - m_{i,0})^2$$

$$E_{\mathrm{pen}} = \sum_i \lambda_i (\cos\theta_i - \cos\theta_{i,0})^2$$

$$\bullet \bullet \bullet$$



$$\mathrm{Min}\, E_{\mathrm{tot}} / \sum_i \left(c_i^{\alpha}\right)^2 = 1 \Rightarrow H = H_0 + H_{\mathrm{pen}}$$

BEYOND STONER MODEL= TB+U(J,B)

$$H = H_0 + H_{\text{int}}$$

$$H_{\text{int}} = \frac{1}{2} \sum_{i\alpha_1\alpha_2\alpha_3\alpha_4} U_{i\alpha_1\alpha_2\alpha_3\alpha_4} c^+_{i\alpha_1\sigma} c^+_{i\alpha_2\sigma'} c_{i\alpha_4\sigma'} c_{i\alpha_3\sigma}$$

$\alpha_n = d(f)$ orbitals

i : atomic site

Hartree Fock decoupling (mean field)

$$H_{mag} = H^{HF}_{\text{int}} = \frac{1}{2} \sum_{\substack{i\alpha_1\alpha_2\alpha_3\alpha_4 \\ \sigma\sigma'}} \left(U_{i\alpha_4\alpha_2\alpha_3\alpha_1} \left\langle c^+_{i\alpha_4\sigma} c_{i\alpha_3\sigma} \right\rangle c^+_{i\alpha_2\sigma'} c_{i\alpha_1\sigma'} - U_{i\alpha_4\alpha_2\alpha_1\alpha_3} \left\langle c^+_{i\alpha_4\sigma} c_{i\alpha_3\sigma'} \right\rangle c^+_{i\alpha_2\sigma'} c_{i\alpha_1\sigma} \right)$$

$$U_{m_1m_2m_3m_4} = \int_{-\infty}^{+\infty} d^3r \int_{-\infty}^{+\infty} d^3r' \varphi^*_{im_1\sigma}(\vec{r}) \varphi^*_{im_2\sigma'}(\vec{r}') \frac{e^2}{|\vec{r} - \vec{r}'|} \varphi_{im_3\sigma}(\vec{r}) \varphi_{im_4\sigma'}(\vec{r}')$$

$$U_{\lambda_1\lambda_2\lambda_3\lambda_4} = \int_{-\infty}^{+\infty} d^3r \int_{-\infty}^{+\infty} d^3r' \varphi_{i\lambda_1\sigma}(\vec{r}) \varphi_{i\lambda_2\sigma'}(\vec{r}') \frac{e^2}{|\vec{r} - \vec{r}'|} \varphi_{i\lambda_3\sigma}(\vec{r}) \varphi_{i\lambda_4\sigma'}(\vec{r}')$$

$$U_{\alpha_1\alpha_2\alpha_3\alpha_4} = \text{linear combination}(A, B, C)$$

Racah parameters:

$$(F_0, F_2, F_4)$$

Slater integrals

PRB **76**, 024412 (2007)

New set of parameters

$(A,B,C) \Rightarrow (U,J,B)$

real orbitals

$$U = \frac{1}{4}\sum_{\mu,\mu\neq\lambda} U_{\lambda\mu\lambda\mu} = A - B + C = F^0 - \frac{1}{49}(F^2 + F^4)$$

$$J = \frac{1}{4}\sum_{\mu,\mu\neq\lambda} U_{\lambda\mu\mu\lambda} = \tfrac{5}{2}B + C = \frac{5}{98}(F^2 + F^4)$$

spherical harmonics (Anisimov)

$$U_A = \frac{1}{25}\sum_{mm'} U_{mm'} = A + \frac{7}{5}C = F^0 \qquad U_A - J_A = \frac{1}{20}\sum_{\substack{mm' \\ m\neq m'}} (U_{mm'} - J_{mm'})$$

$$J_A = \frac{7}{2}B + \frac{7}{5}C = \frac{1}{14}(F^2 + F^4)$$

$$\begin{cases} U_A = U + \dfrac{2J}{5} \\ J_A = \dfrac{7}{5}J \end{cases} \Leftrightarrow \begin{cases} U = U_A - \dfrac{2J_A}{7} \\ J = \dfrac{5}{7}J_A \end{cases}$$

From HF to Stoner

$$n_{i,\lambda\sigma,\mu\sigma'} = \bar{n}_{i,\sigma}\delta_{\lambda\sigma,\mu\sigma'} \qquad \bar{n}_{i,\sigma} = \tfrac{1}{5}\sum_{\lambda} n_{i,\lambda,\sigma}$$

$$H_{\text{int}} \rightarrow H = \sum_{i\lambda\sigma}(U_{eff}\, n_{i\lambda\sigma} - \tfrac{1}{2} I_{dd} m_{i\lambda\sigma}) c^{\dagger}_{i\lambda\sigma} c_{i\lambda\sigma}$$

$$U_{eff} = (9U - 2J)/5 \qquad I_{dd} = (U + 6J)/5$$

I_{dd} Controls the spin-moment

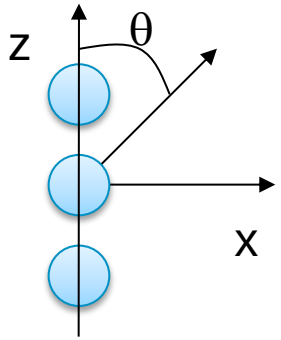
$U - J, B$ Controls the orbital-moment and anisotropy

SPIN-ORBIT COUPLING

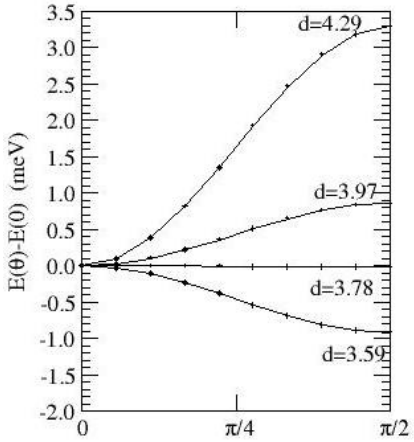
$$H_{SOC} = \sum_i \xi_i(r)\vec{L}_i.\vec{S}_i$$

We keep d orbitals only

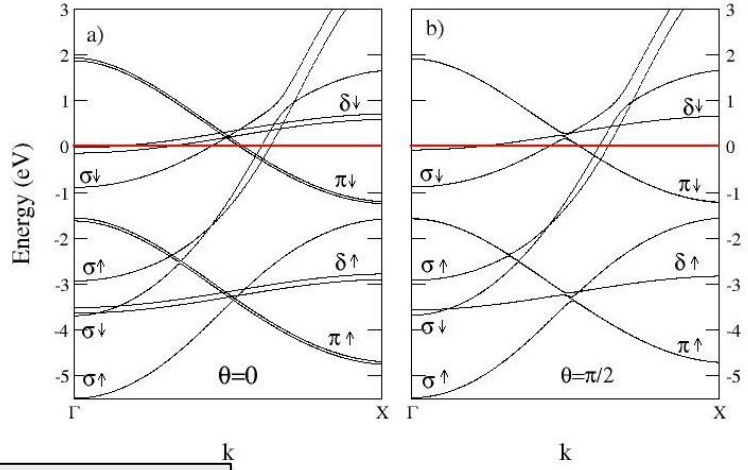
$$\xi_{i,d} = \int R_{i,d}^2(r) r^2 dr$$



Magnetic anisotropy



Band structure anisotropy



Orbital moment

$M_L \sim 0.1\mu_B$ (bulk)
$M_L \sim 0.5-1\mu_B$ (wire)

J. Phys.: Cond. Matter **18**, 6785 (2006)

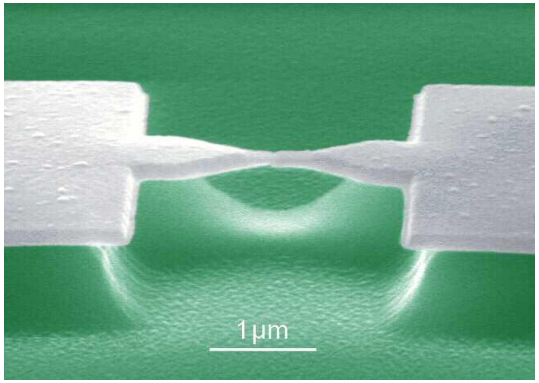
TB PARAMETERS

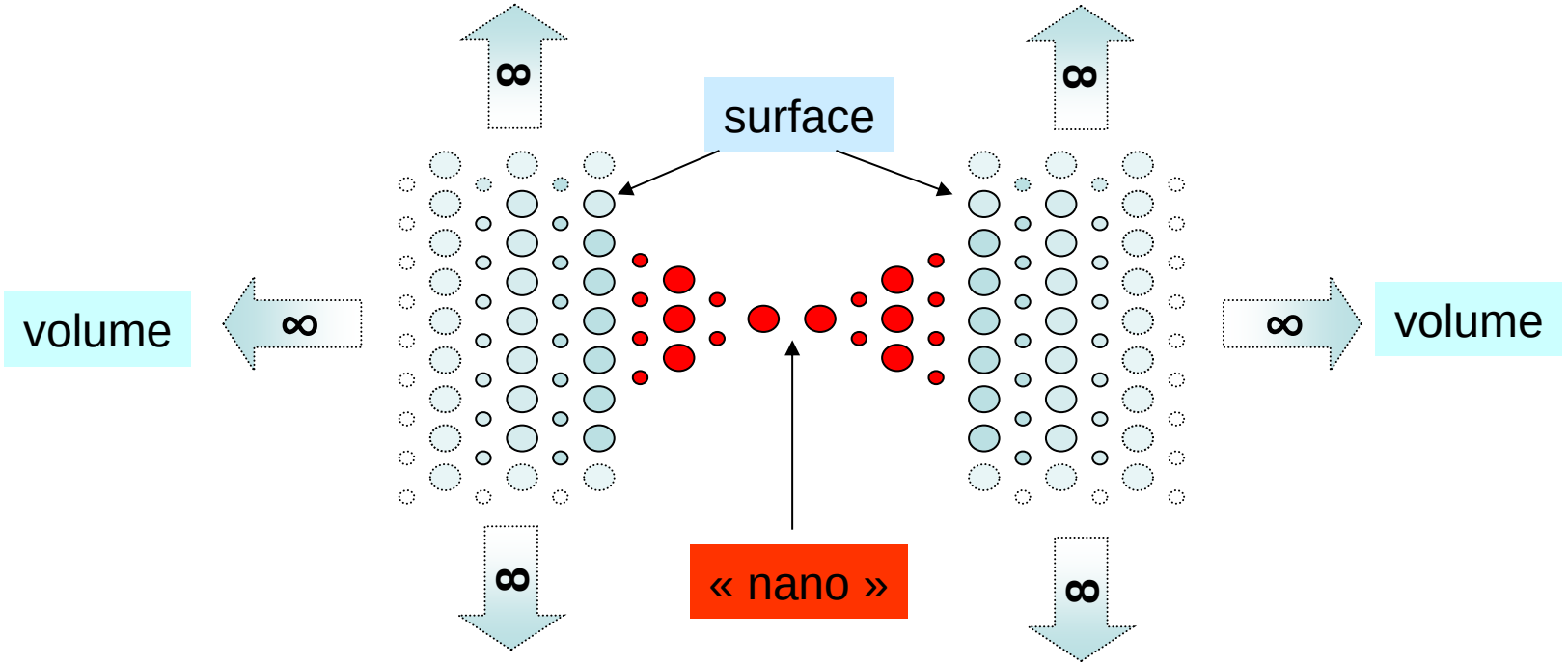
TB Hamiltonian

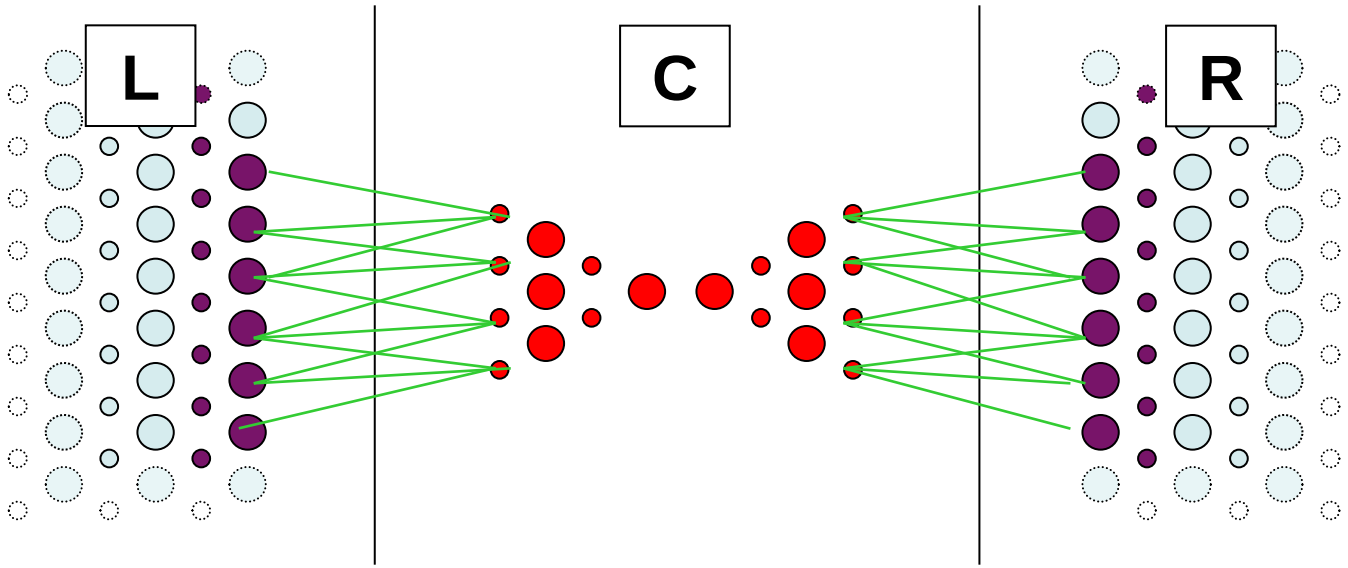
$$H = H_0 + H_{LCN} + H_{mag} + H_{SOC}$$

- H_0: Fitted on ab-initio Bulk band structures and E(V) curve
- H_{LCN}: Local charge neutrality (LCN)
- H_{mag}: Stoner parameter Adjusted on M(d) curve
- H_{SOC}: Fitted on ab-initio Band structure

TRANSPORT MODELLING

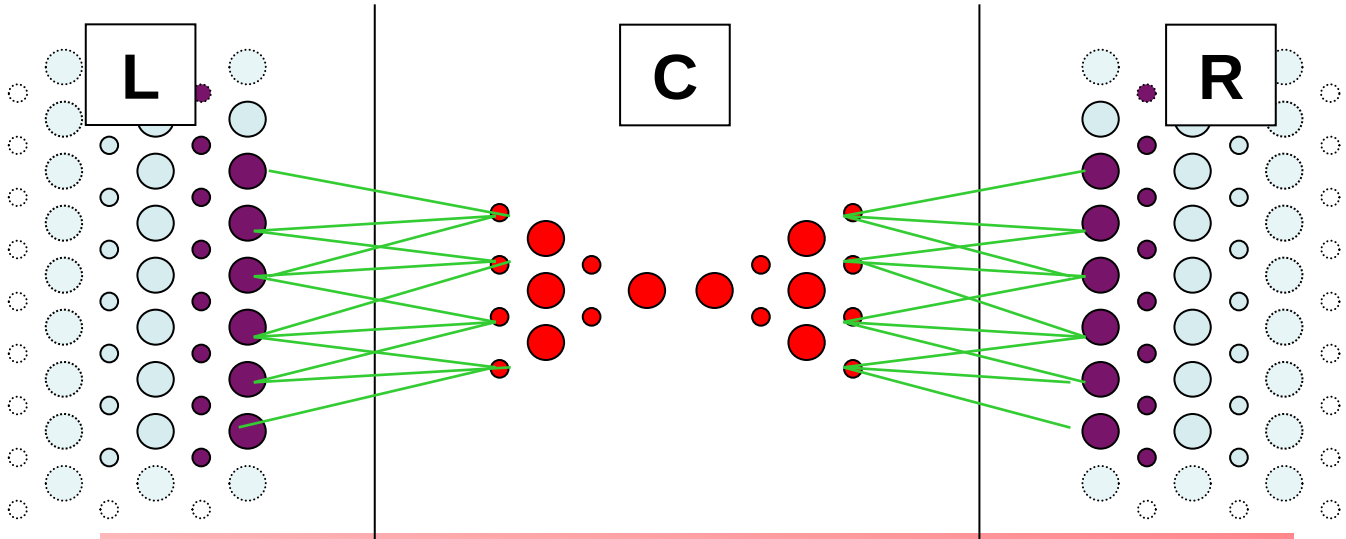






$H_L \quad V_{LC} \quad H_C \quad V_{CR} \quad H_R$

$$H = \begin{pmatrix} H_L & V_{LC} & 0 \\ V_{CL} & H_C & V_{CR} \\ 0 & V_{RC} & H_R \end{pmatrix} \begin{matrix} \updownarrow \infty \\ \updownarrow N = N_{at} N_{orb} \\ \updownarrow \infty \end{matrix}$$



$$T(E) = Tr(\Gamma_L G_C \Gamma_R G_C^+) = Tr(t(E)t^\dagger(E))$$

$$G_C = (E - H_C - \Sigma_L - \Sigma_R)^{-1}$$

« Contact » Green function

$$\Gamma_{L/R} = i(\Sigma_{L/R} - \Sigma_{L/R}^*)$$

Scattering rate matrix

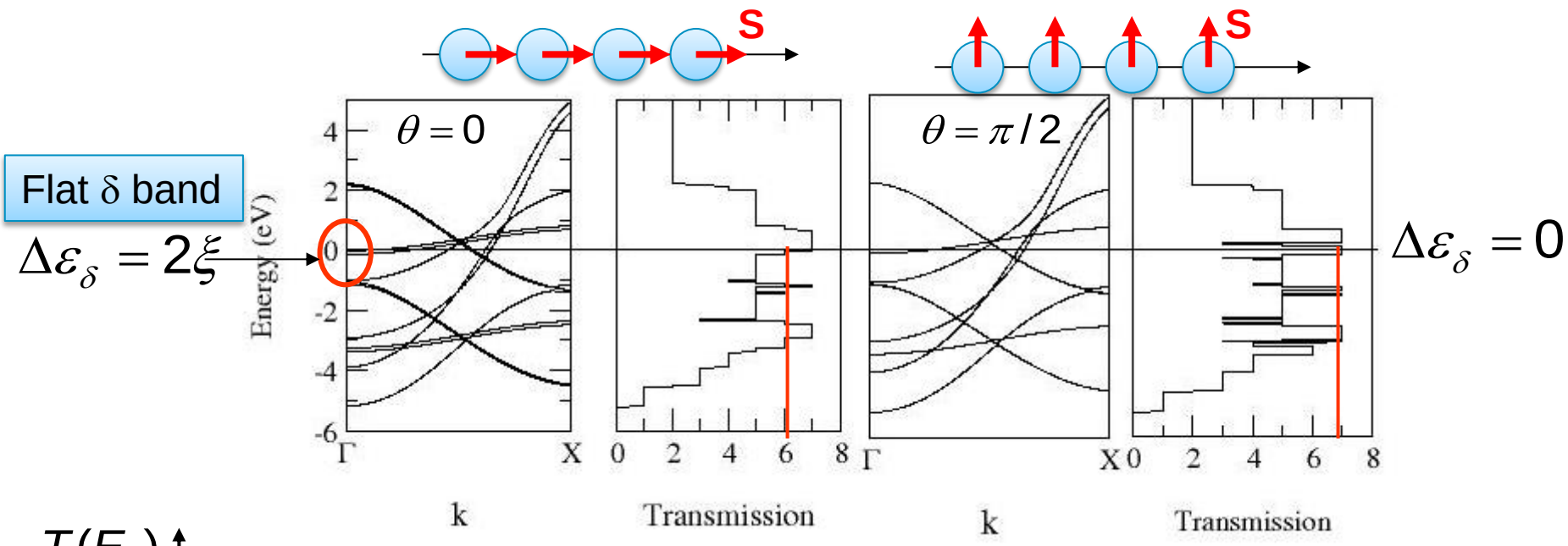
$$\Sigma_{L/R} = V_{C-L/R}(E - H_{L/R})^{-1} V_{L/R-C}$$

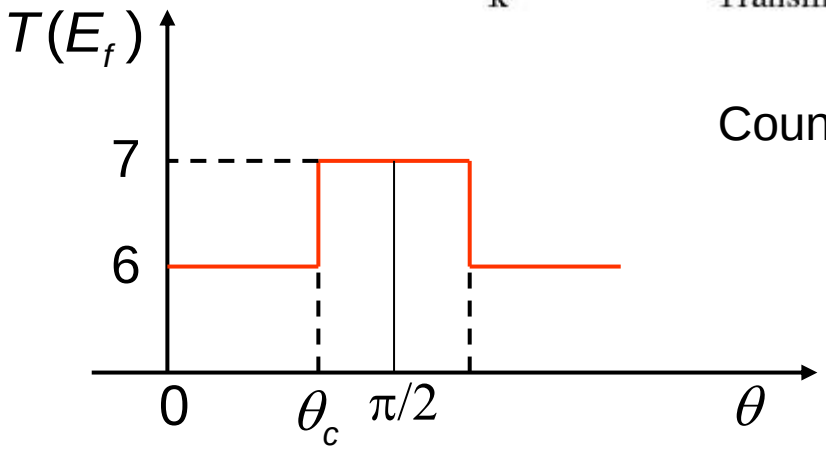
« Electrode » Self-energy

$g^S_{L/R}(E)$ Surface Green function
(iterative Lopez-Sancho algorithm)

SIMPLE ATTRACTIVE (BUT WRONG) MODEL OF AMR IN NANOCONTACTS

BAMR of a monatomic wire

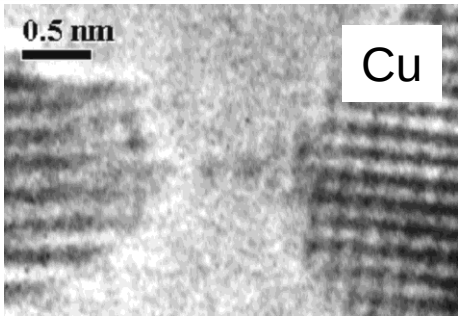




Counting bands at the Fermi level

« REALITY » IS MUCH MORE COMPLEX

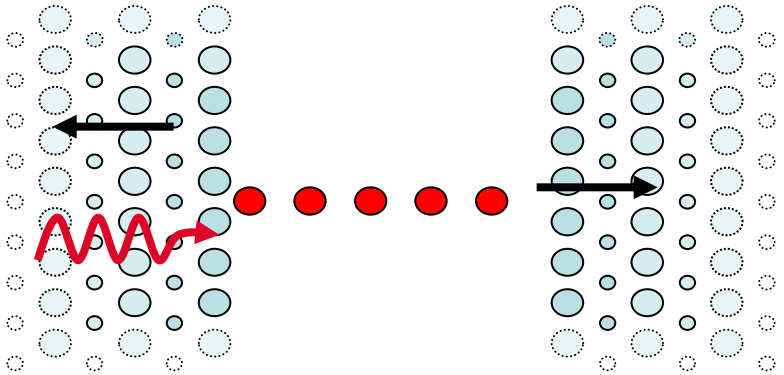
No wire in 3d elements



Role of contacts

Reflexion at interfaces

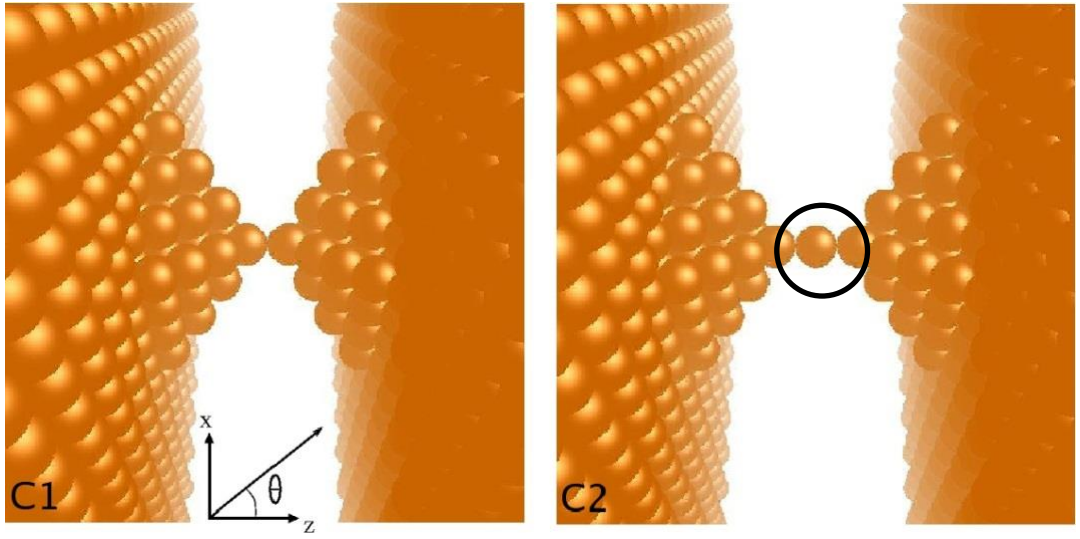
Magnetic coupling with electrodes



« Filtering » of δ-like electrons at the interface

« REALISTIC » MODEL

Realistic atomic contact

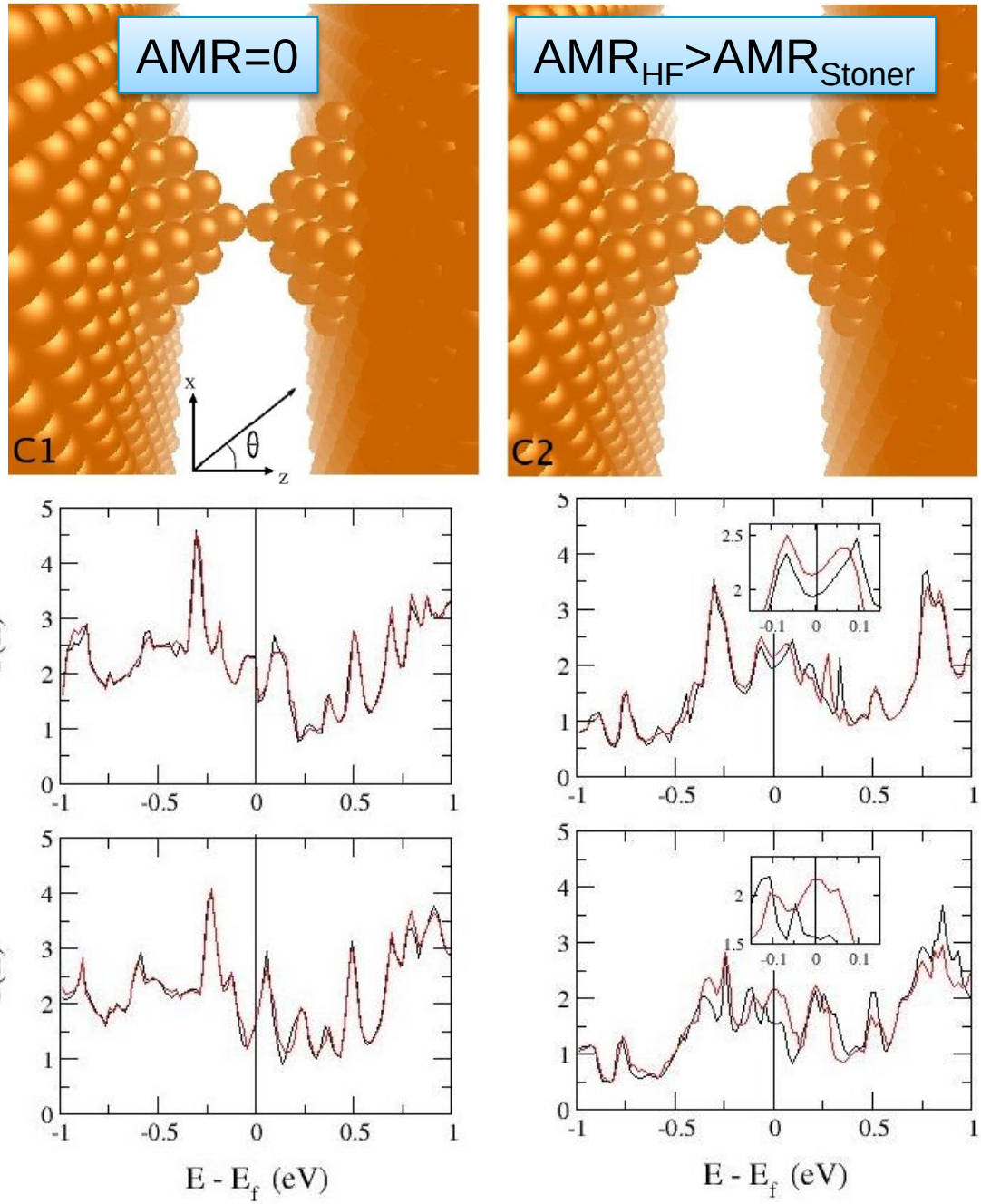


Realistic electronic structure

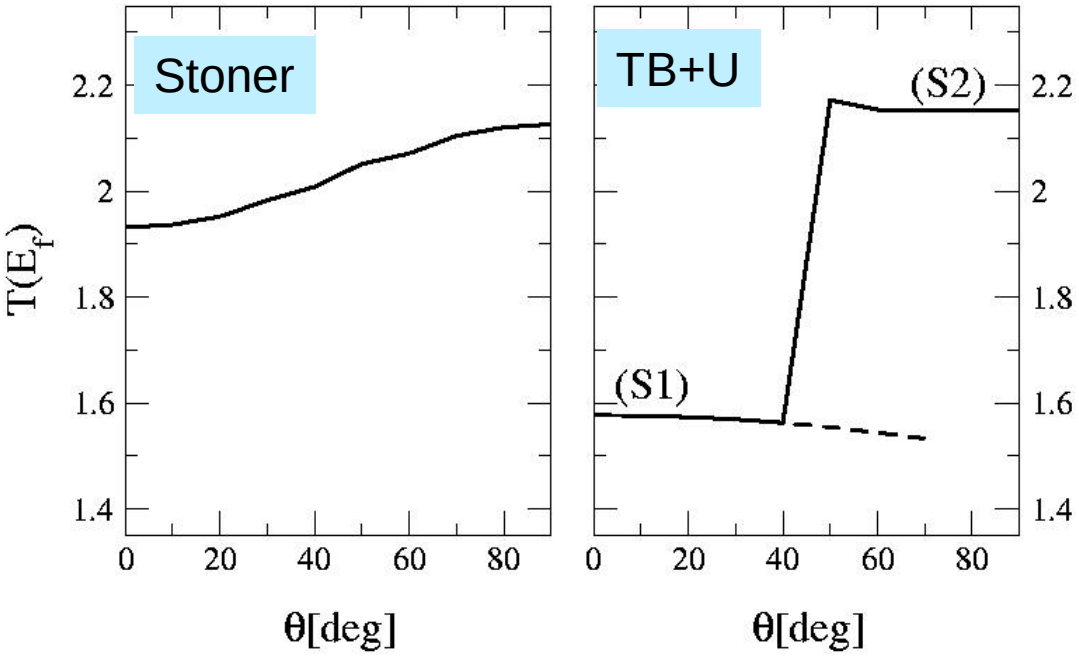
non collinear magnetism + SOC + U(J,B)

$$H = H_0 + H_{LCN} + H_{mag} + H_{SO}$$

$$H_{mag} \begin{cases} H_{Stoner} \\ H_{HF} \end{cases}$$


AMR=0
AMR$_{HF}$>AMR$_{Stoner}$
C1
C2
x
θ
z
T(E)
T(E)
E - E$_f$ (eV)
E - E$_f$ (eV)

AMR(θ)



Right sign of AMR

AMR~10%

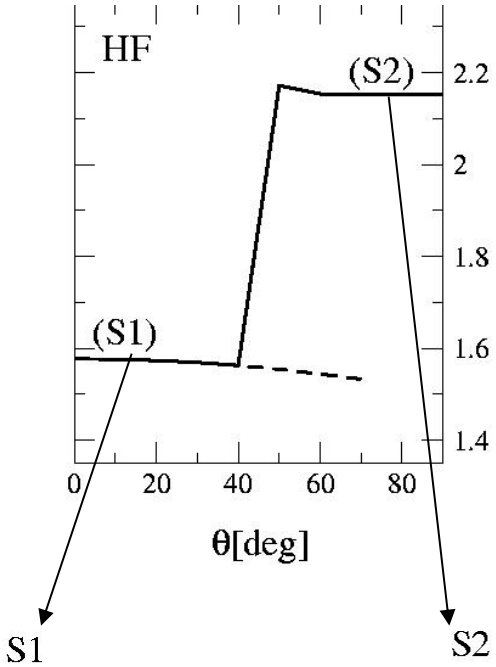
Continuous AMR

Right sign of AMR

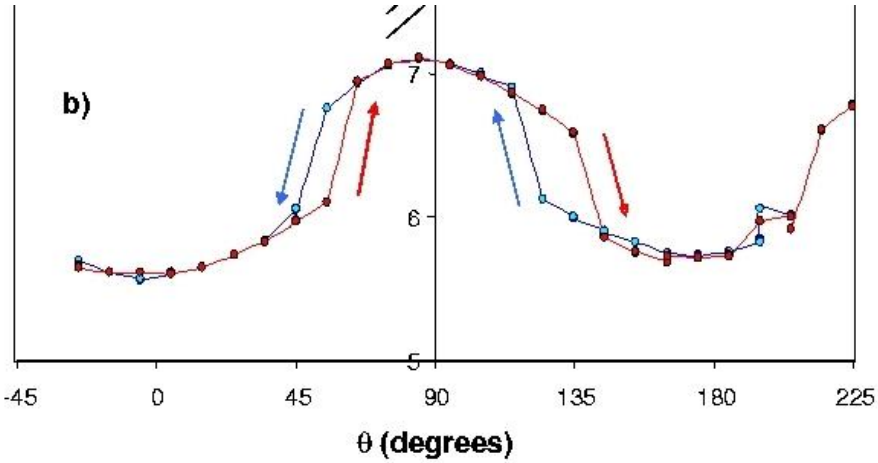
AMR~30%

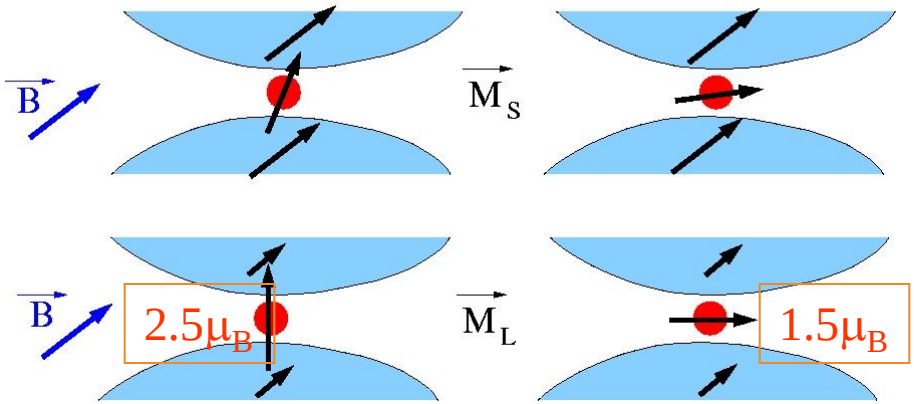
Two-level AMR

From BAMR to AAMR



Hysteresis

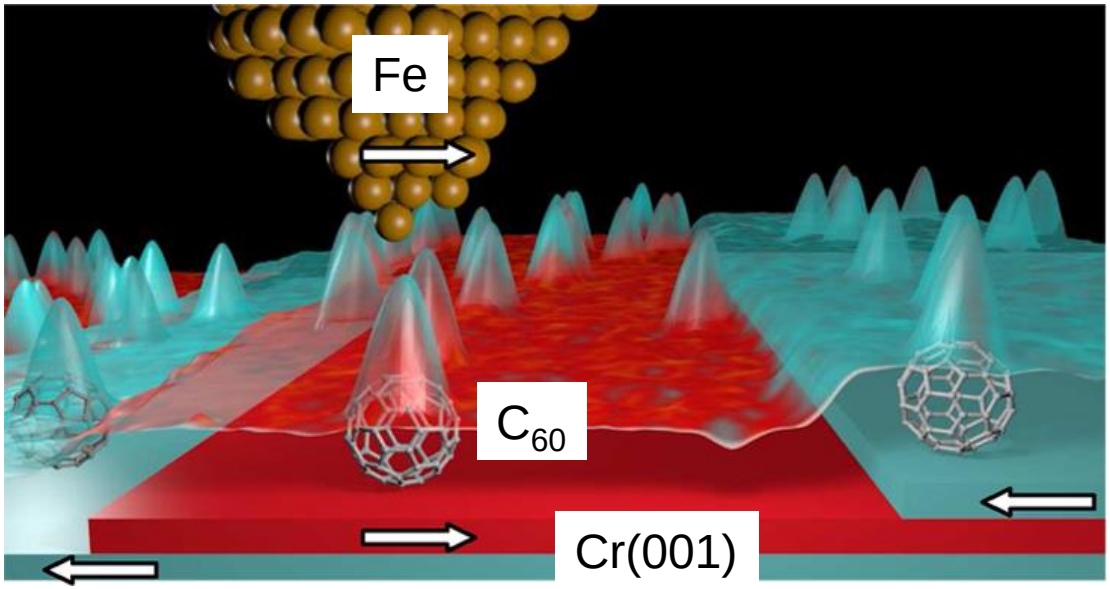




*EPL **83**, 17010 (2008)*

SPIN-POLARIZED TRANSPORT THROUGH C60/CR(001)

SP-STM/STS EXPERIMENTS

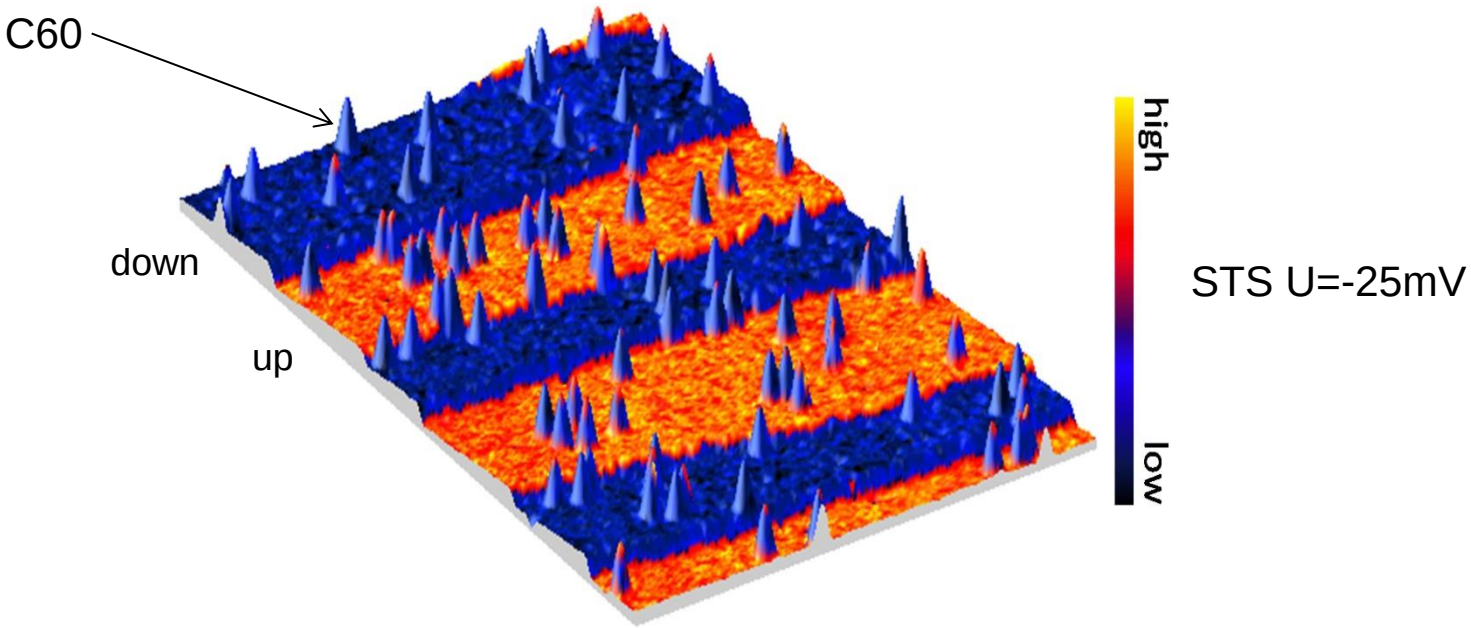


Topographic mode $I(z(x,y), U = cte) = cte \Rightarrow z(x,y)$

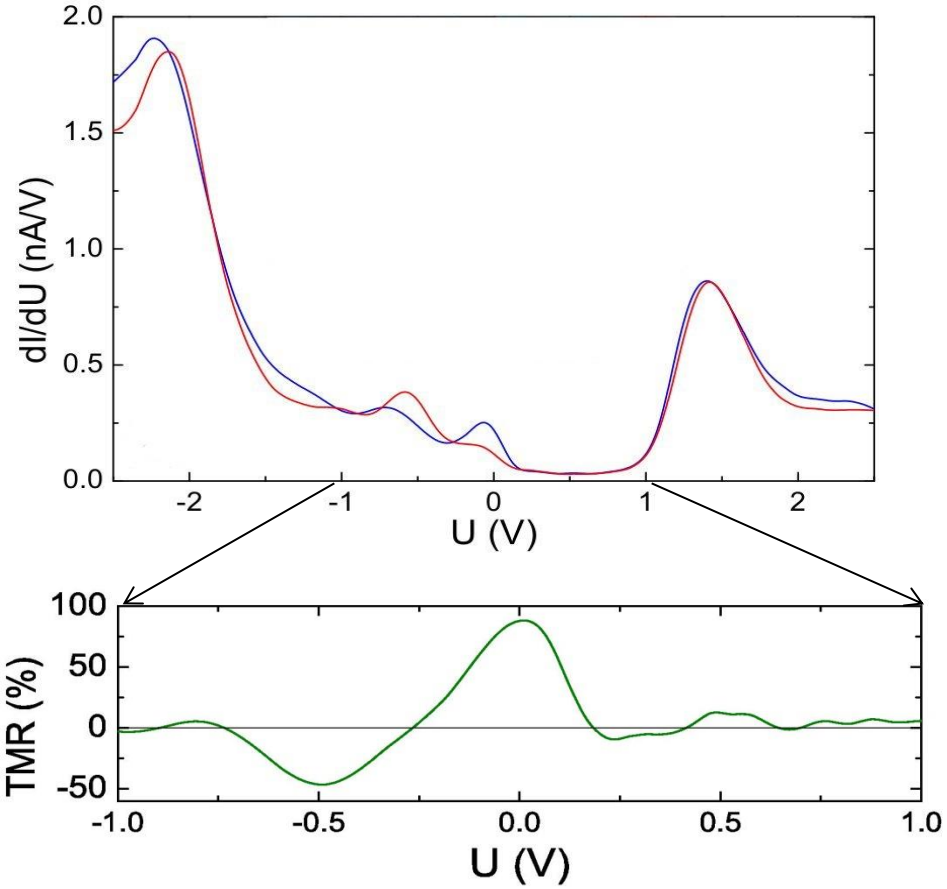
Spectroscopic mode $G = \frac{dI}{dU}(U)$ $z(x,y)$ fixed

Topographic mode

$$U_{bias} = 1V \qquad I = 300\,pA$$

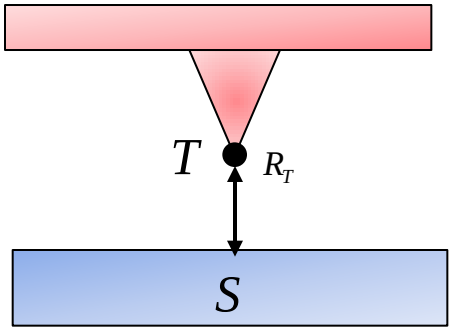


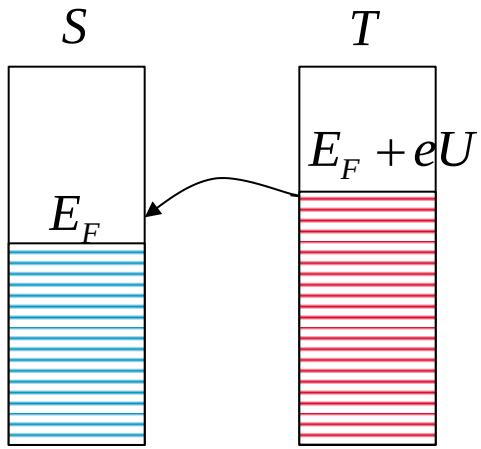
Spectroscopic mode



$$TMR = \frac{G_{up} - G_{down}}{G_{down}} \times 100\%$$

TUNNEL CURRENT
TERSOFF-HAMANN APPROXIMATION





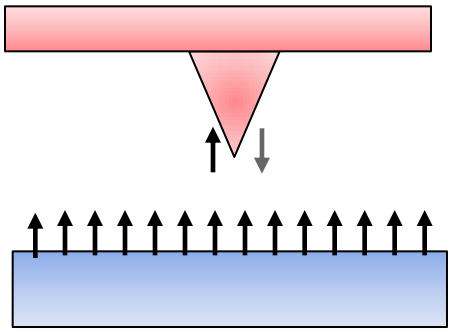
$$I_t(U) \sim D_T \int_{E_F}^{E_F+eU} D_S(E, R_T)\,dE \qquad D_T(E) \sim cte$$

$$G = \frac{dI}{dU} \sim D_T D_S(E_F + eU, R_T)$$

$$D(E_F + eU, R_T) = \sum_{\alpha,k} \left|\Psi_{\alpha,k}(R_T)\right|^2 \delta(E_F + eU - \varepsilon_\alpha(k))$$

Spin-Polarized Tunnel Current

$$G = \frac{dI}{dU} \sim \sum_{\sigma} D_T^{\sigma} D_S^{\sigma} (E_F + eU, R_T)$$



Fully polarized tip

$$D_T^{\uparrow} = 0 \quad D_T^{\downarrow} \neq 0$$

(or vice-versa if the tip and the substrate have opposite polarization)

$$G \sim D_S^{\uparrow\downarrow} (E_F + eU, R_T)$$

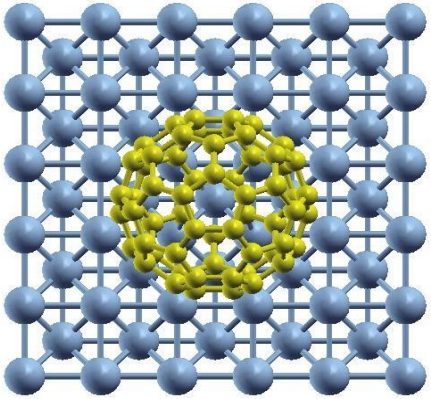
DFT MODELIZATION

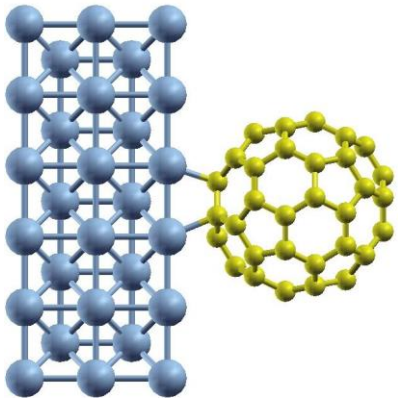
ab initio electronic structure package Quantum-ESPRESSO

Main ingredients

- Density Functional Theory (DFT)
- plane waves basis set for wave functions;
- ultrasoft pseudo potentials to describe electron-ion interactions;

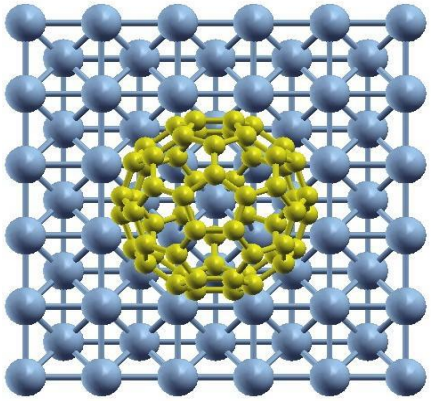
SUPERCELL

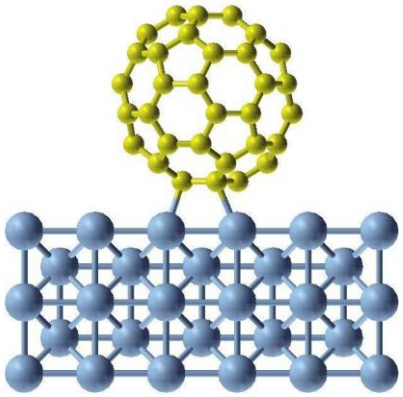




- 185 atoms, size 15Ax15Ax30A, FFT grid = (150,150,320);
- “running time” 30 h on 32 CPU

Atomic relaxation

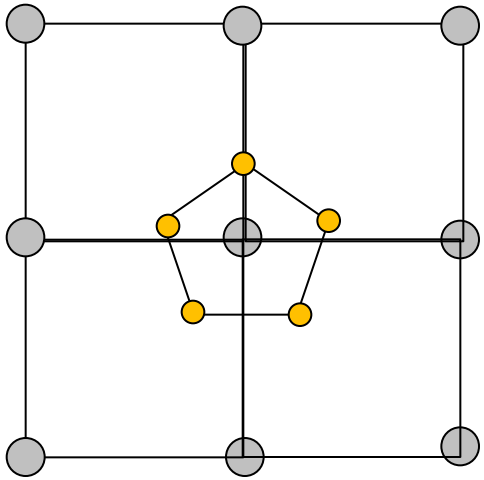




Most stable position:

« on top » on a pentagonal ring

« flatenning » of the molecule in contact with Cr

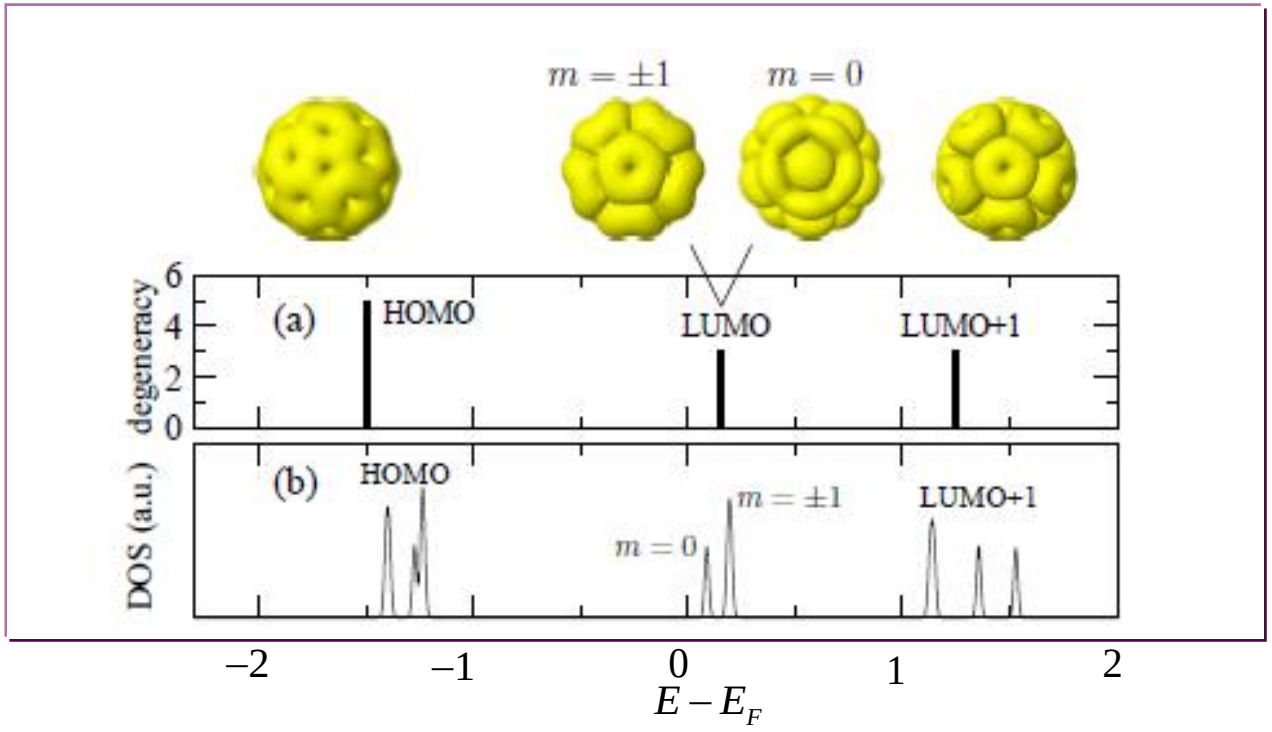


Small anecdote:
Non magnetic C60/Cr(001) is unstable because Cr(001) « wants » to reconstruct
Same as Mo(001) and W(001)

“isolated” C60 molecular spectrum

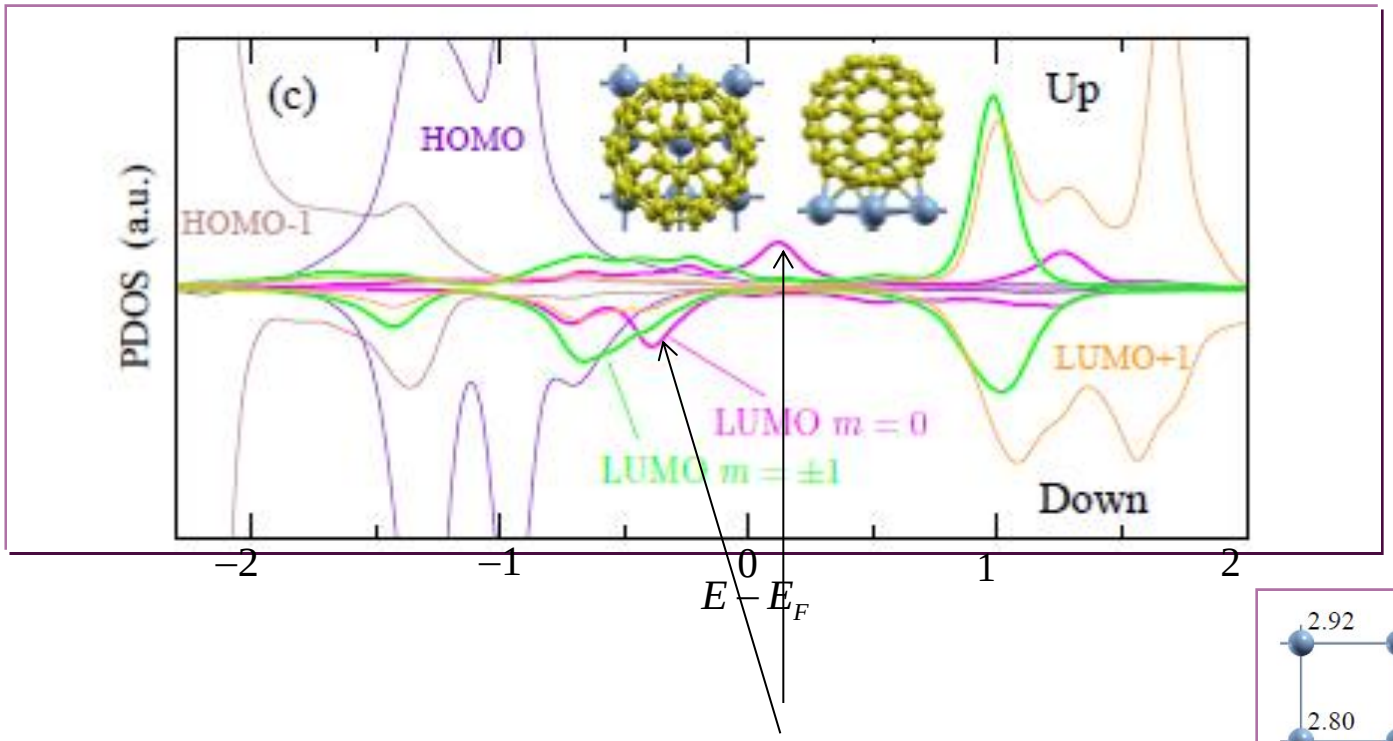
« Spherical » C60

« flattened » C60



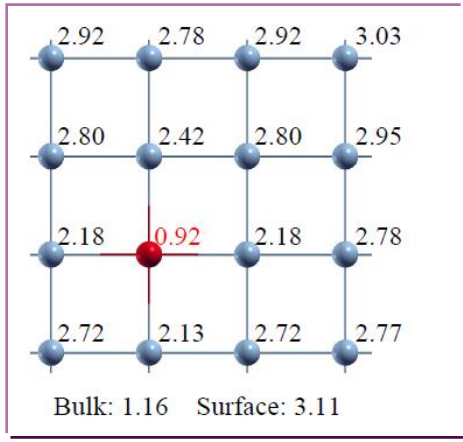
Degeneracy lifting due to the distorsion provoked by the interaction with the substrate

PDOS on molecular orbitals



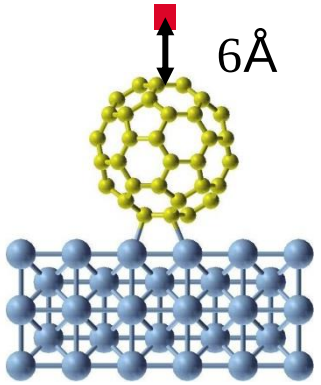
Exchange Splitting of LUMO (m=0)

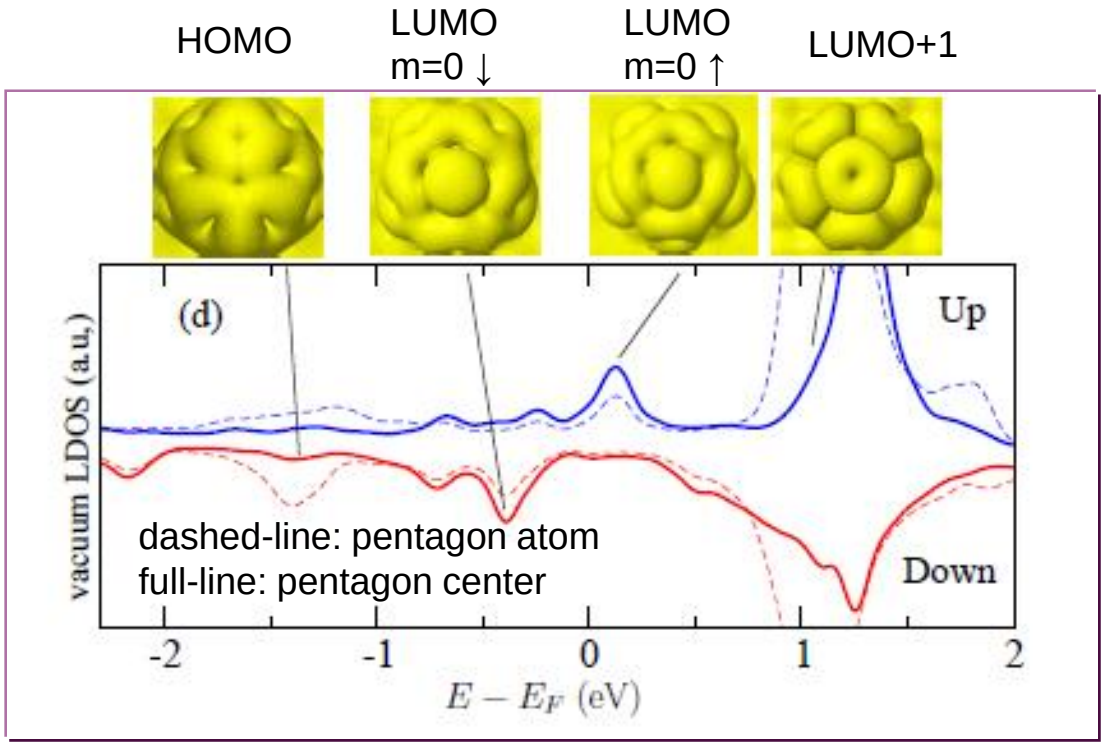
Opposite polarization to the Cr surface
(AF coupling)
Attenuation of Cr surface magnetism



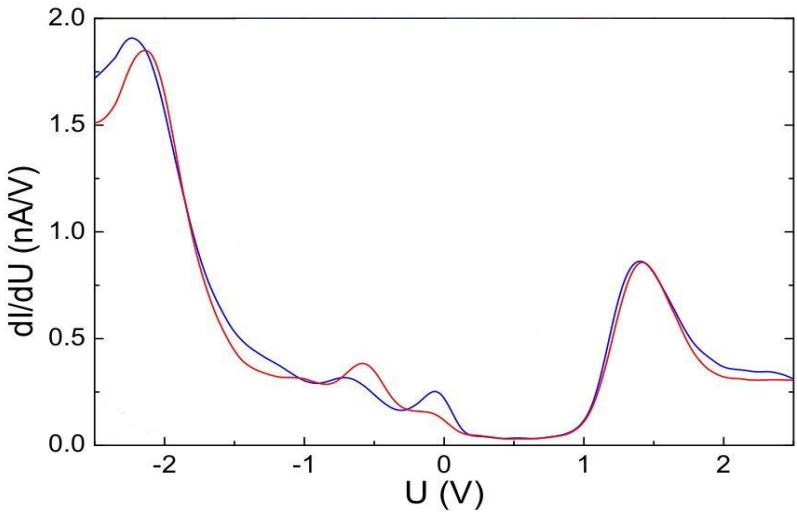
Vacuum LDOS

$$D(E_F + eU, R_T) = \sum_{\alpha,k} \left|\Psi_{\alpha,k}(R_T)\right|^2 \delta(E_F + eU - \varepsilon_\alpha(k))$$





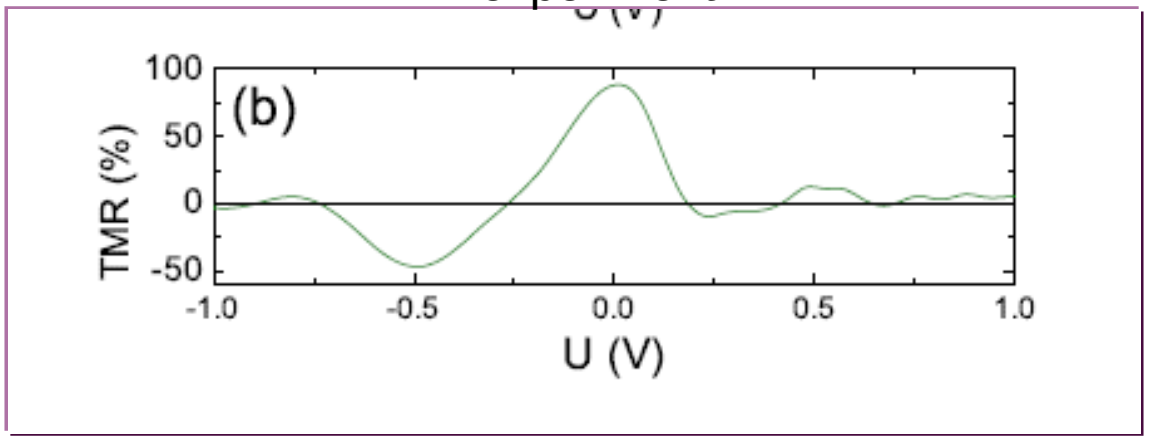
SP-STS (exp)



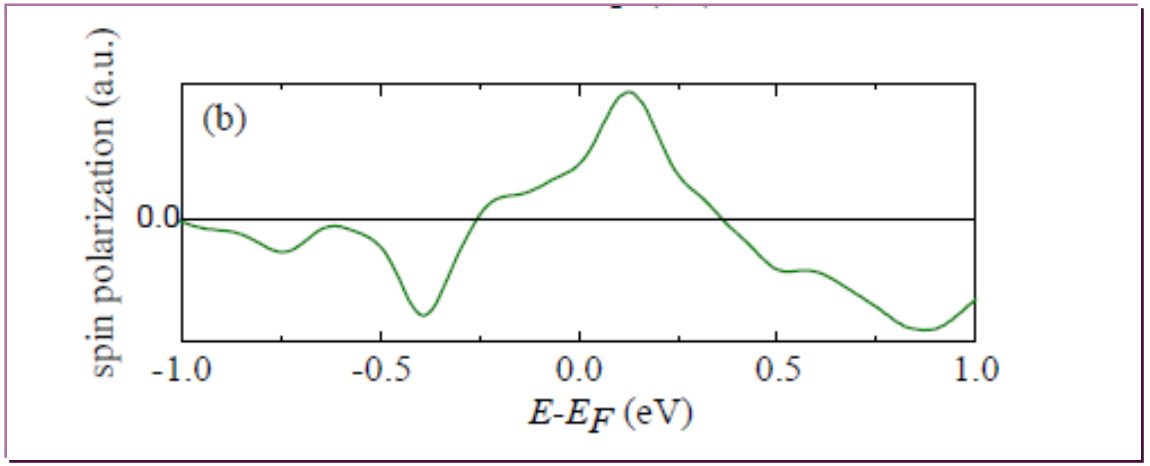
TMR

$$TMR = \frac{\left.\frac{dI}{dU}\right|_{up} - \left.\frac{dI}{dU}\right|_{down}}{\left.\frac{dI}{dU}\right|_{down}}$$

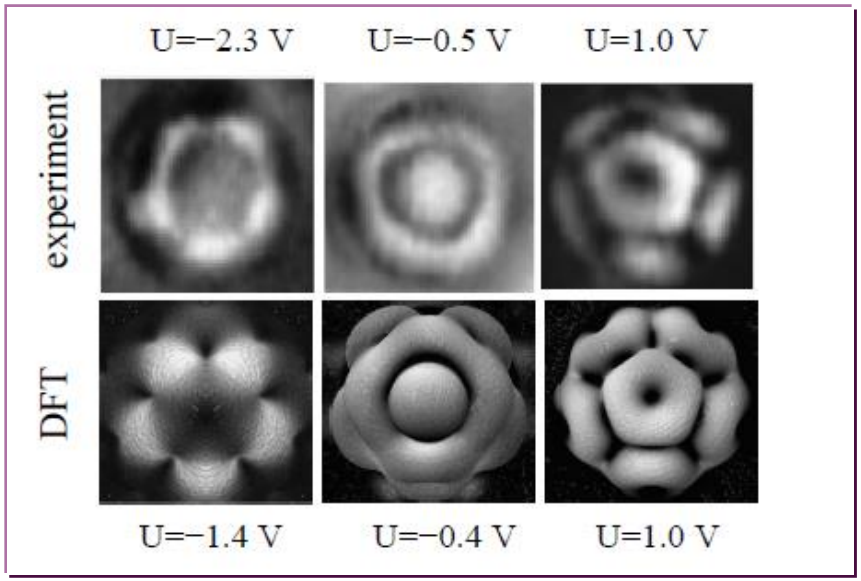
experiment



theory



SP-STS+ topography



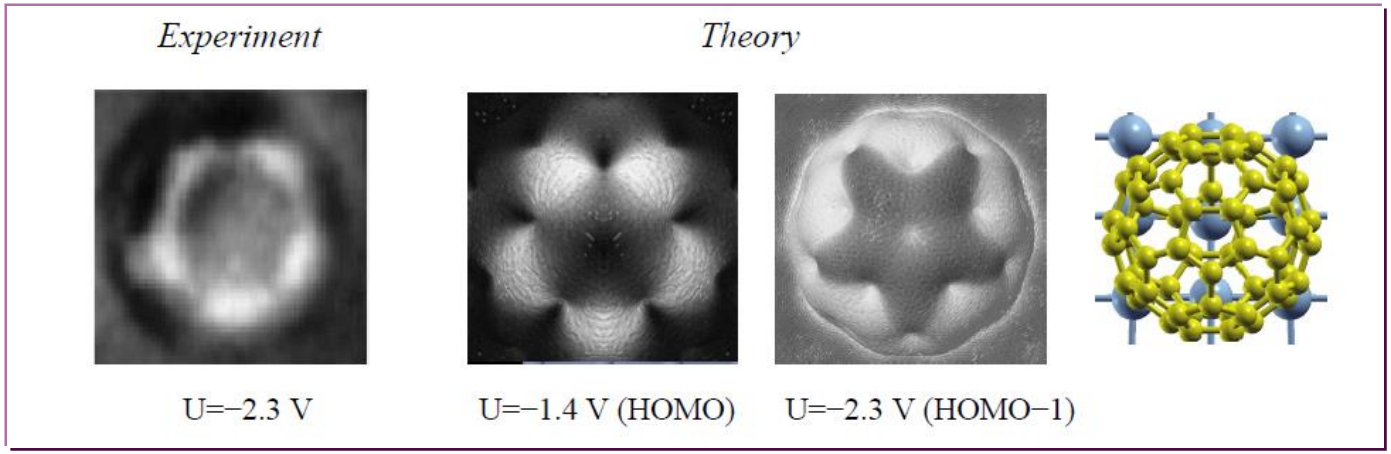
Topography + spectroscopy

$$I(z(x,y),U) = cte$$

$$G = \frac{dI}{dU}(U)$$

$$I_t(U) \sim \int_{E_F}^{E_F+eU} D_S(E, R_T(x,y,z))dE$$

$$G \sim D_S(E_F + eU, R_T(x,y,z))$$



CONCLUSIONS & PERSPECTIVES

nanocontacts

❶ Enhanced magnetoresistive effect

❷ Correlation & orbital polarization can be important

❸ Take into account potential drop in the contact

❹ Nanocontacts of alloys

Molecular spintronics

❶ Large TMR effects are possible

❷ Strong dependence on the atomic geometry

❸ Can we control spin transport with an external parameter

❹ Can we optimize the system to enhance TMR?

Special Thanks to

M.C. Desjonquères D. Spanjaard

G. Autès A. Smogunov Spin polarizedTransport

M. Viret Break junction experiment

J. Lagoute & V. Repain SP-STM experiment

THANK YOU FOR YOUR ATTENTION

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