

Single magnetic clusters embedded in matrix

Véronique Dupuis

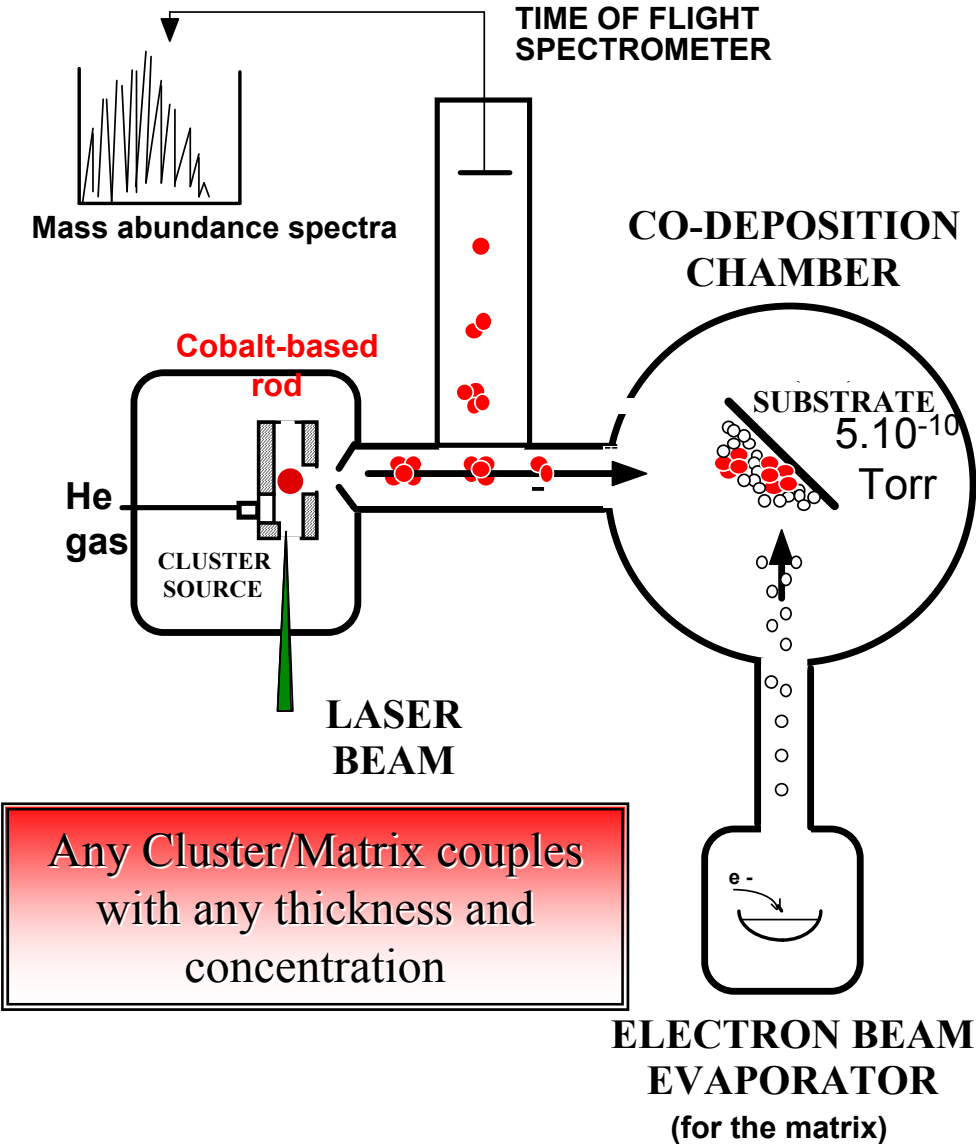
*Laboratoire de physique de la matière condensée et nanostructures
UMR 5586 Université Claude Bernard Lyon I – CNRS Villeurbanne, France*



- **Synthesis, Structure and Superparamagnetism
on Assembled clusters films**
- **Measurements one single nanoparticle**

co-deposition of clusters

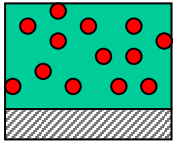
Low Energy Clusters Beam Deposition



D₁ Thick samples :

Assembled clusters films

500 nm matrix + < 5% cluster concentration



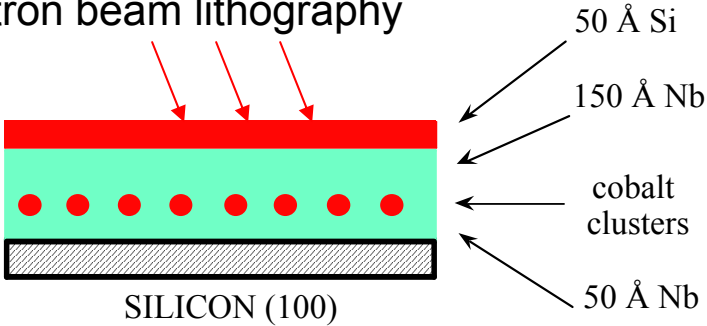
D₂ Thin samples :

One single particle

ex : MicroSQUID :

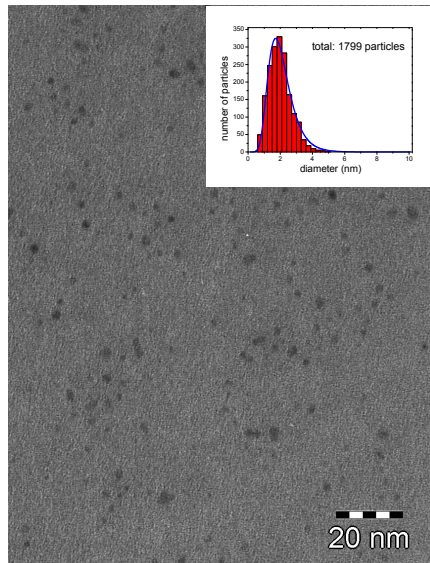
20 nm Nb + < 0.1% cluster conc.
(< 5 clusters/micro-bridge)

Electron beam lithography



M. Jamet, V. Dupuis et al. PRB (2000)

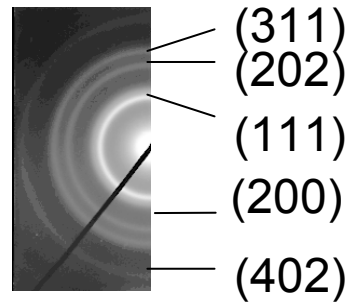
TEM observations on mixed CoPt clusters



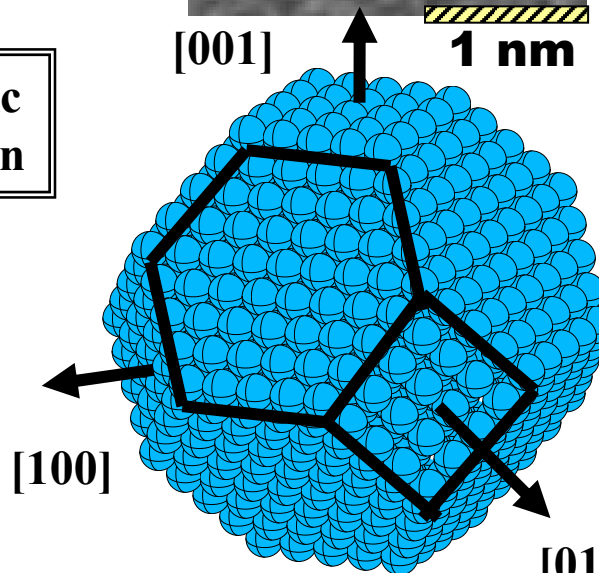
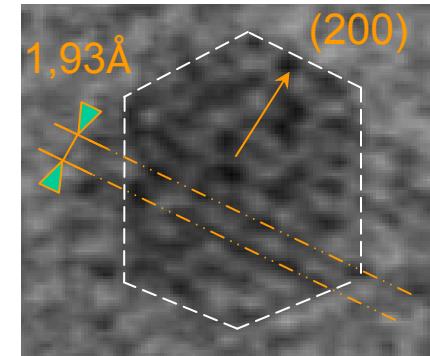
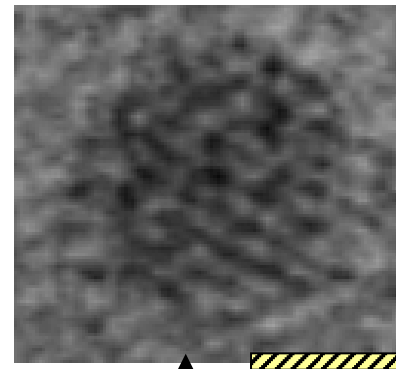
Log-normal size distribution

$$f(D) = \frac{1}{D\sqrt{2\pi\sigma^2}} \exp\left(-\ln^2\left(\frac{D}{D_m}\right) \cdot \frac{1}{2\sigma^2}\right)$$

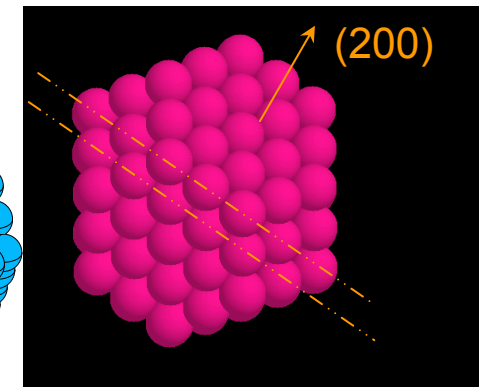
Supported CoPt clusters are fcc
With a mean diameter ϕ CoPt ≈ 2 nm



Electronic diffraction



Truncated octahedron
with (100) and (111) faces



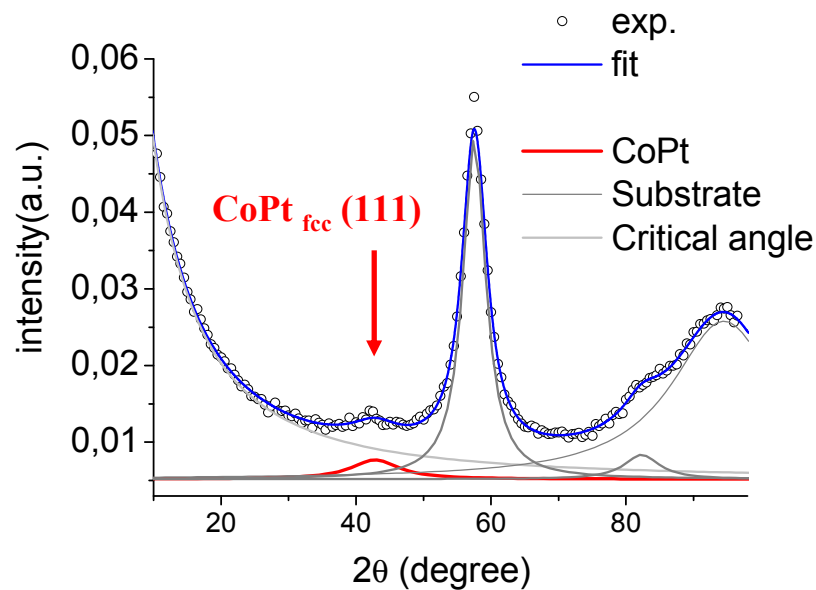
Isolated cluster
along [110]

GIWAXS on mixed CoPt clusters

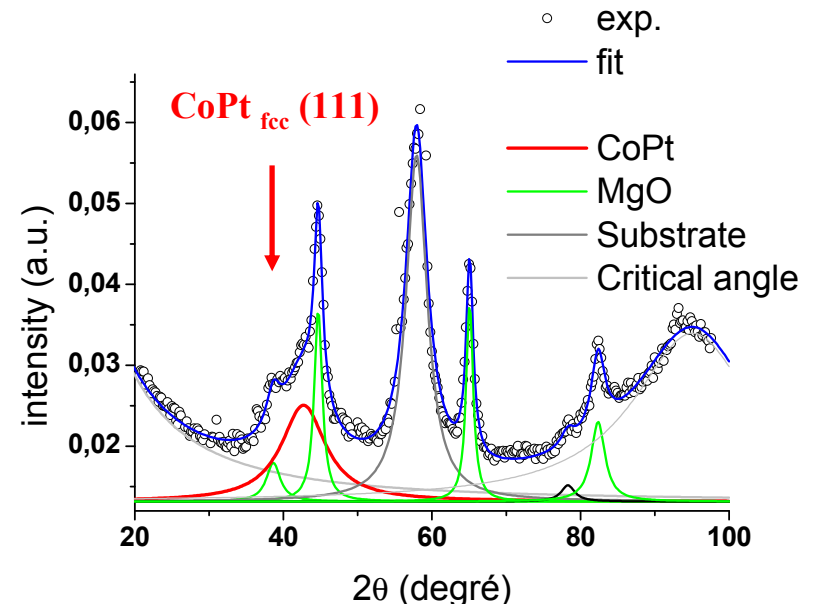
D2AM

(ESRF, Grenoble)

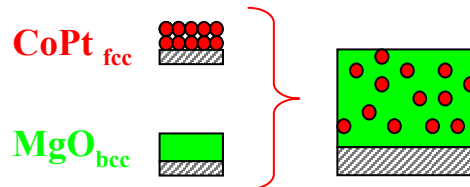
Supported **CoPt** clusters (without matrix)



CoPt clusters in **MgO** matrix



Embedded in MgO matrix
fcc CoPt is conserved



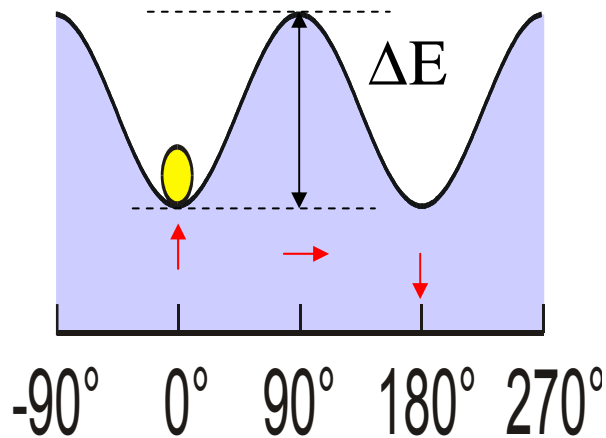
But from Debye Scherrer,
diameter ϕ CoPt \approx 1.3 nm
is reduced : \Rightarrow Diffuse interface
in MgO

L. Favre, V. Dupuis et al. Submitted to Phys. Rev. B (2005)

One magnetic nanoparticle

Stoner and Wohlfarth model

ΔE is the energy barrier at $T=0K$ and $H_{\text{appl}}=0T$



$$\Delta E = K D^\alpha$$

Anisotropy constant

Cluster diameter

$\alpha = 2$, surface anisotropy
 $\alpha = 3$, volume anisotropy

Superparamagnetism :

τ is the relaxation time at $T \neq 0K$ and $H_{\text{appl}} = 0T$

$$\tau = \tau_0 e^{\Delta E / k_B T}$$

with $\tau \approx 10^{-9}$ s for cluster

If $\tau_{\text{mes}} \gg \tau$: $\langle \mathbf{M} \rangle = 0$, superparamagnetic state

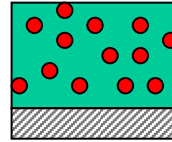
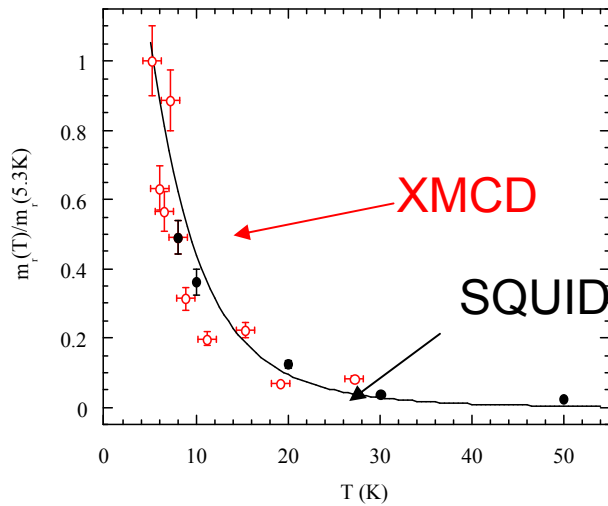
When $\tau_{\text{mes}} = \tau$: One define T_B (resp. D_B), Blocking Temperature (resp. Diameter)

Possible to extend to a clusters assembly without interaction

Magnetic clusters assembly in matrix : Effective Anisotropy

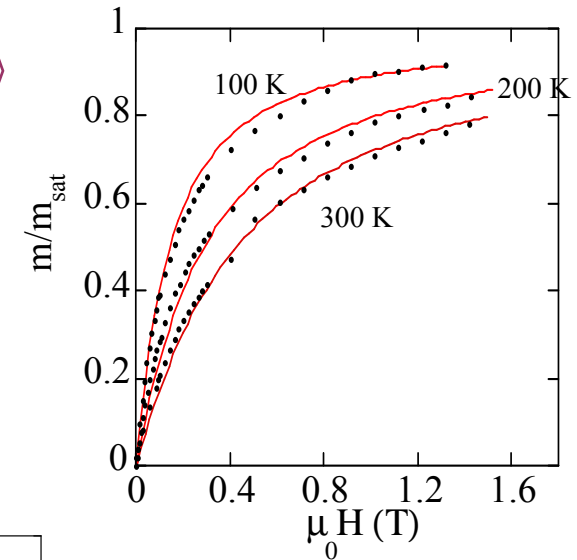
At low temperatures

- From M_r (T)



In the superparamagnetic state

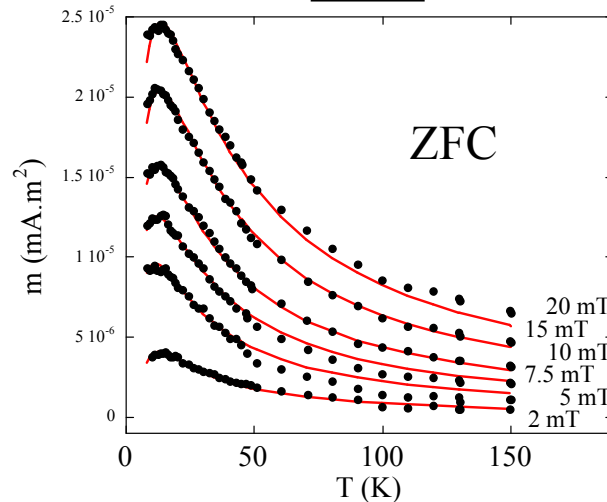
-From M/M_{sat} (T)



$$\Delta E = K_{eff} D \alpha$$

- From ZFC

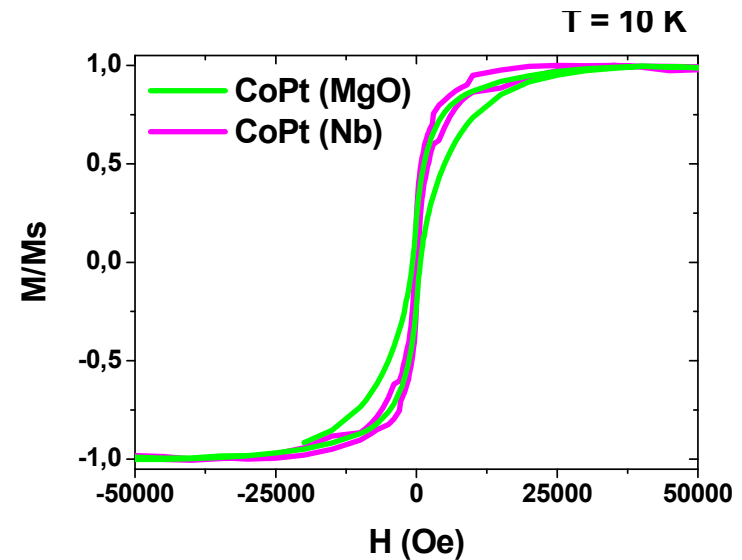
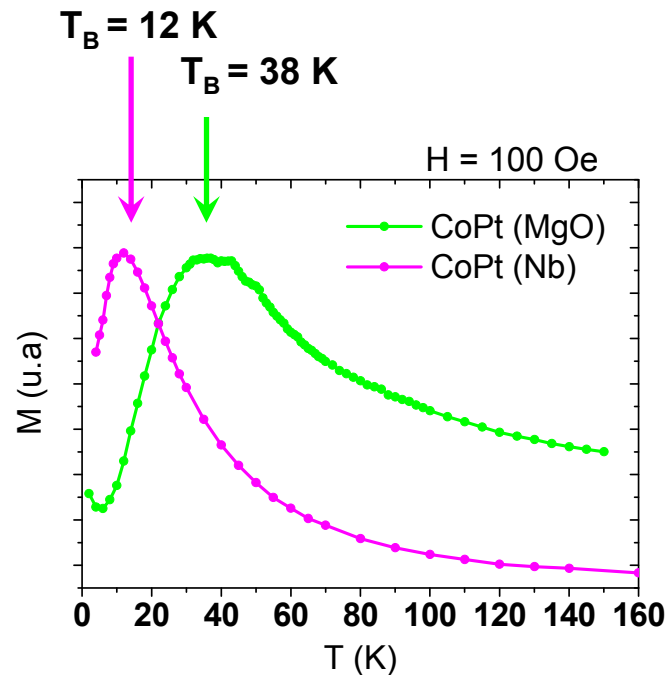
$$T_B$$



$$K_{eff} = \frac{6k_B T}{\pi D_B^3 \alpha(T)} \ln \left(\frac{\tau_{mes}}{\tau_0} \right)$$

M. Jamet et al. PRB (2000)

Magnetic Anisotropy of CoPt clusters in various matrices:



Interface effects

Blocking Temperature : $T_B \propto K D^\alpha$:

⇒ **Higher MAE in MgO than in Nb**

From ZFC simulations:

$K_{\text{Surf CoPt (in MgO)}} > K_{\text{Surf CoPt (in Nb)}}$

From XMCD and GIWAXS experiments:

$D_{\text{mag (in Nb)}} < D_{\text{mag (in MgO)}} < D_{\text{TEM}}$

Matrix	H_C	T_B
Nb	150 Oe	12 K
MgO	750 Oe	38 K

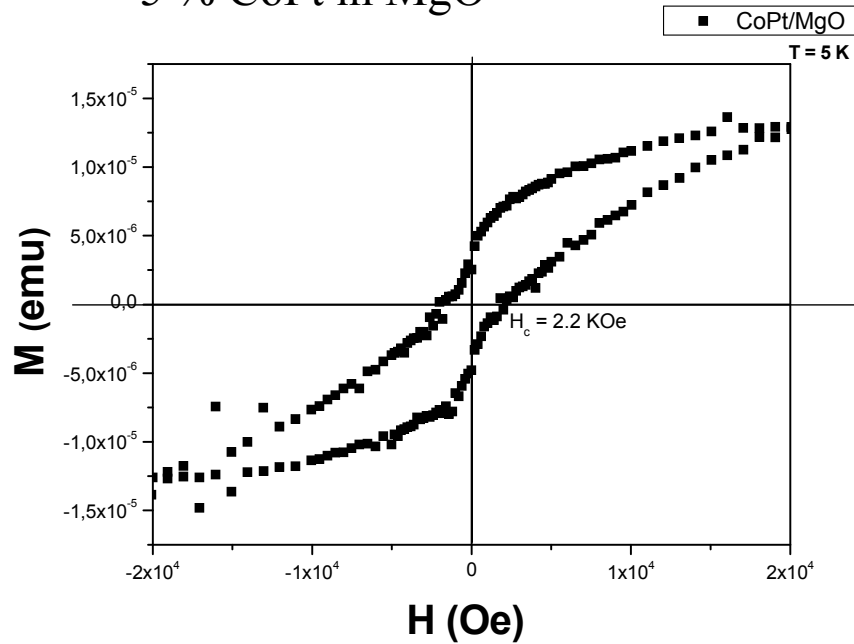
Cf Poster:

«Structural and magnetic properties of $\text{Co}_x\text{Pt}_{1-x}$ mixed clusters... »

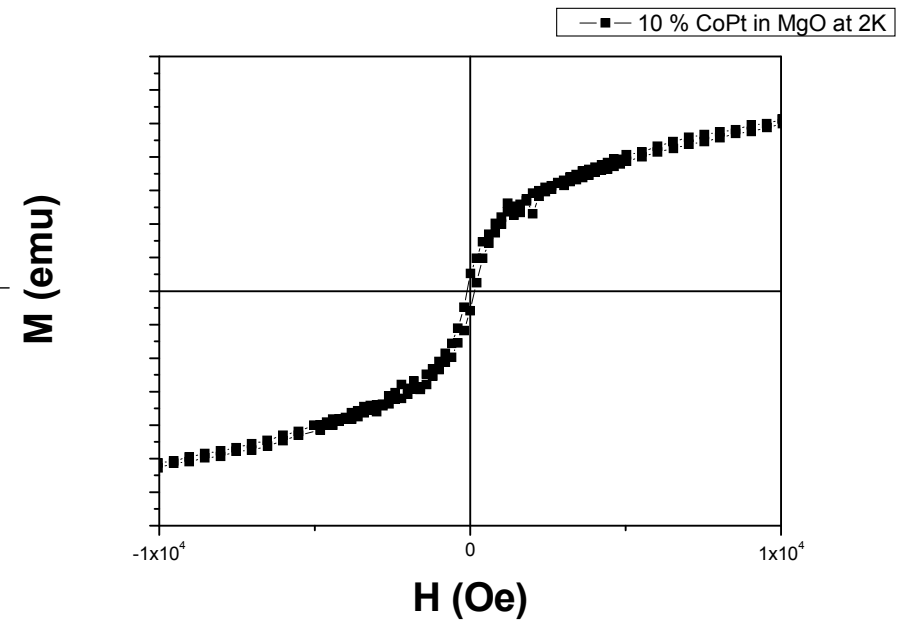
Stanislas Rohart, Cécile Raufast et al.

Magnetization of CoPt clusters in MgO versus concentration

5 % CoPt in MgO



10 % CoPt in MgO



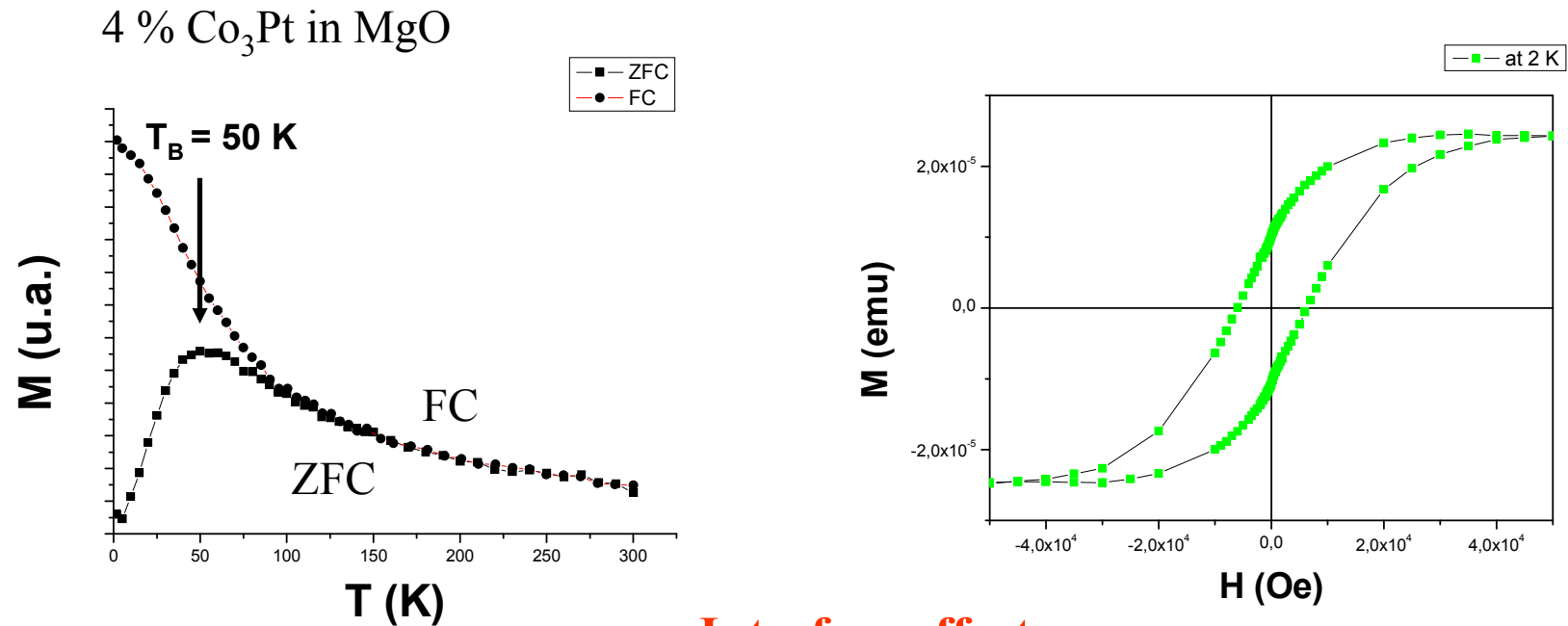
MgO	H_C (at 2K)	T_B
5% CoPt	2.2 kOe	23 K
10% CoPt	750 Oe	38 K

$T_B \nearrow$ and $H_C \searrow$ versus CoPt concentration \nearrow

\Rightarrow Dipolar interactions between clusters \nearrow
(~ Super-cluster)

Inter-particles effects

Magnetization of diluted $\text{Co}_x\text{Pt}_{1-x}$ clusters in MgO matrix



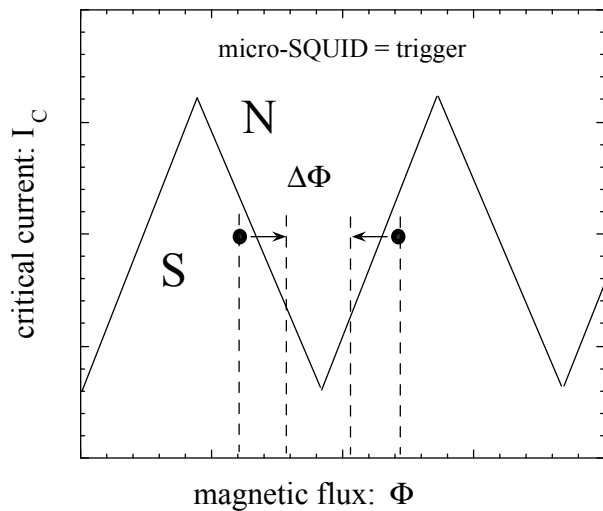
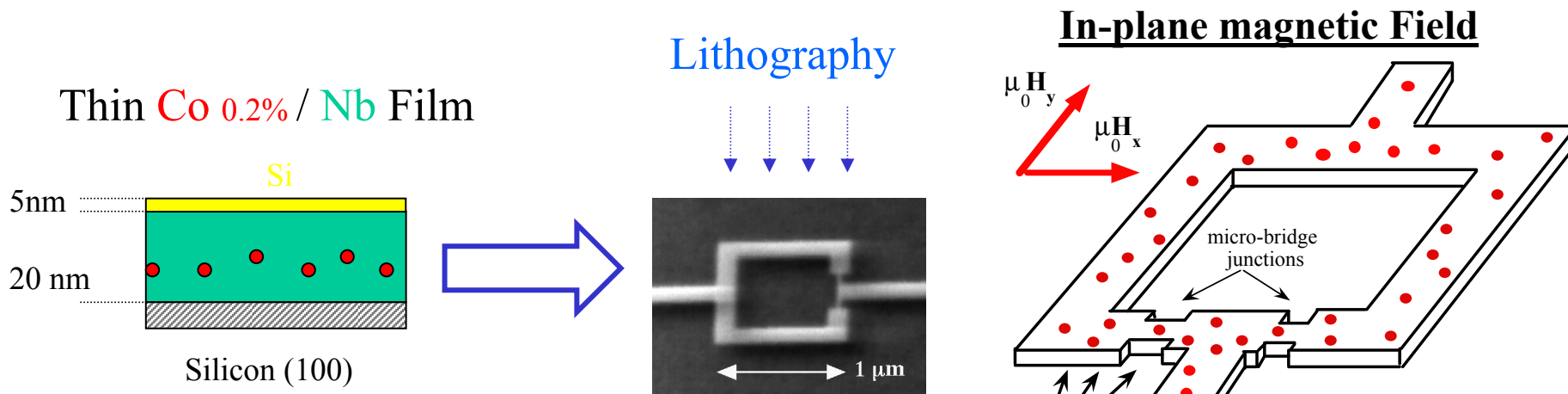
Interface effects

in MgO	H_C (at 2K)	$T_B \propto K D^\alpha$	Structure and diameter
5% CoPt	2.2 kOe	23 K	CoPt fcc : 2 nm
4% Co_3Pt	6 kOe	50 K	Core CoPt + Shell Co : 2 nm
2.5% Co	5 kOe	87 K	Co fcc : 3 nm

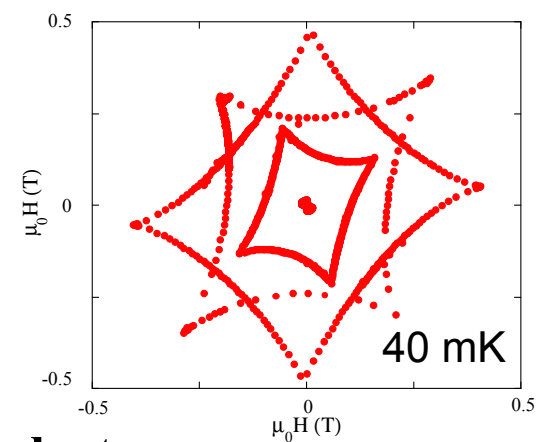
in Progress...

- **Synthesis, Structure and Superparamagnetism
on Assembled clusters films**
- **Measurements one single nanoparticle**

MicroSQUID measurements on single clusters



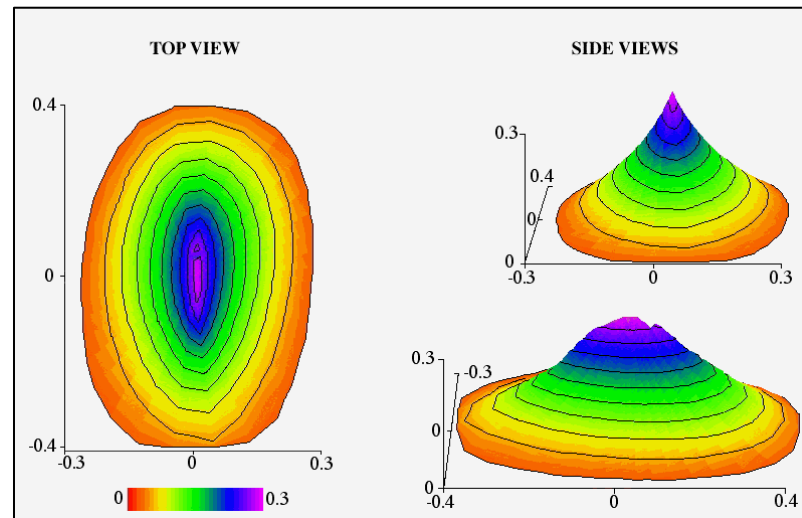
Cold mode



**3 Co-clusters
in the micro-bridge**

Anisotropy energy:

$$G(\vec{m}) = K_1 m_z^2 + K_2 m_y^2 + K_4 (m_x'^2 m_y'^2 + m_x'^2 m_z'^2 + m_y'^2 m_z'^2) + K_6 m_x'^2 m_y'^2 m_z'^2 + \dots$$



From 3 D Astroid simulations : Experimental anisotropy constants

$$K_1 = -2.1 \cdot 10^5 \text{ J/m}^3$$

$$K_2 = 0.8 \cdot 10^5 \text{ J/m}^3$$

$$K_4 = -0.1 \cdot 10^5 \text{ J/m}^3$$

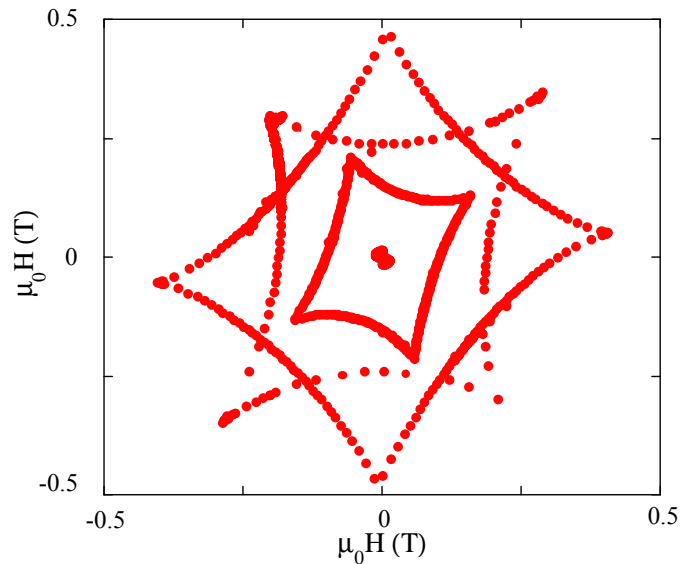


**Second order terms dominate for pure Co clusters :
bi-axial Surface Anisotropy**

M. Jamet, W. Wernsdorfer et al. PRL (2001)

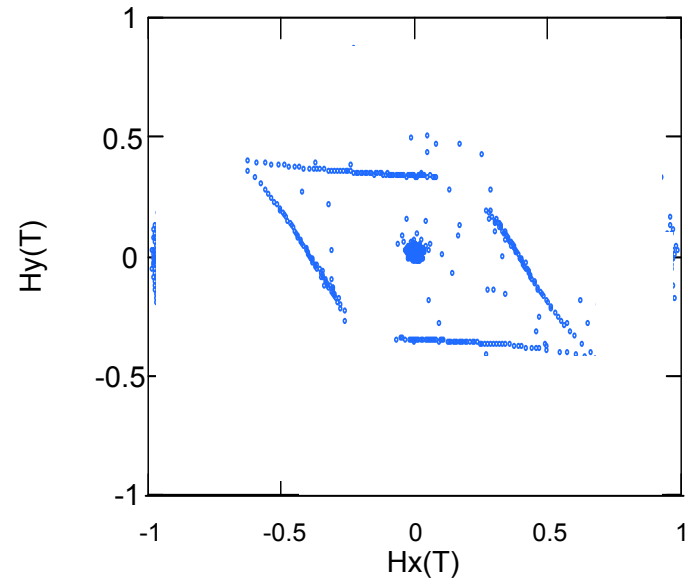
Comparative 2D astroid on Co and CoPt clusters

Three Co clusters in Nb



M. Jamet et al. Phys. Rev. Lett. 86 (2001) 4676

One single CoPt cluster in Nb



C. Raufast et al. (2005) unpublished

With equivalent Blocking Temperature : $T_B \sim 12$ K

For $\varnothing_{Co} \sim 3$ nm and $\varnothing_{CoPt} \sim 2$ nm,

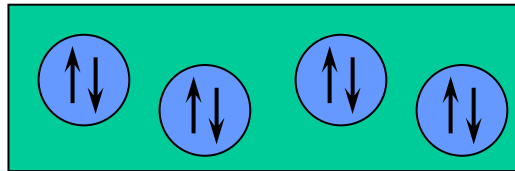
$H_{sw} (CoPt) \gg H_{sw} (Co)$

Thus **$K_{CoPt} (in Nb) > K_{Co} (in Nb)$**

Proximity Effects : F cluster in S matrix

Superconductivity/Ferromagnetism: Two antagonist orders of Spin

Cooper pairs



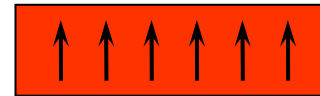
Type II Superconductor (Nb)

Coherence length:

$$\xi_S \approx 30 \text{ nm}$$

Order temperature:

$$T_{\text{critical}} \approx 9 \text{ K}$$



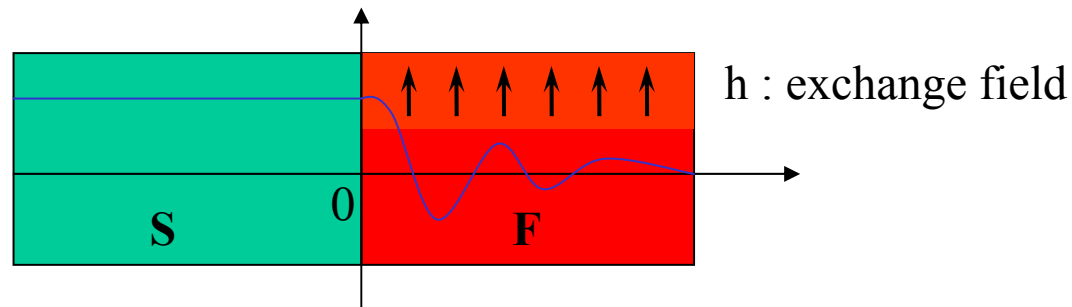
Ferromagnetic metal (Co)

$$\xi_F \approx 2 - 4 \text{ nm}$$

$$T_{\text{Curie}} \approx 1388 \text{ K}$$

→ **Coexistence: Mutual Influence on Electronic properties ?**

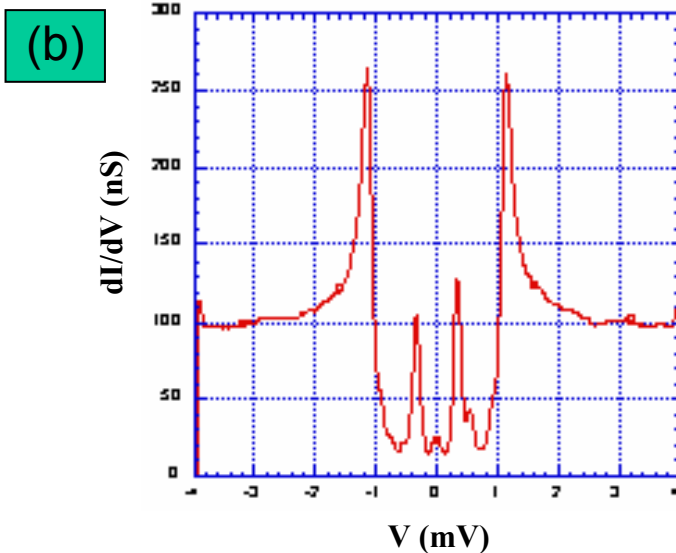
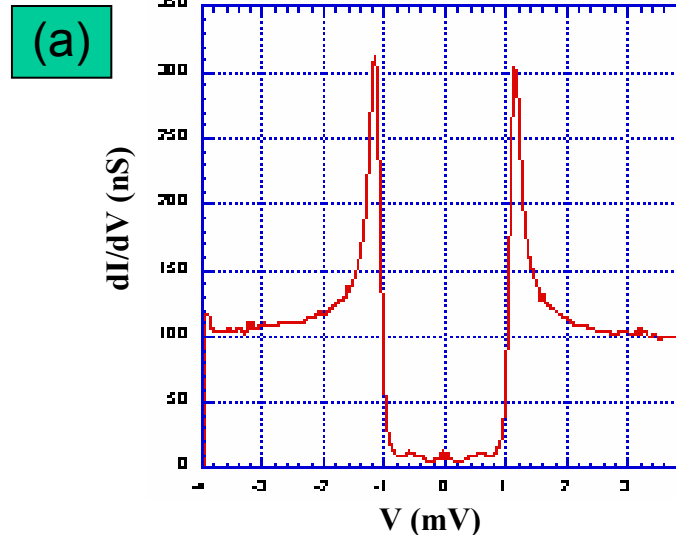
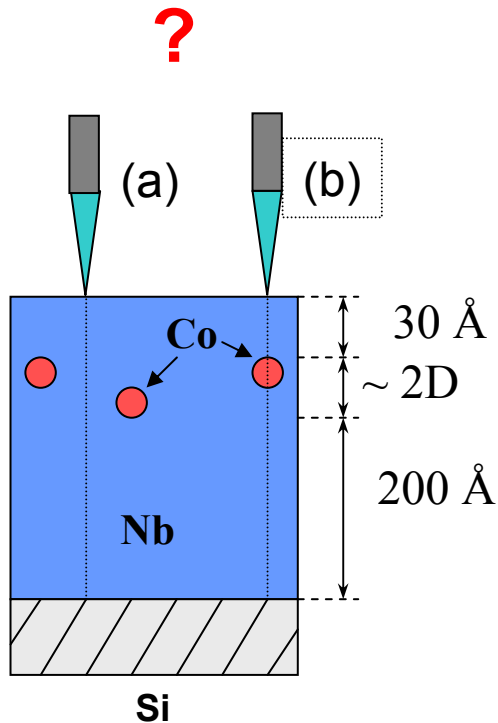
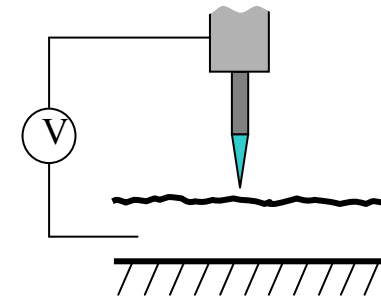
Superconductivity order parameter on the S/F interface



The cooper pair entering in the F region becomes an oscillating evanescent state

Local density of State

Very low temperature STM measurements



Density of state of Nb :
Superconducting gap

$$2\Delta \approx 2 \text{ meV}$$

$$T_{\text{mes}} = 60 \text{ mK}$$

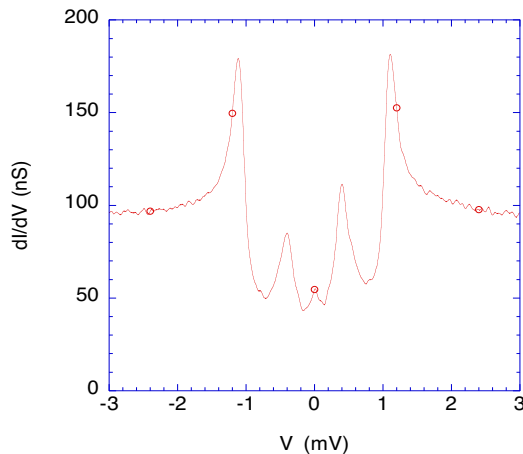
Asymmetric peaks
in S gap to correlate
with F clusters

STREP *SFINX*
(2004-2007)

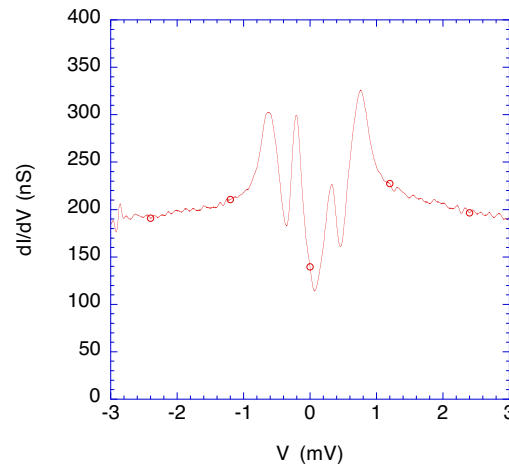
Perspectives : Theoretical and experimental works to explain S/F proximity effects

Systematic and reproducible experiments

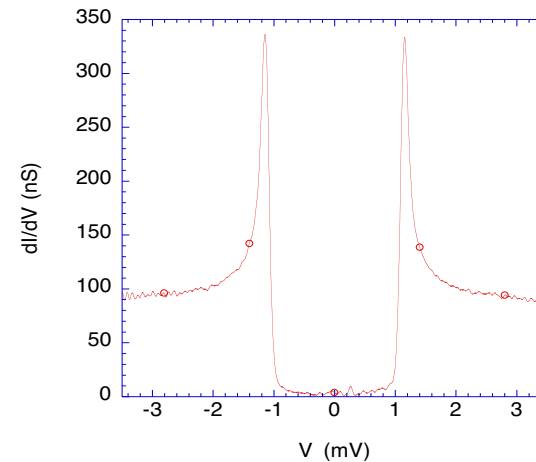
(Co) clusters in Nb
(150 clusters/ μm^2)



(Co) clusters in Nb
(12000 clusters/ μm^2)



(Ag) clusters in Nb
(370 clusters/ μm^2)



Improvement in Matrix crystallinity and Monodisperse clusters

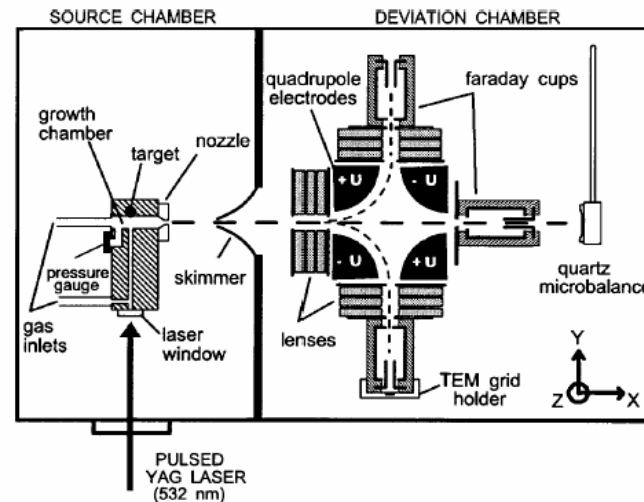
New UHV M.B.E chamber
(Heating substrate, RHEED,...)

UHV Transfer suitcase
(Compatible with very low T STM)

and



Since October 2005 :
SFINX Post doc
Stanislas Rohart



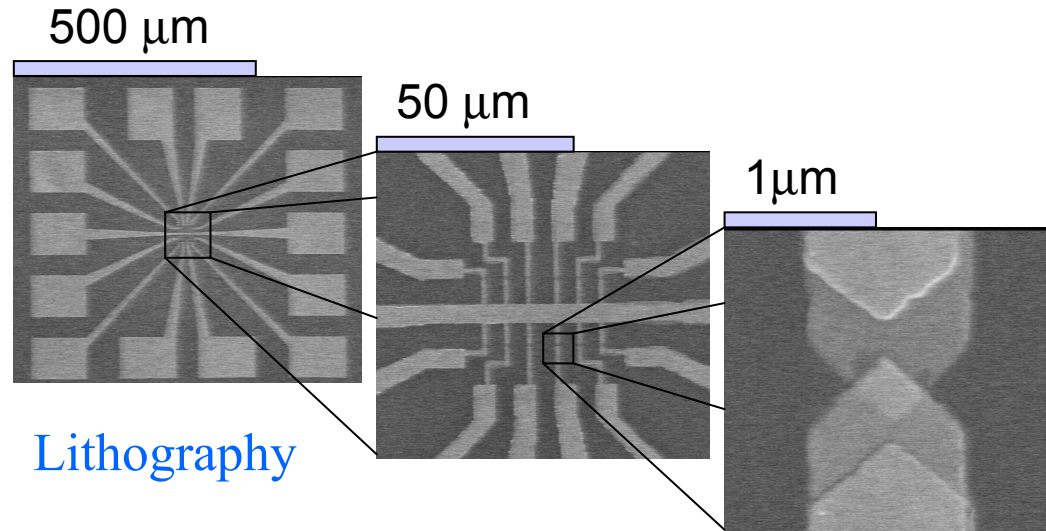
**Electrostatic
quadrupole deviator:**

$$\Delta d / \langle d \rangle = 25\% \rightarrow 5\%$$

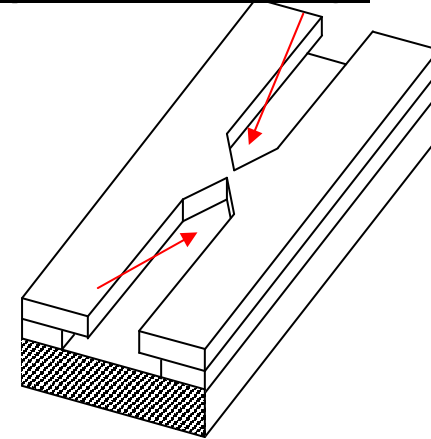
Rev. Sc. Instrum. **75**, 2461 (2004)

Magnetic cluster embedded in MgO matrix

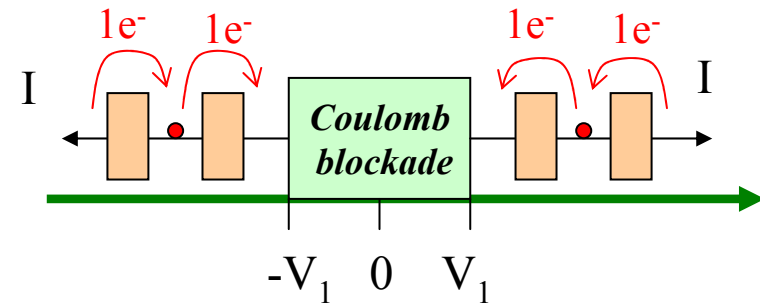
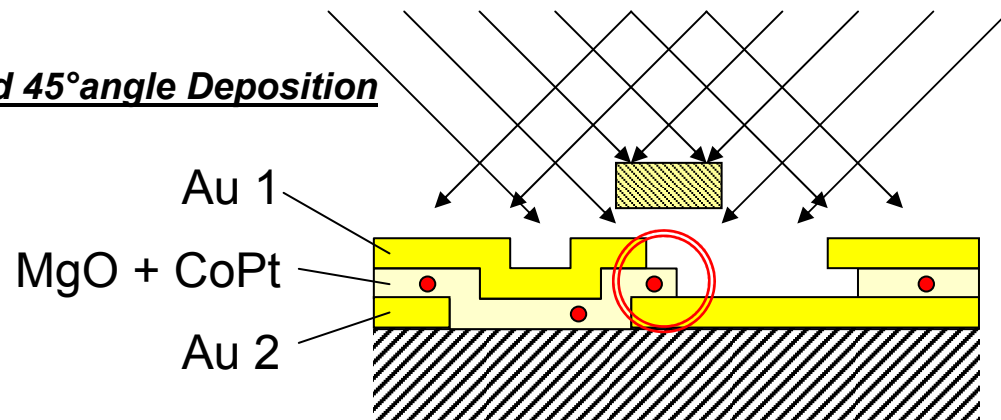
Tunnel junction with one CoPt cluster



Using a Suspended nano-bridge

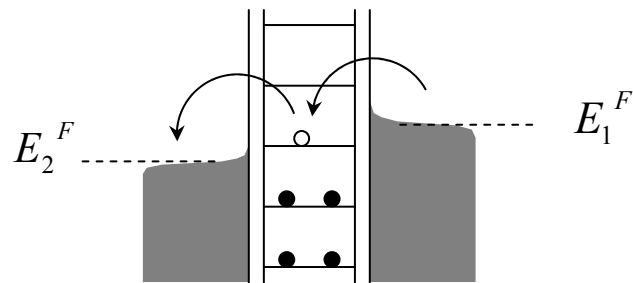


And 45° angle Deposition



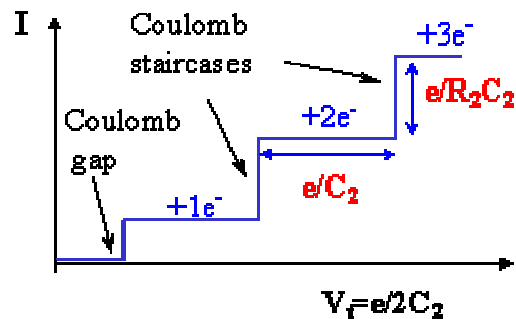
Spin-dependent tunneling and Coulomb Blockade

Principle



Coulomb Blockade

$$E = E_0 + \frac{e^2}{2C}$$

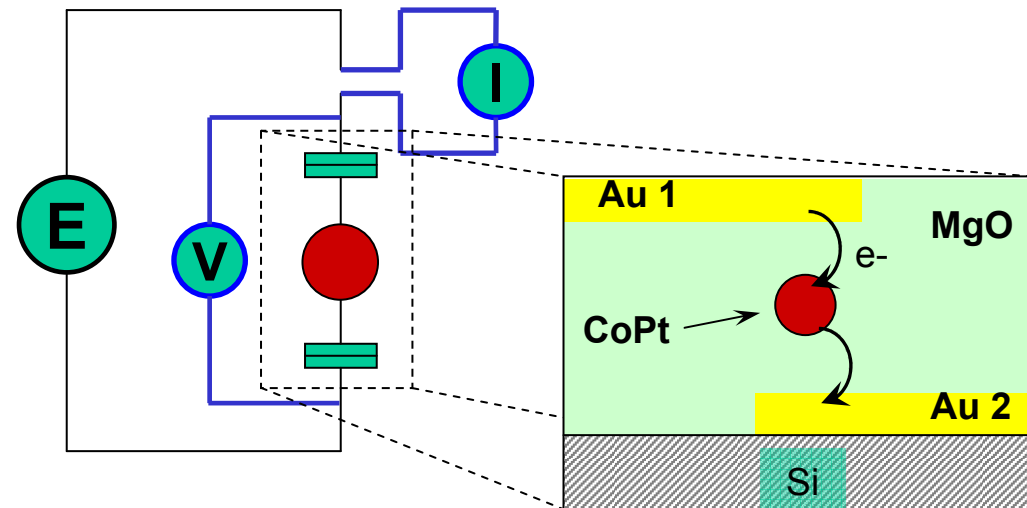


Charge Quantization

Nano-junction

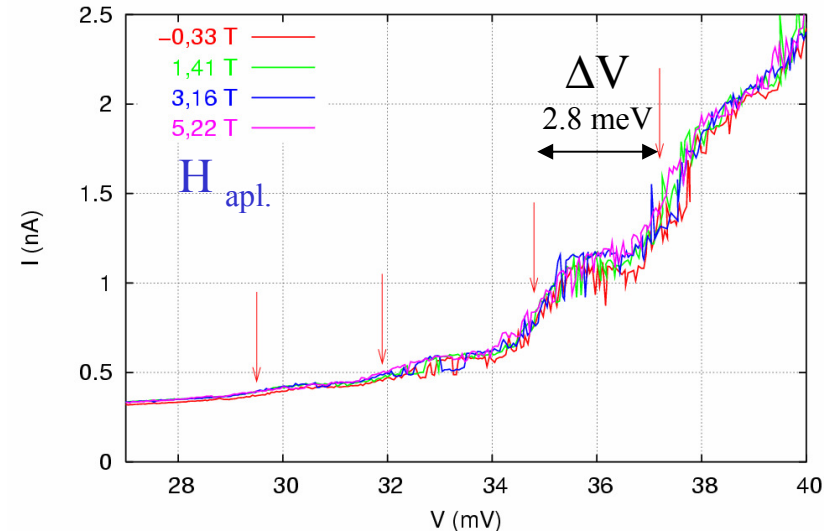
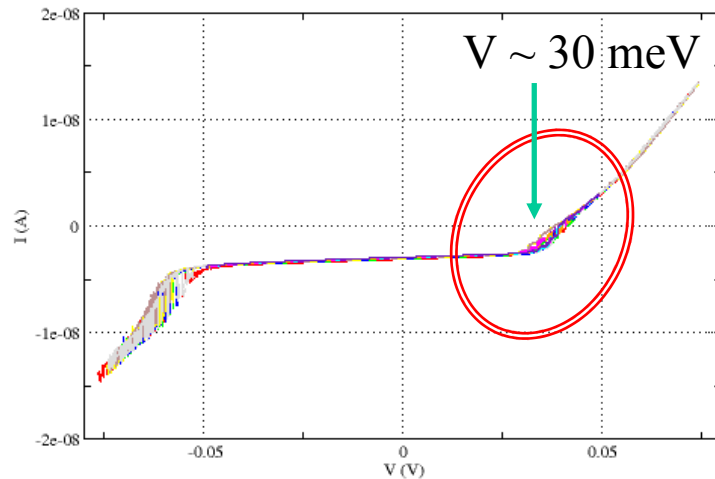
Charge transfer of one single e^- from Au_1 to Au_2 electrode via one CoPt cluster in MgO insulator

Measurement $I = f(V, H)$



For a fixed bias and various external magnetic fields, energy levels must be spin-dependent \rightarrow magnetic properties of one cluster

Quantum size effect but Spin-independent tunneling



Coulomb blockade :
Energy Step due to
Tunnelling via one cluster

*in progress with
Cécile Raufast' thesis...*

Zoom on the first Energy Step
- Elementary Energy Step due to
Tunnelling via discrete electronic levels
in one 2nm-diameter CoPt cluster (300
atoms)

- But **Independent on** applied magnetic
Fields H_{apl} for CoPt cluster in MgO:
nonmagnetic cluster?

Aknowledgments

Main results from the M. Jamet (2001) and L. Favre (2004) Thesis, Lyon, France

In progress, results from the C. Raufast PhD thesis and S. Rohart SFINX post Doc

Close collaborations:

- micro-squid and tunnel junction measurements (W. Wernsdorfer and E. Bonet, LLN Grenoble, France)*
- density of state STM measurement at very low temperature (H. Courtois, CRTBT Grenoble, France)*
- Tunneling Magneto-transport measurement (C. Marrow, Leeds, UK)*