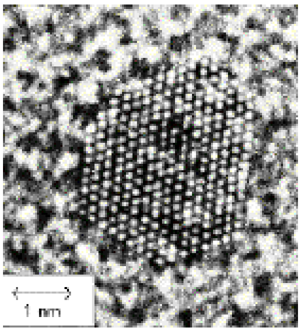
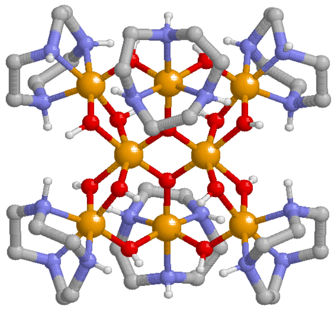
Magnetization reversal in nanostructures:
Quantum dynamics in nanomagnets and single-molecule magnets



S = 10^2 to 10^6

Wolfgang Wernsdorfer,
Laboratoire de Magnétisme Louis Néel
C.N.R.S. - Grenoble



S = 1/2 to ≈ 30

CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE

GRENOBLE 1 UNIVERSITE JOSEPH FOURIER

Mesoscopic physics in magnetism

macroscopic *nanoscopic*

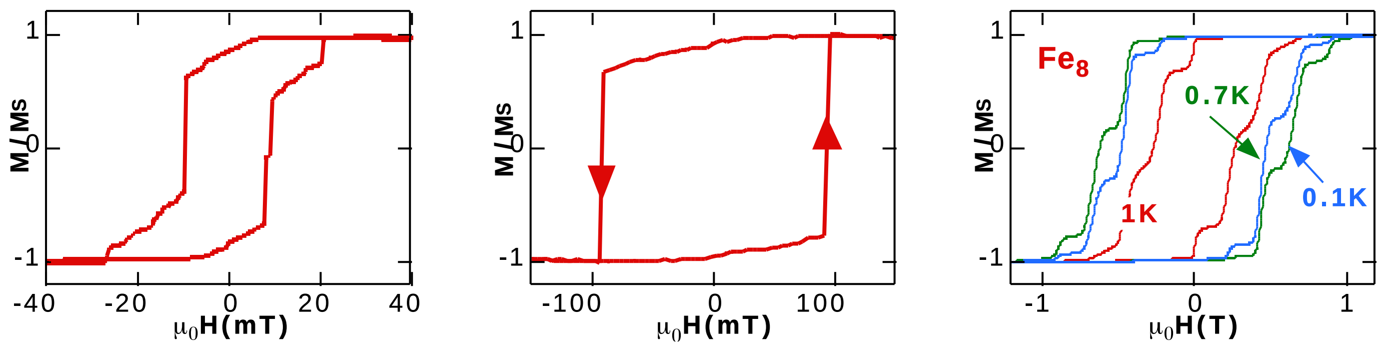
permanent magnets	micron particles	nanoparticles	clusters	molecular clusters	individual spins

S = 10^{20} 10^{10} 10^{8} 10^{6} 10^{5} 10^{4} 10^{3} 10^{2} 10 1

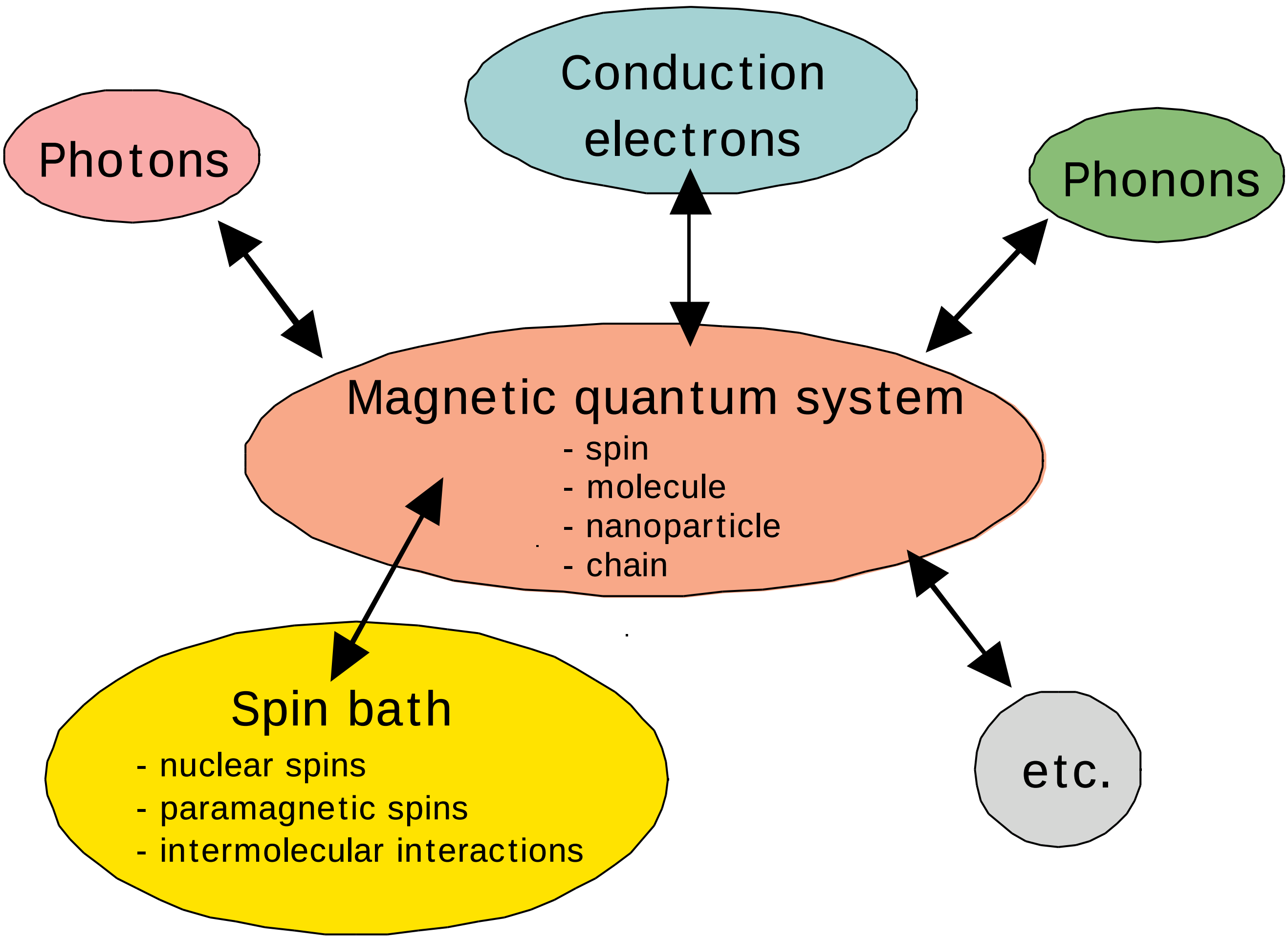
multi - domain
nucleation, propagation and annihilation of domain walls

single - domain
uniform rotation
curling

giant spin
quantum tunneling,
quantization
quantum interference



Interactions in magnetic mesoscopic systems



Outline

Part I: *classical magnetism*

1. Magnetization reversal by uniform rotation
   (Stoner-Wohlfarth model)
   - theory
   - experiment (3 nm Co clusters)
2. Influence of the temperature on the magnetization reversal
   (Néel-Brown model)
3. Magnetization reversal dynamics
   (Landau-Lifshitz-Gilbert)
   - magnetization reversal via precession
   - dynamical astroid

Outline

Part II: *quantum magnetism*

1. A simple tunnel picture
    - Giant spin model
    - Landau Zener tunneling
    - Spin parity
    - Berry phase

2. Interactions with the environment
    - Intermolecular interactions
    - Interaction with photons

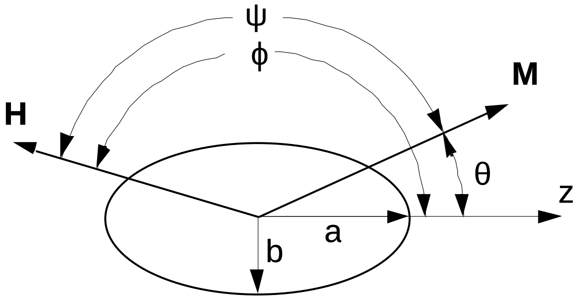
Conclusion

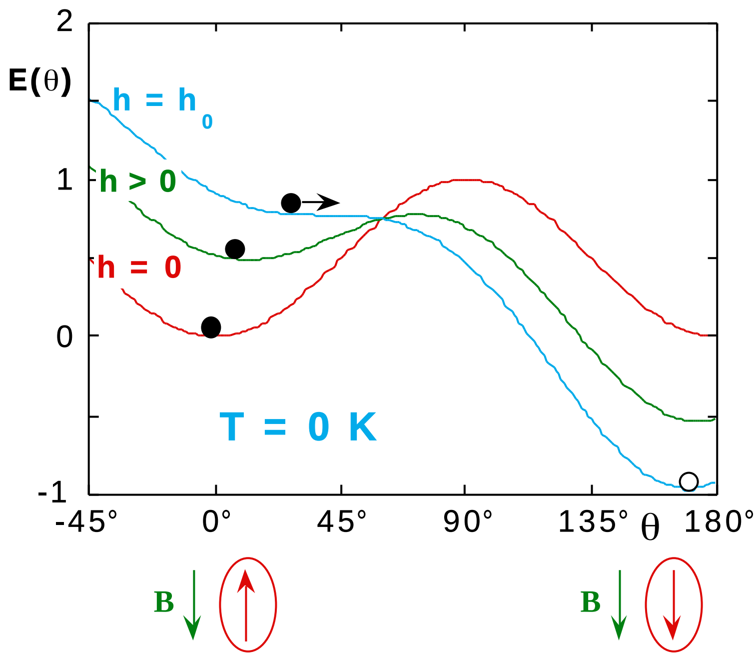
Uniform rotation of magnetization:
Stoner -Wohlfarthmodel (1949)

- **single domain magnetic particle**
- **one degree of freedom:** orientation θ of magnetization M
- **potential:**

$$E = K \sin^2\theta - \mu_0 M_S H \cos(\theta - \phi)$$

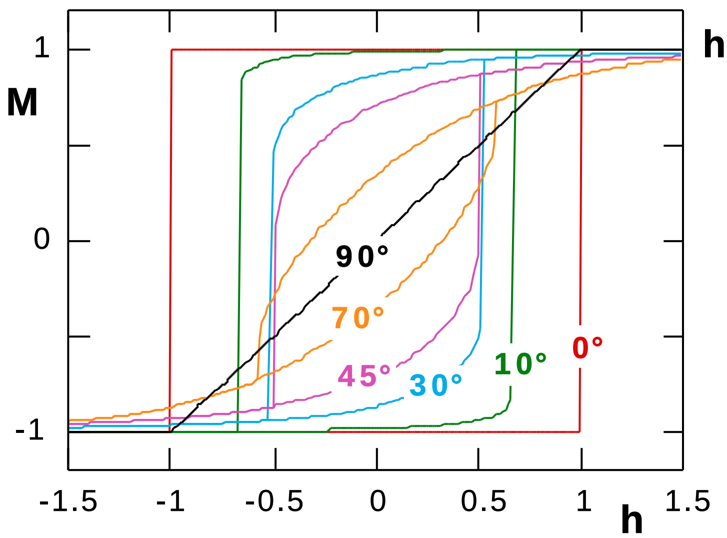
$$K = K_1 + \frac{1}{2}\mu_0 M_S^2 (N_b - N_a)$$

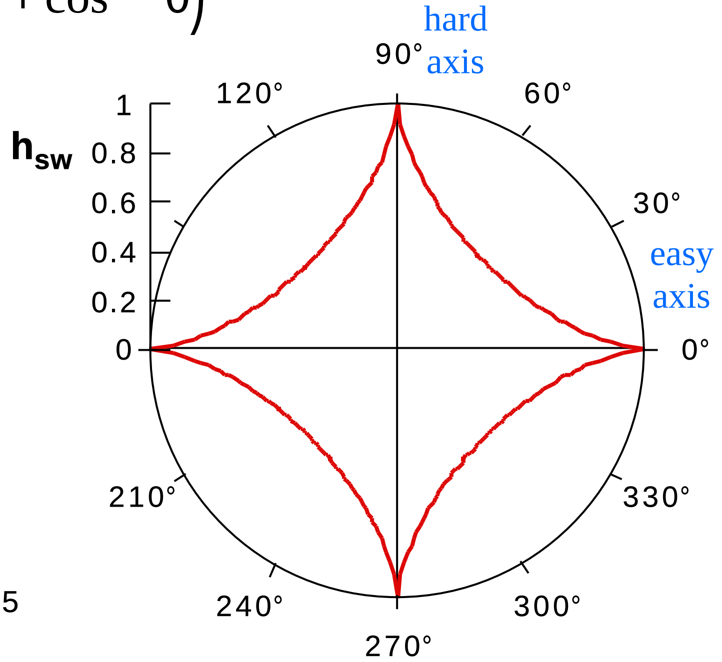




Stoner - Wohlfarth switching field

$$h_{sw} = \left(\sin^{2/3}\theta + \cos^{2/3}\theta\right)^{-3/2}$$





Stoner - Wohlfarth astroid

Generalisation of the Stoner - Wohlfarth model

- **from 2D to 3D**
- **for arbitrary anisotropy functions**

André Thiaville, JMMM **182**, 5 (1998)
PRB **61**, 12221 (2000)

- **potential St-W:** $E = K \sin^2\theta - \mu_0 M_S H \cos(\theta - \phi)$

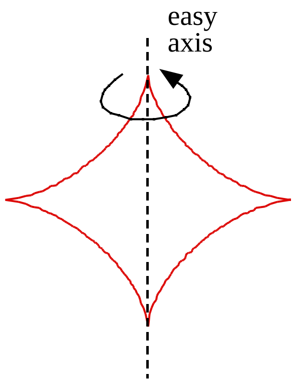
- **potential 3D:** $E = E_0\left(\theta, \varphi\right) - \mu_0 \vec{M}\vec{H}$

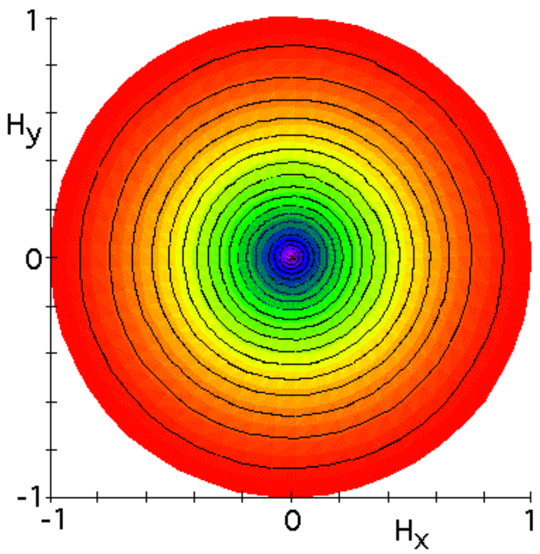
$$E = E_0\left(\vec{m}\right) - \mu_0 M_S \vec{m}\vec{H}$$

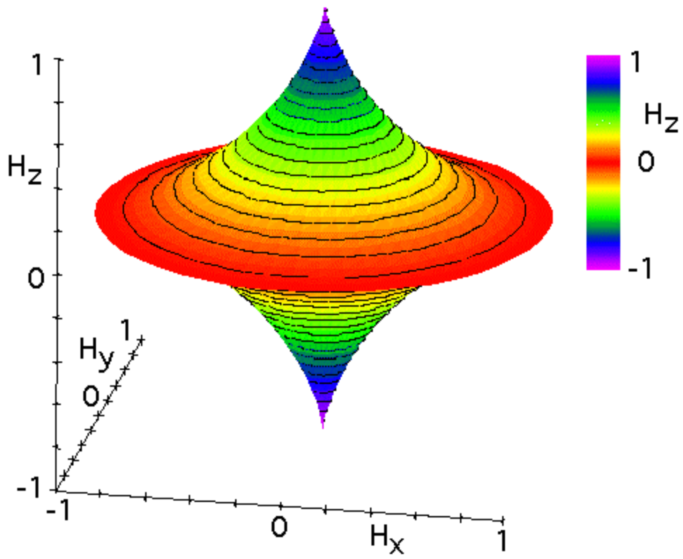
$$E_0(\vec{m}) = E_{shape}(\vec{m}) + E_{cristal}(\vec{m}) + E_{surface}(\vec{m}) + E_{mag.elastic}(\vec{m})$$

Switching fields of uniaxial anisotropy

$$E_0 = -m_z^2$$

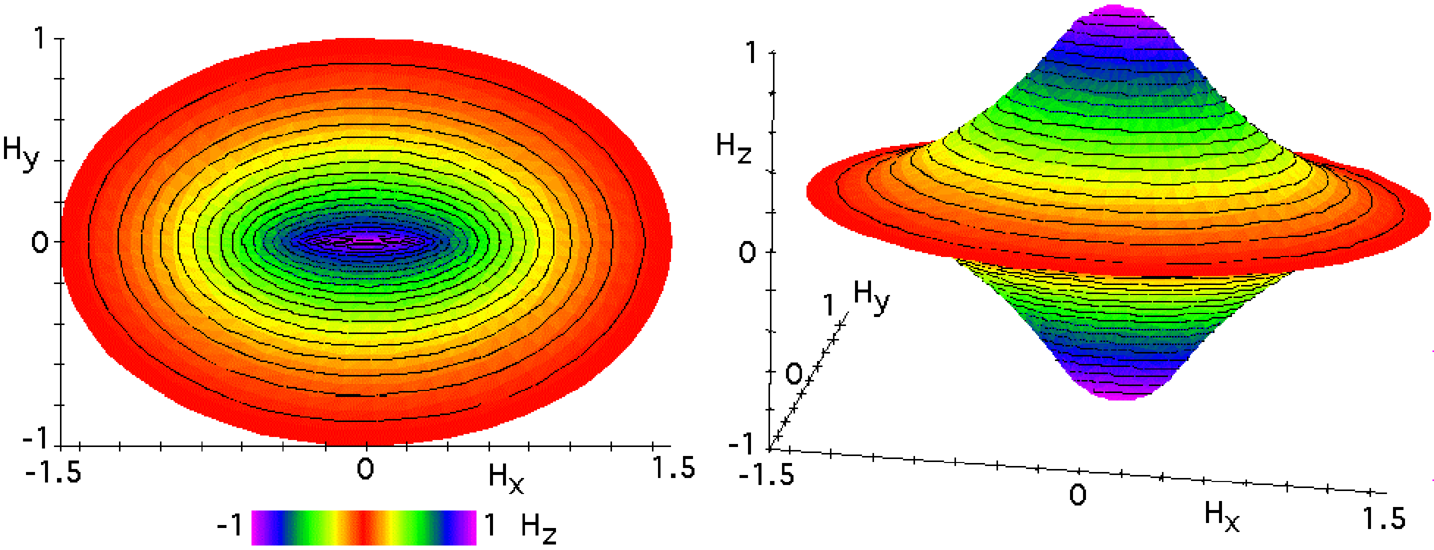






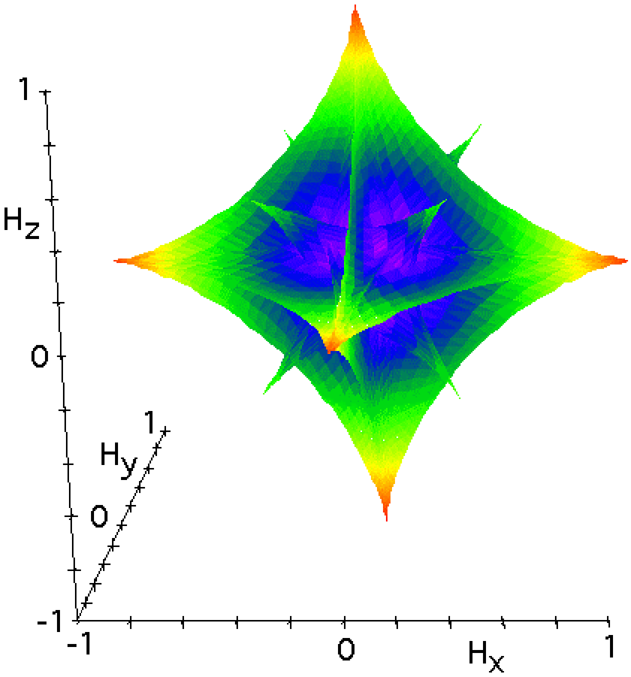
Switching fields of biaxial anisotropy

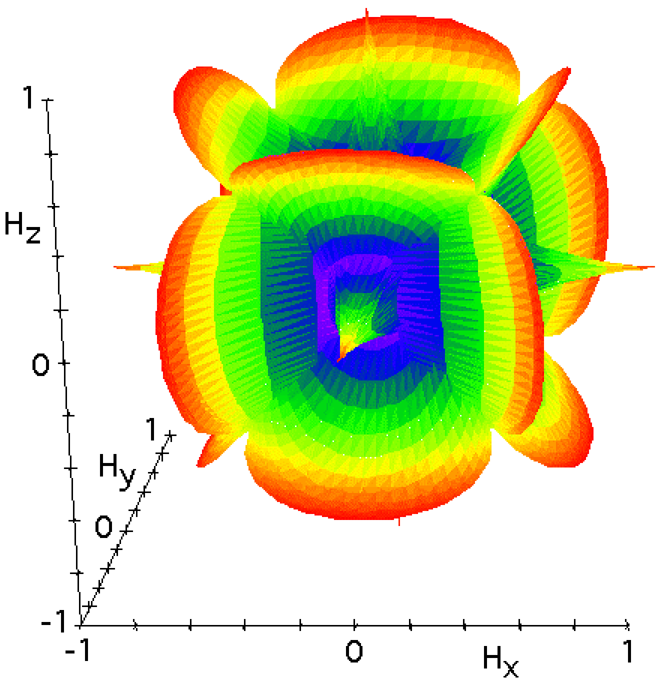
$$E_0 = -m_z^2 + 0.5m_x^2$$



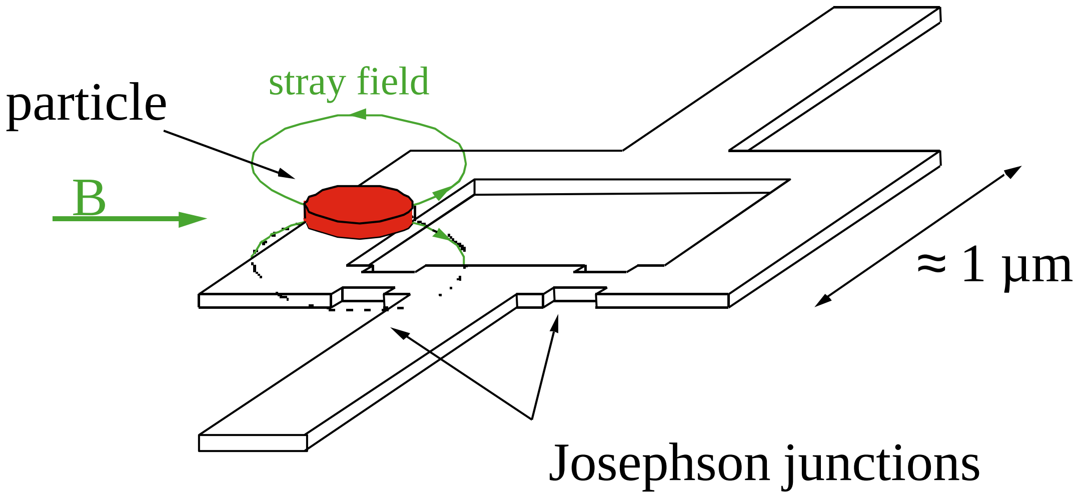
Switching fields of cubic anisotropies

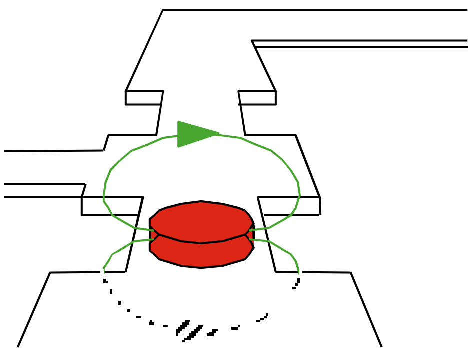
$$E_0 = -\left[m_x^2 m_y^2 + m_x^2 m_z^2 + m_y^2 m_z^2\right] \qquad E_0 = +\left[m_x^2 m_y^2 + m_x^2 m_z^2 + m_y^2 m_z^2\right]$$





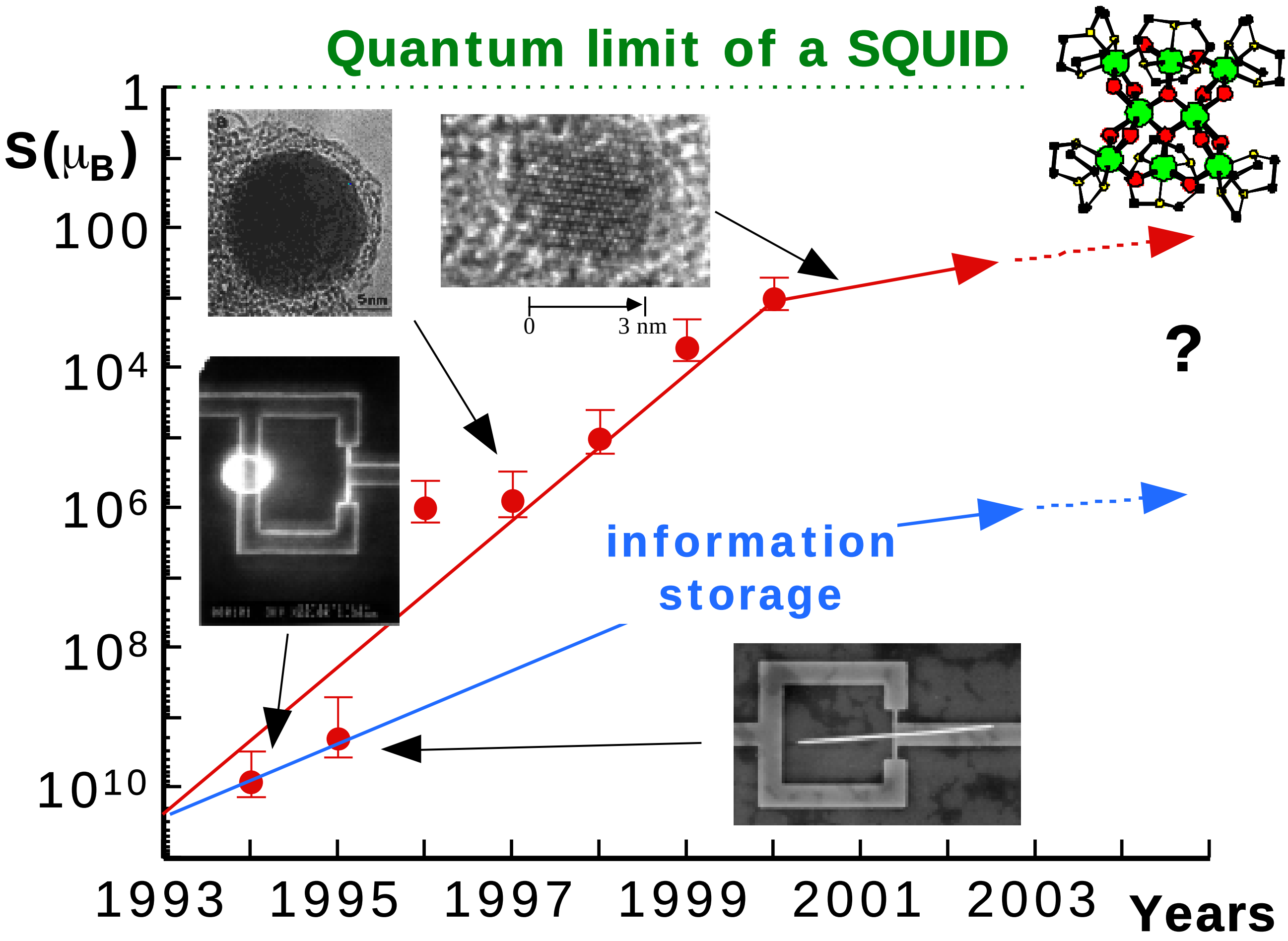
Micro-SQUID magnetometry





- **fabricated by electron beam lithography**
  (D. Mailly, LPM, Paris)
- **sensitivity :** $10^{-4}\ \Phi_o$
  $\approx$ $10^2 - 10^3\ \mu_B$ **i.e. (2 nm)^3 of Co**
  $\approx$ $10^{-18} - 10^{-17}$ **emu**

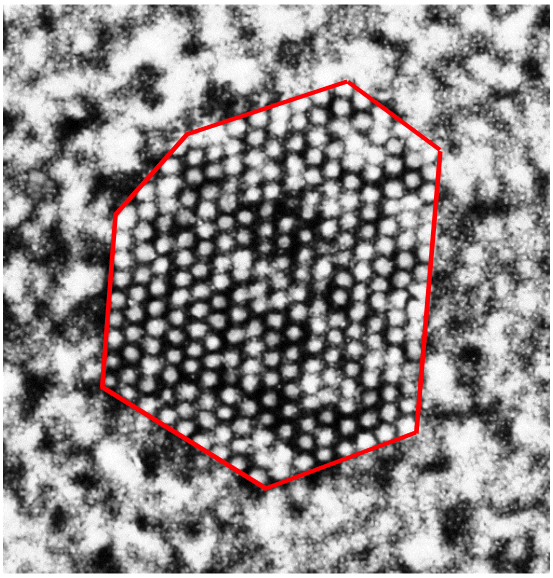
Roadmap of the micro-SQUID technique



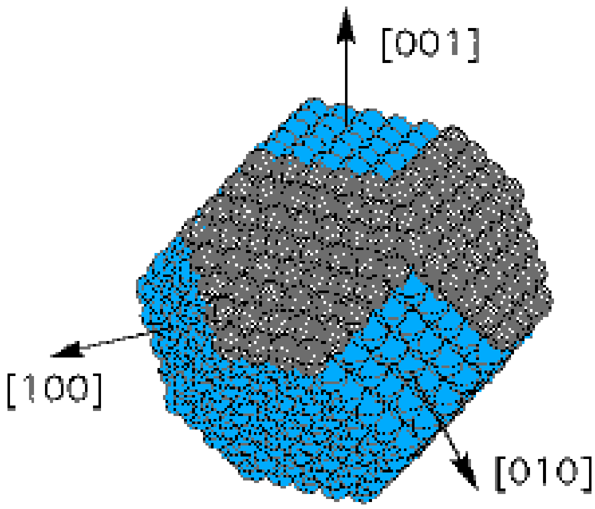
3 nm cobalt cluster

DPM - Villeurbanne: LASER vaporization and inert gas condensation source

Low Energy Cluster Beam Deposition regime



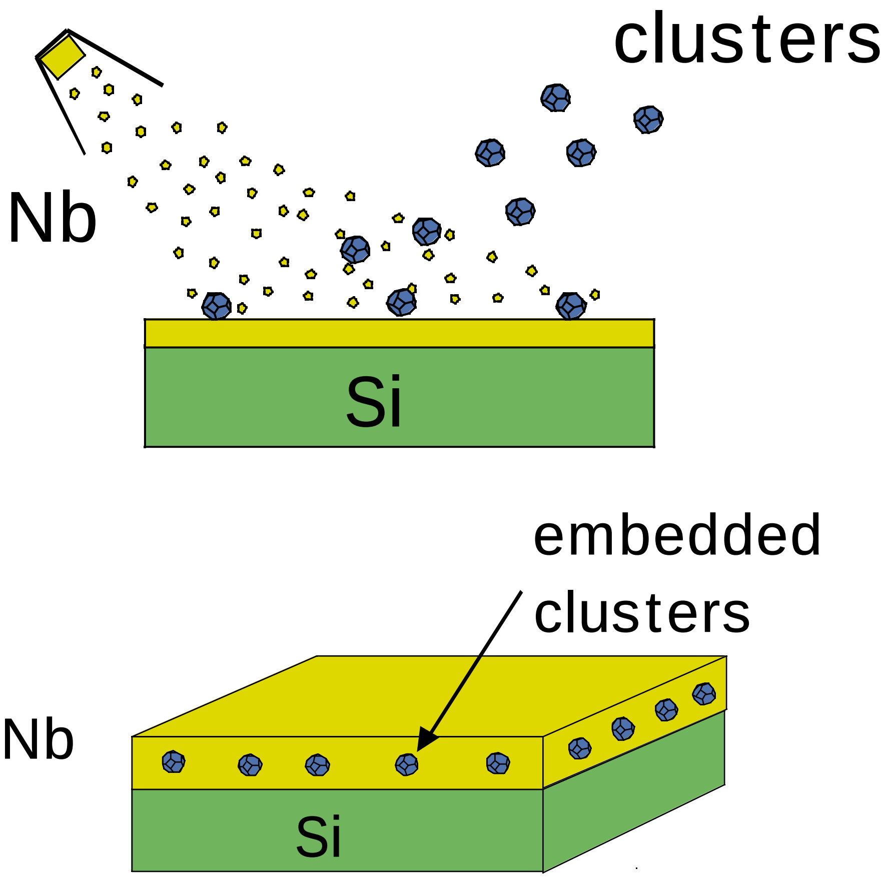
HRTEL along a [110] direction
fcc - structure, faceting



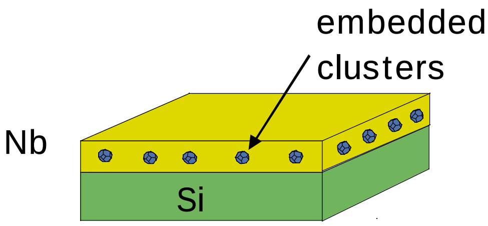
blue: 1289-atoms truncated octahedron
grey: added atomes, total of 1388 atomes

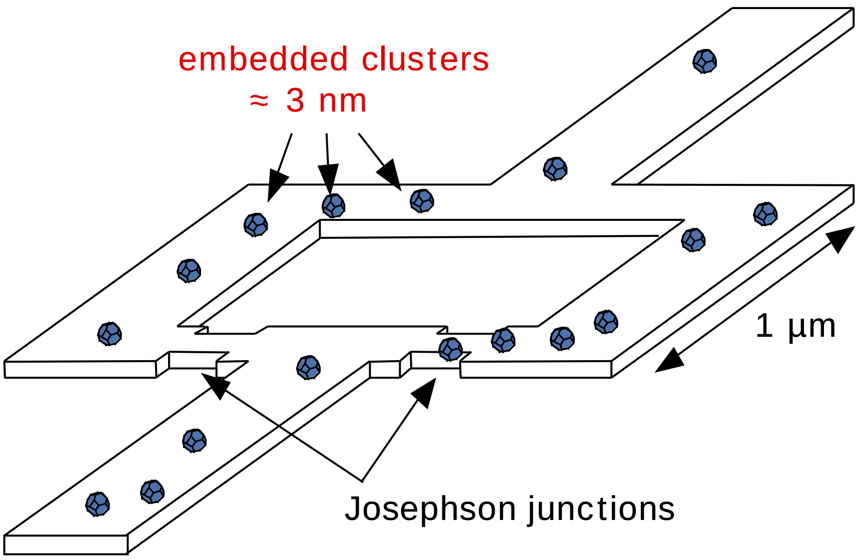
Ideal case: truncated octagedron with 1289 or 2406 atoms for diameters of 3.1 or 3.8 nm

Low energy cluster beam deposition



Micro-SQUID magnetometry





SQUID is fabricated by electron beam lithography

D. Mailly, LPN-CNRS

sensitivity : ≈ **10^2 - 10^3 μ_B** i.e. $(2\ nm)^3$ of Co

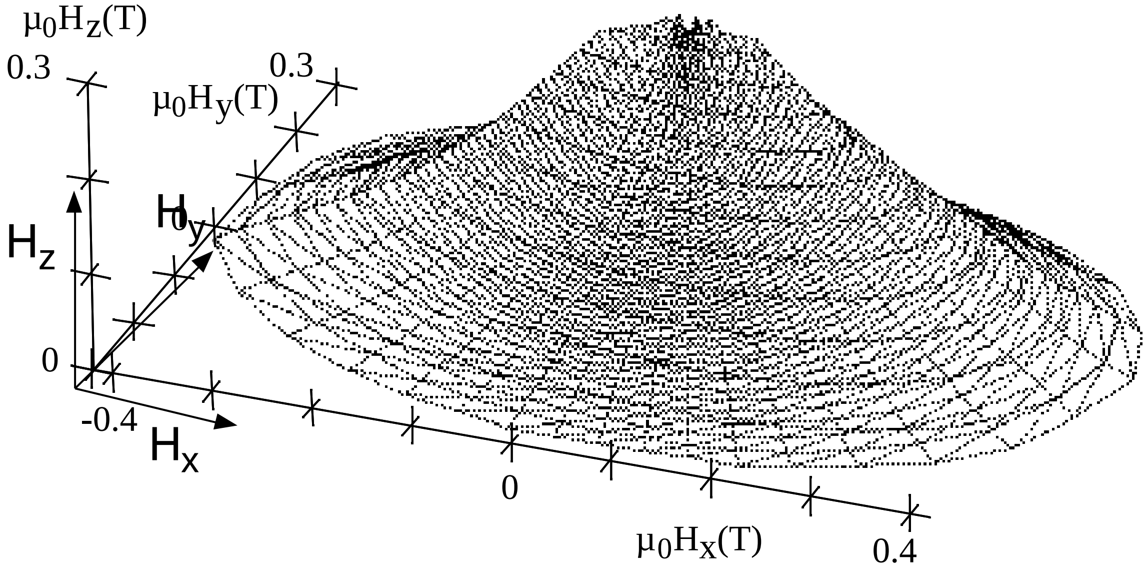
i.e. ≈ 10^{-18} - 10^{-17} emu

clusters in Nb - matrix

M. Jamet, V. Dupuis, A. Perez, DPM-CNRS, Lyon

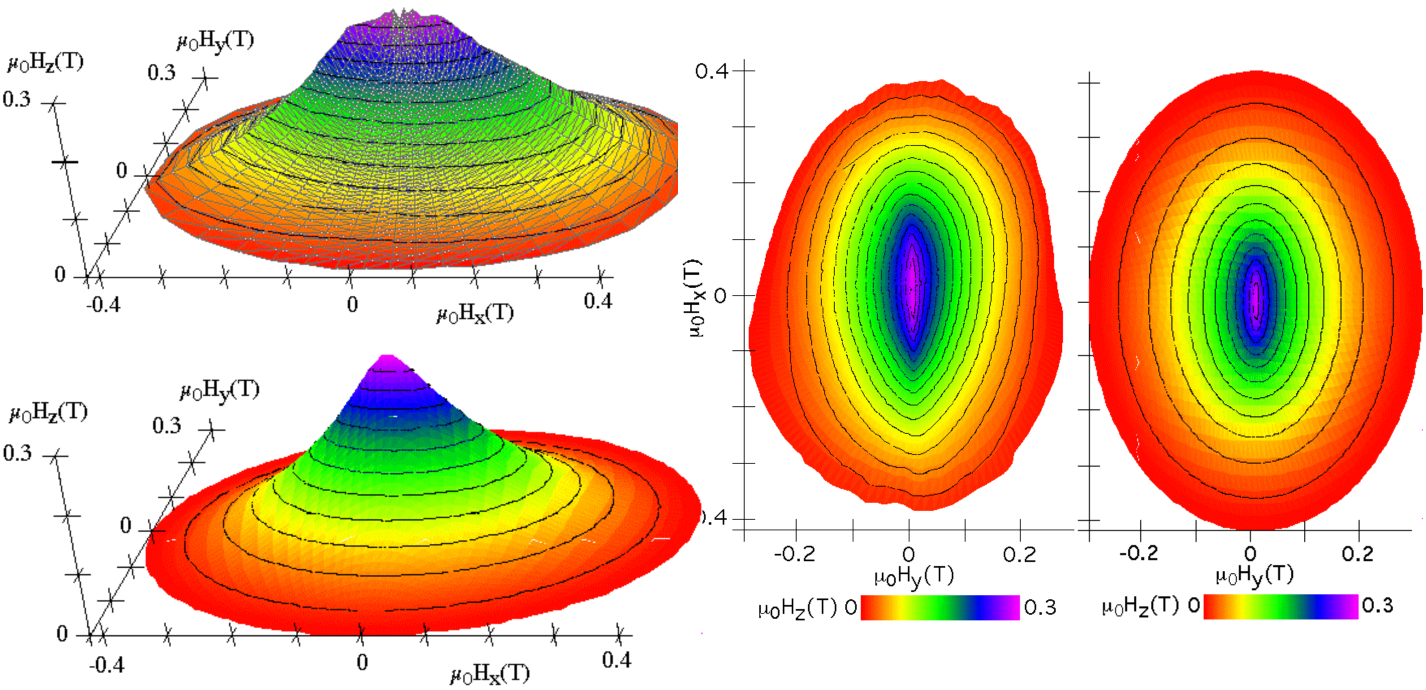
Acknowledgment: B. Pannetier, F. Balestro, J.-P. Nozières

3D switching fields of a 3nm Co cluster



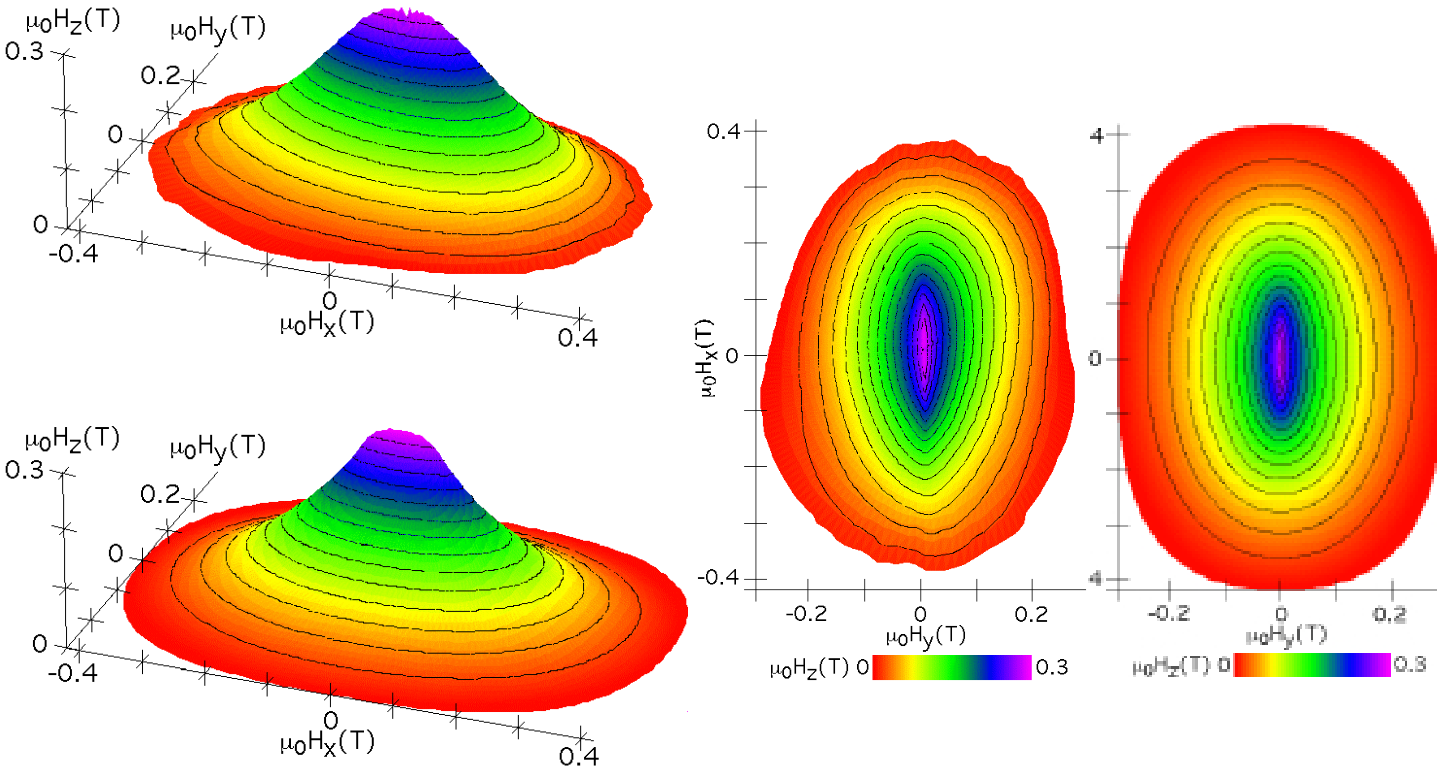
Finding the anisotropy function from 3D switching field measurements

$$H = -m_z^2 + 0.4 m_x^2$$



Finding the anisotropy from **3D switching field measurements**

$$E_0 = -m_z^2 + 0.4 m_x^2 - 0.1\left[m_x^2 m_y^2 + m_x^2 m_z^2 + m_y^2 m_z^2\right]$$



Magnetic anisotropy energy

$$E_0(\vec{m}) = E_{shape}(\vec{m}) + E_{cristal}(\vec{m}) + E_{surface}(\vec{m}) + E_{mag.elastic}(\vec{m})$$

$$E_{experiment}/v = -K_1\, m_z^2 + K_2\, m_x^2 \qquad K_1 \approx 2.5 \times 10^5\, J/m^3 \quad \left(2.5\times10^6\, erg/cm^3\right)$$

$$K_2 \approx 0.7 \times 10^5\, J/m^3 \quad \left(0.7\times10^6\, erg/cm^3\right)$$

$$E_{shape}/v = -K_{||}\, m_z^2 + K\quad m_x^2 \qquad K_{||} \approx K \quad \approx 0.1 - 0.3 \times 10^5\, J/m^3$$

$$E_{cristal}/v = -K_{cristal}\left(m_x^2 m_y^2 + m_x^2 m_z^2 + m_y^2 m_z^2\right) \quad K_{cristal}^{Co-bulk} \approx 1.2 \times 10^5\, J/m^3$$

however the measured 4th order is much smaller

$$K_{mag.elastic} < 0.1 \times 10^5\, J/m^3$$

$E_{surface}$

seems to be the main contribution to the magnetic anisotropy

Model systems for studying the magnetisation reversal

Quasi-static measurements

• hysteresis loops

• switching fields (angles)

Dynamical measurements

• relaxation (T,t)

• switching fields (T,dH/dt)

• switching probabilities (T,t)

Magnetic anisotropy

• forme

• crystal

• surface

• • •

Activation volume

Damping factor

• material

• defects

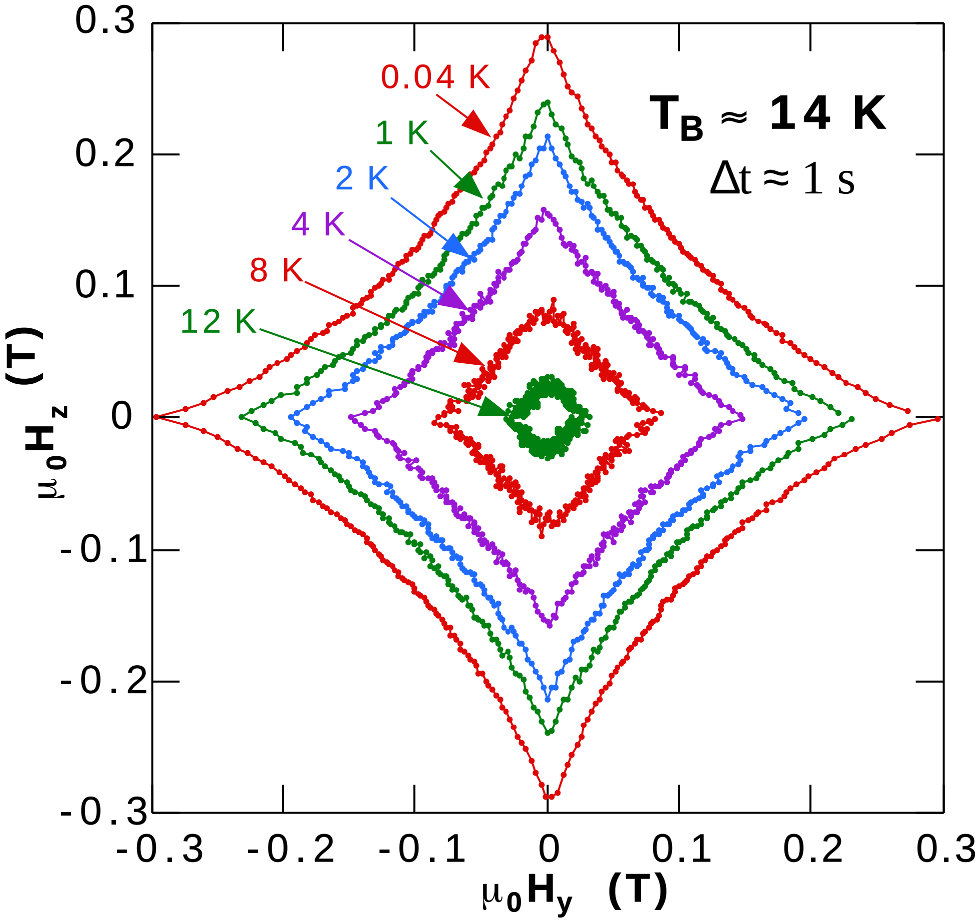
Magnetisation reversal mode

• nucleation, propagation, annihilation of domain walls

• uniform rotation, curling, nucleation in a small volume

• thermal activation, tunneling

Temperature dependence of the switching fields of a 3nm Co cluster



=> in agreement with the Néel Brown theory

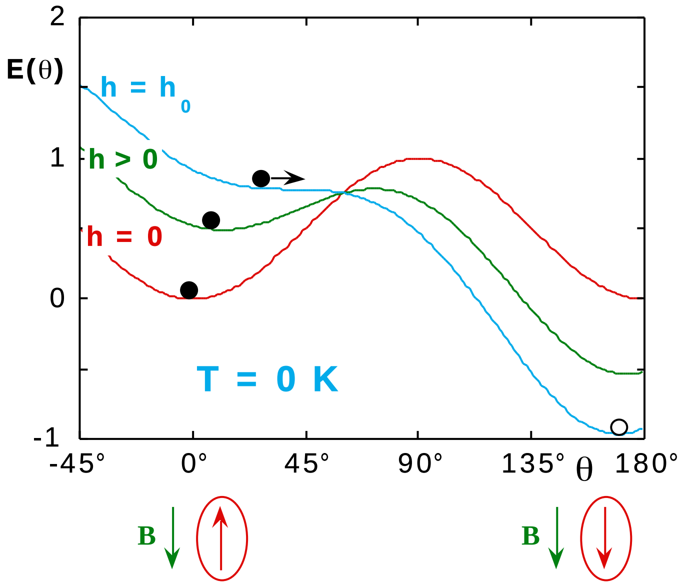
Temperature dependence of the magnetisation reversal

-- Escape from a metastable potential well --

Stoner - Wohlfarth model

$$\Delta E = E_0\left(1 - \frac{H_{sw}}{H_{sw}^0}\right)^{\alpha}$$

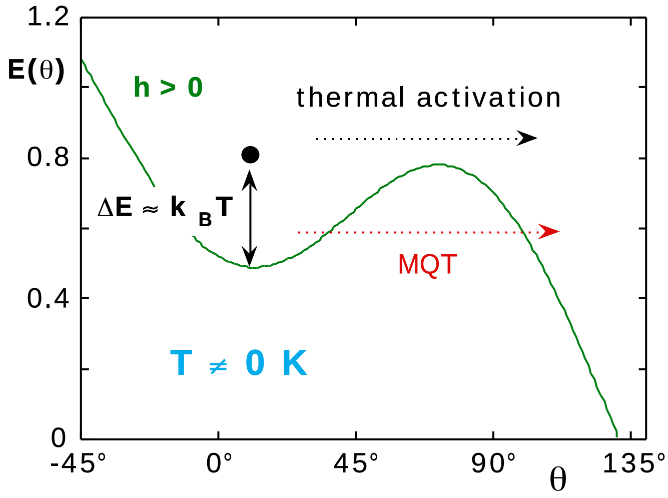
$$\alpha(angle) = 1.5 \leftrightarrow 2$$



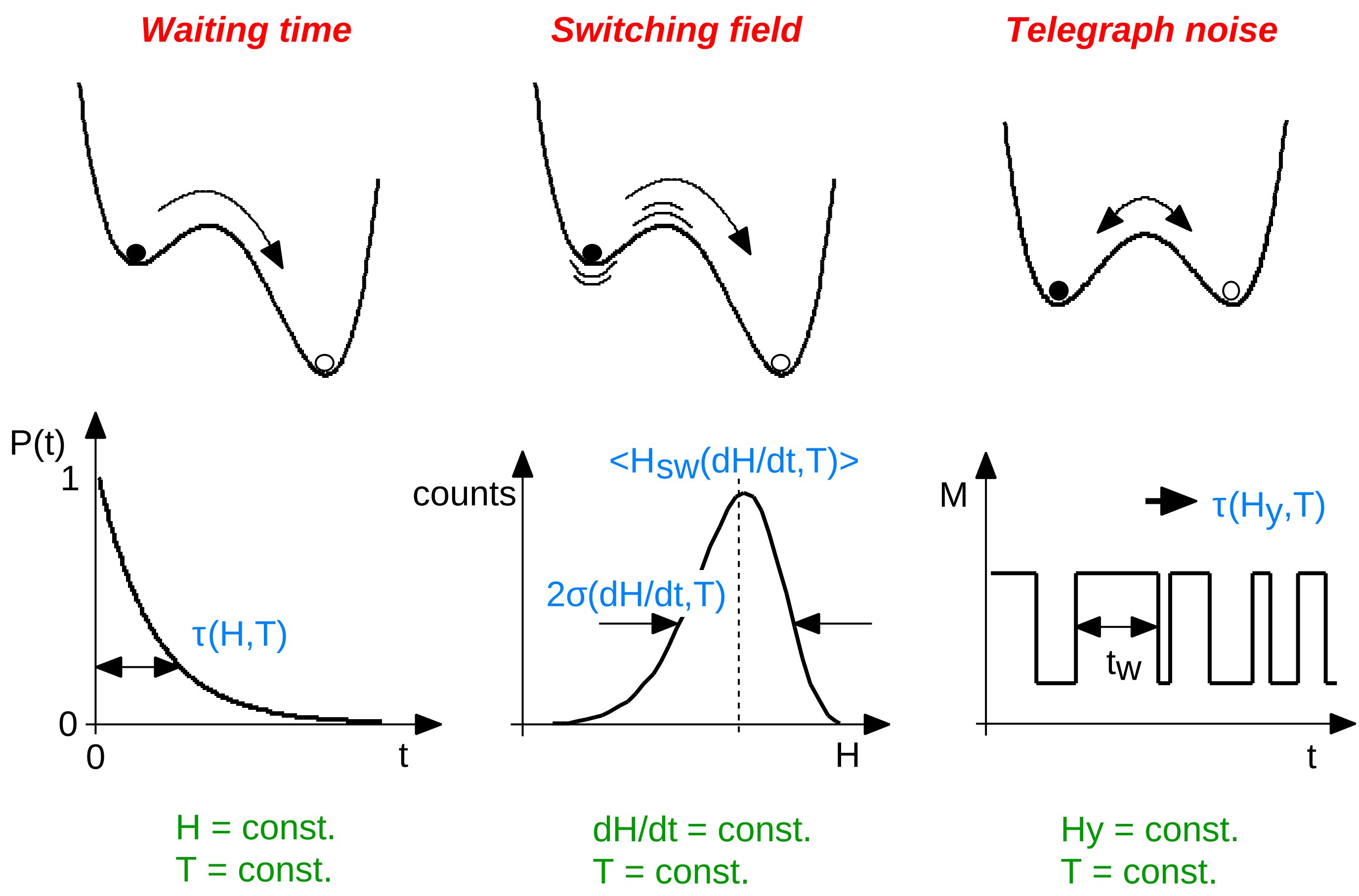
Néel Brown model

$$P(t) = e^{-t/\tau}$$

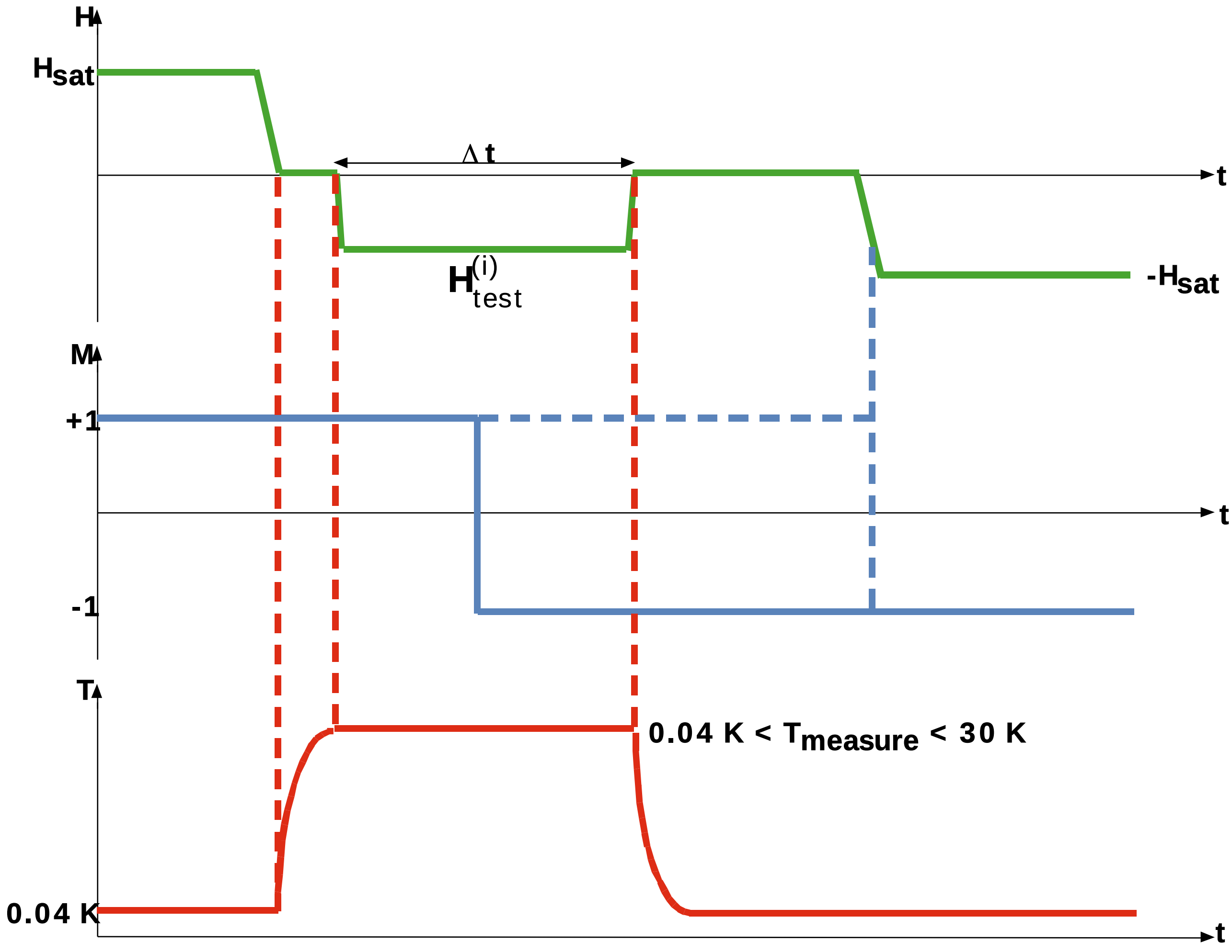
$$\tau = \tau_0 e^{\Delta E/kT}$$



Experimental methods for the study of the Néel–Brown model



Synopsis of the switching field measurement at a given temperature



Temperature dependence of the switching fields of a 3 nm Co cluster

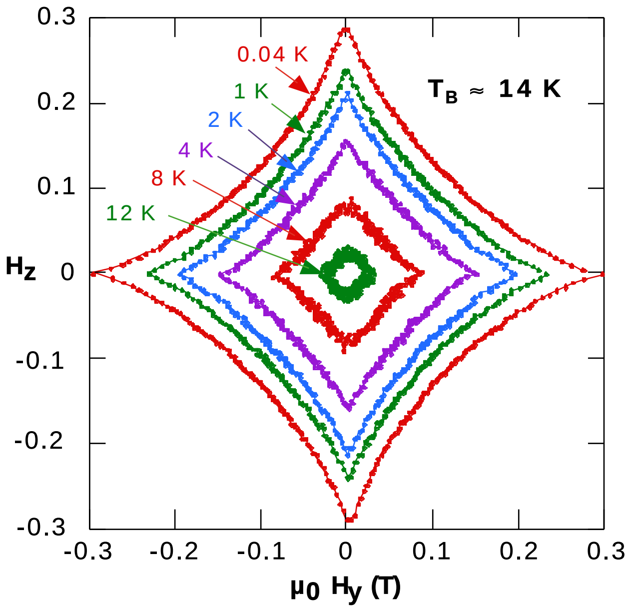
Néel Brown theory:

$$P(t) = e^{-t/\tau}$$

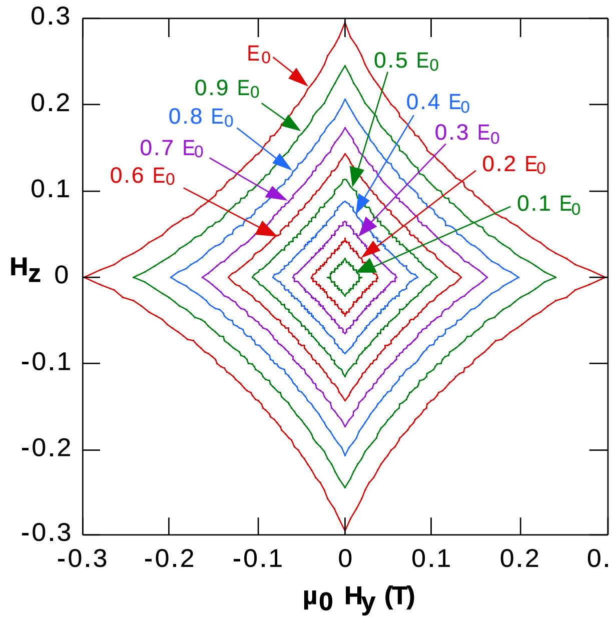
$$\tau = \tau_0 e^{\Delta E/kT}$$

$$\Delta E = E_0\left(1 - \frac{H_{sw}}{H_{sw}^0}\right)^{\alpha}$$

$$\alpha(angle) = 1.5 \leftrightarrow 2$$



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



$$\Delta E = \ln(\tau/\tau_0)\, k_B T$$

Temperature dependence of the switching fields of a 3 nm Co cluster

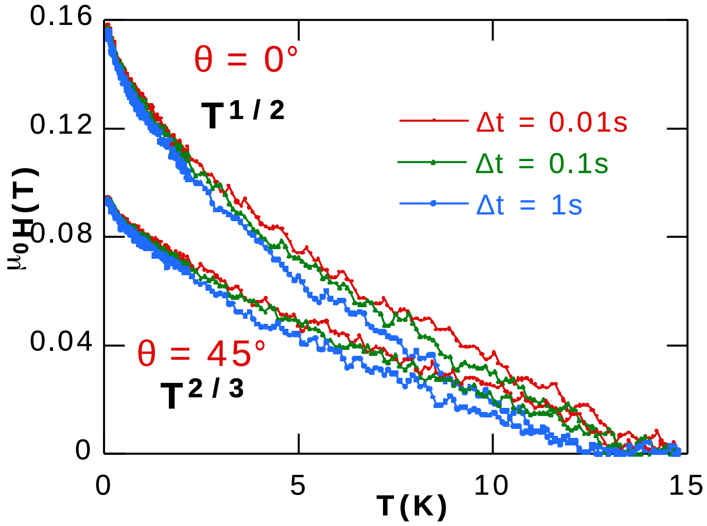
Néel Brown theory:

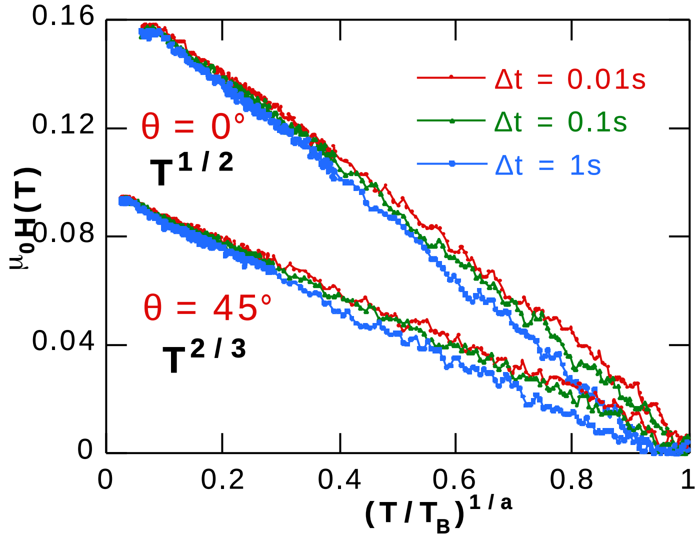
$$P(t) = e^{-t/\tau}$$

$$\tau = \tau_0 e^{E/kT}$$

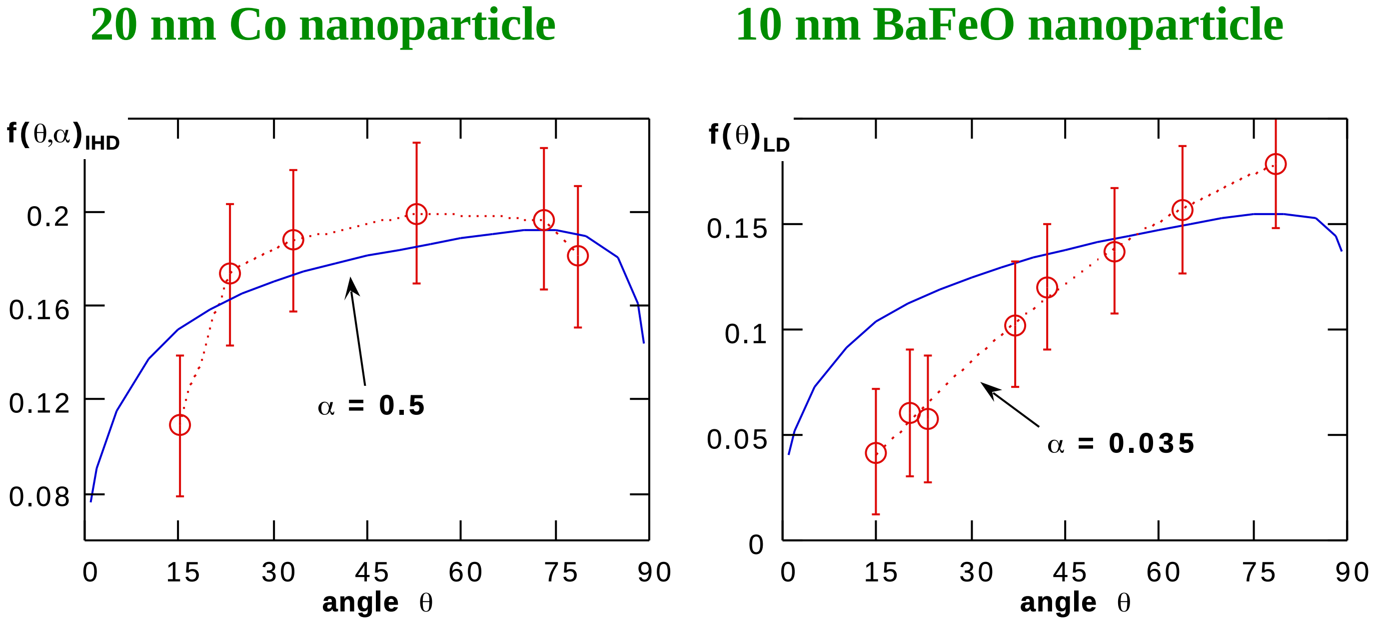
$$E = E_0\left(1 - \frac{H_{sw}}{H_{sw}^0}\right)^{\alpha}$$

$$\alpha(\text{angle}) = 1.5 \leftrightarrow 2$$





Application of the Néel-Brown-Coffey model to find the damping parameter α



W.T. Coffey, W. Wernsdorfer, et al, PRL, 80, 5655 (1998)

Magnetization reversal dynamics

$$\frac{d\vec{M}}{dt} = -\gamma \left(\vec{M} \times \vec{H}_{eff}\right) + \frac{\alpha}{M_S}\left(\vec{M} \times \frac{d\vec{M}}{dt}\right)$$

Landau-Lifshitz-Gilbert equation

St.-W. particle (macrospin) : $\|M\| = \vec{M}_s$

γ : gyromagnetic ratio

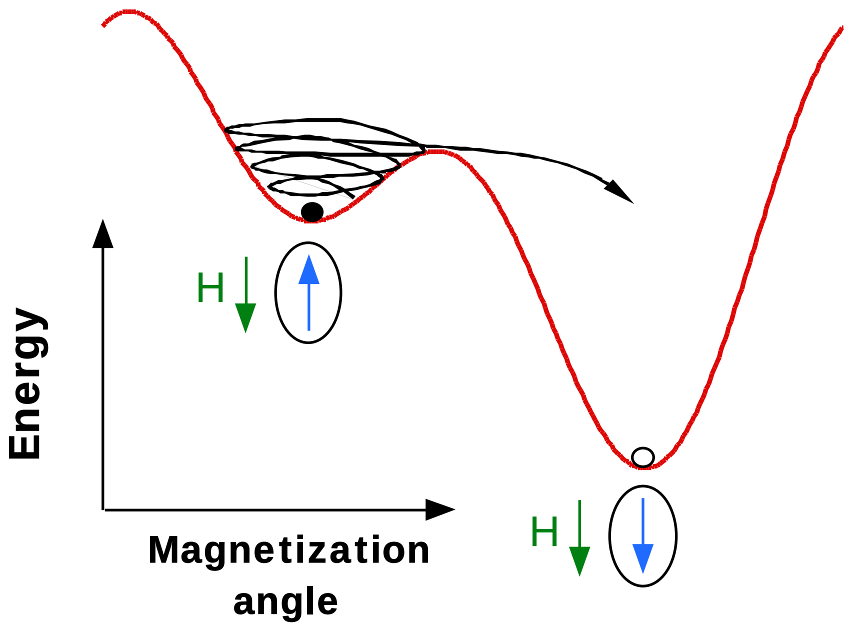
M_s : saturation magnetization

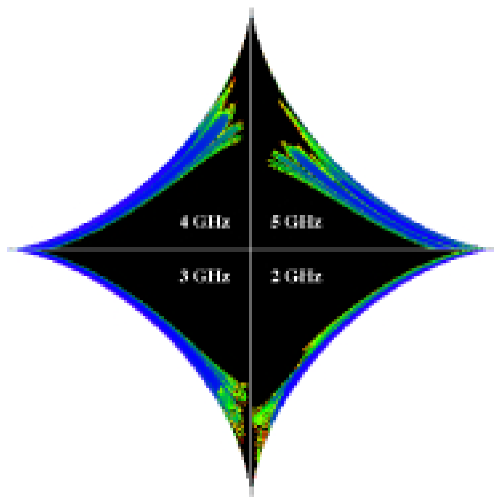
α : the damping parameter

H_{eff} : effective field (derivative of the free energy density with respect to M)

Numerical integration using Runge-Kutta

Switching of magnetization by non-linear resonance studied in single nanoparticles

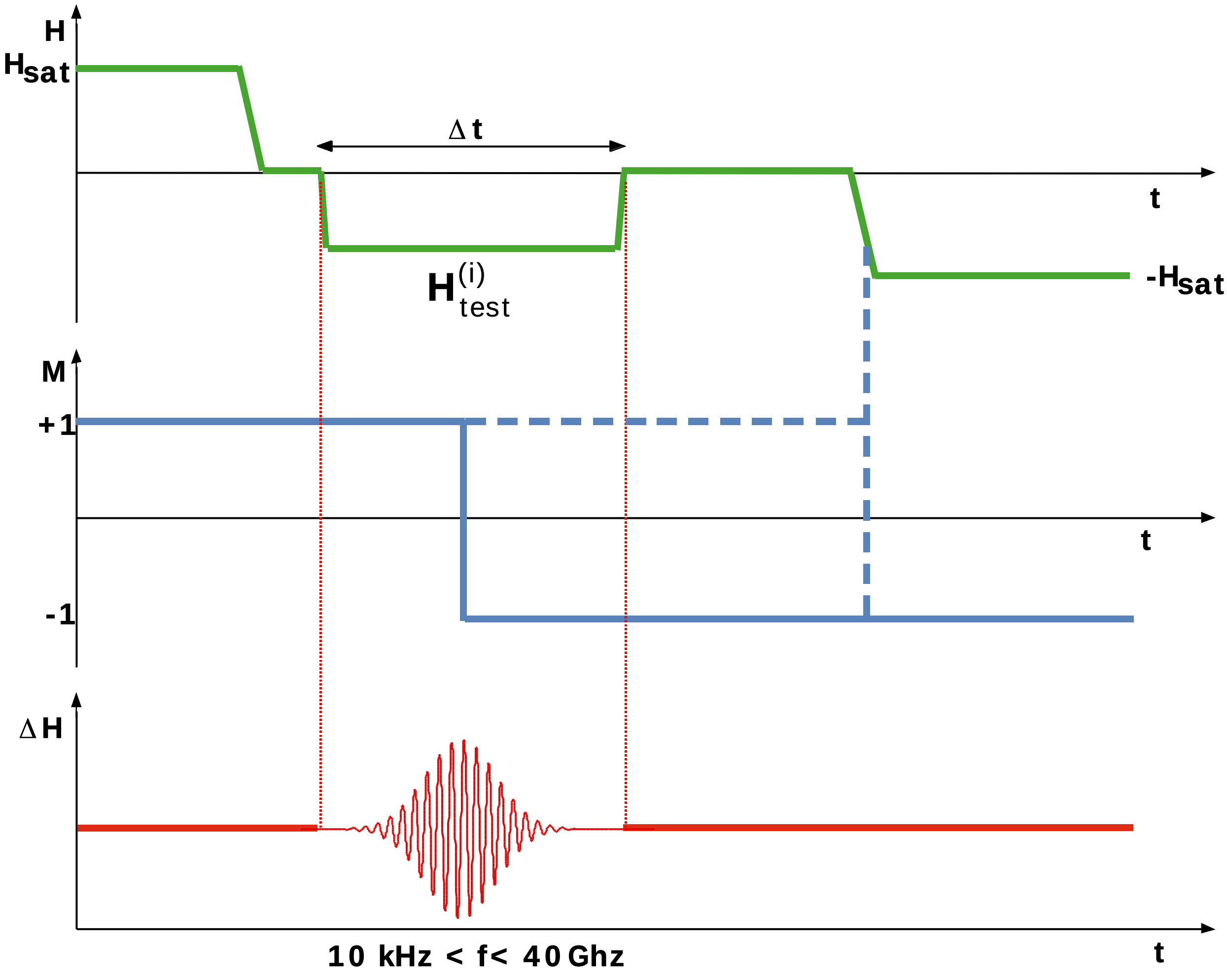




Dynamical astroid

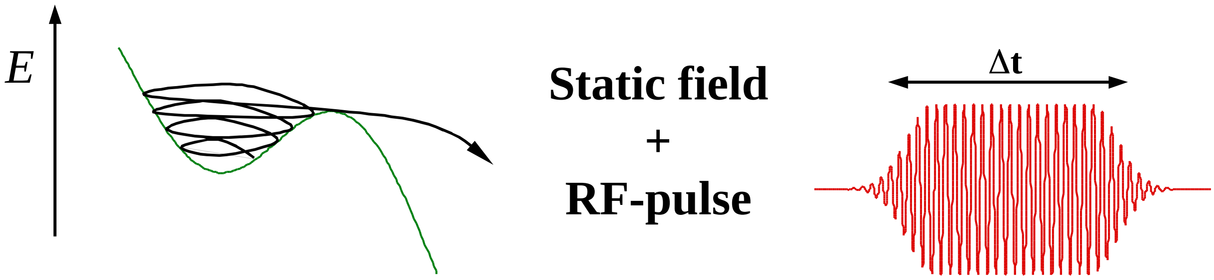
C. Thirion,
W. Wernsdorfer,
D. Mailly
Nature Mat. (1.Aug. 2003)

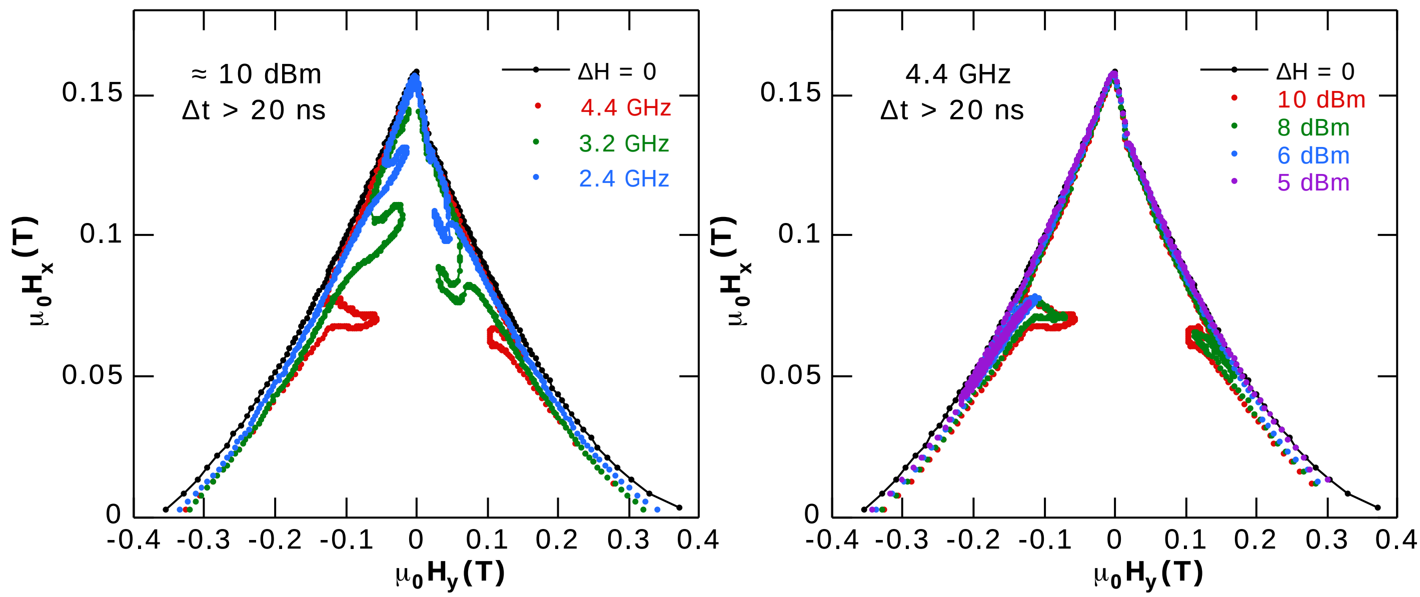
Synopsis of the switching field measurement with microwaves



Switching field measurement with microwaves

Magnetization reversal via precession





Magnetization reversal dynamics

$$\frac{d\vec{M}}{dt} = -\gamma\left(\vec{M} \times \vec{H}_{eff}\right) + \frac{\alpha}{M_S}\left(\vec{M} \times \frac{d\vec{M}}{dt}\right)$$

Landau-Lifshitz-Gilbert equation

St.-W. particle (macrospin) : $\|\vec{M}\| = M_s$

γ : gyromagnetic ratio

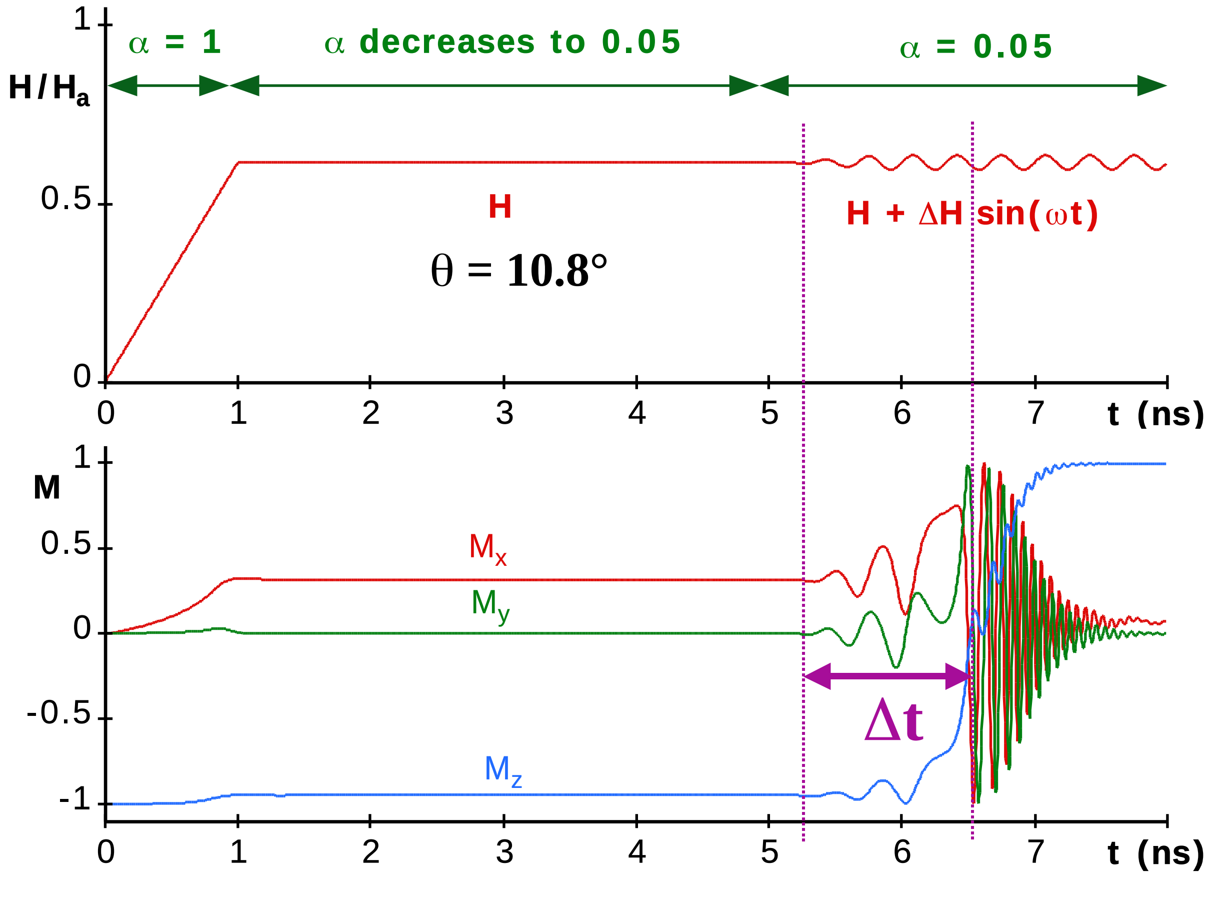
M_s : saturation magnetization

α : the damping parameter

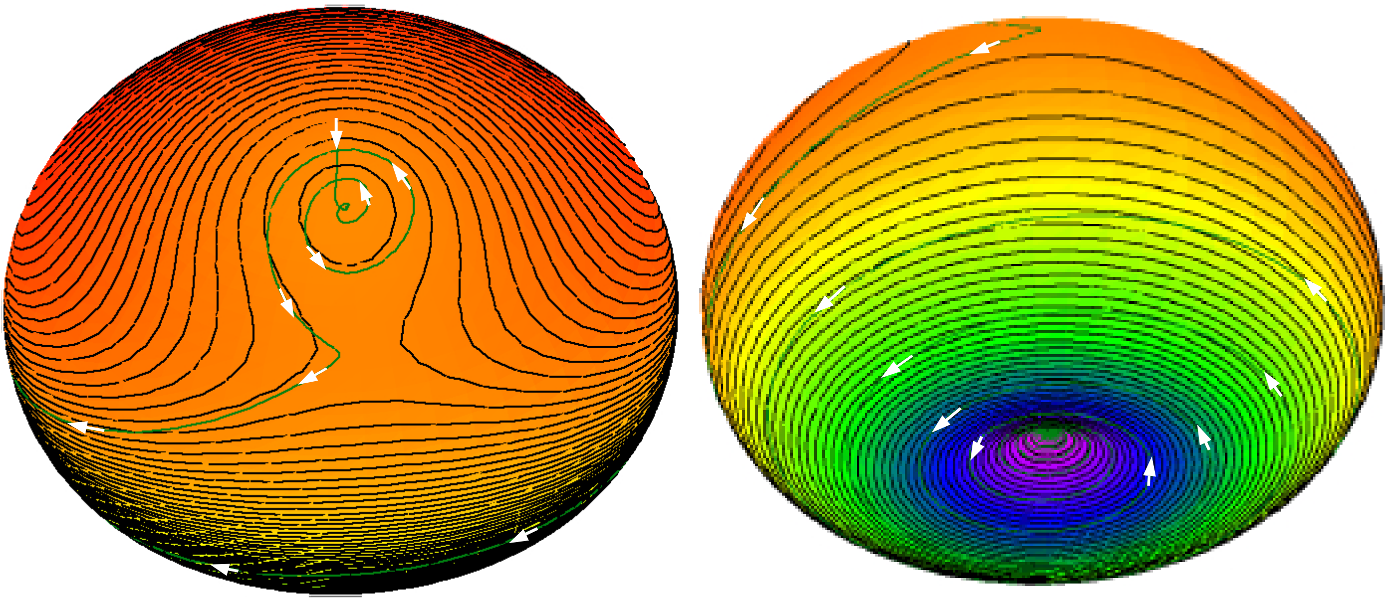
H_{eff} : effective field (derivative of the free energy density with respect to M)

Numerical integration using Runge-Kutta

Simulation of magnetization reversal via precession

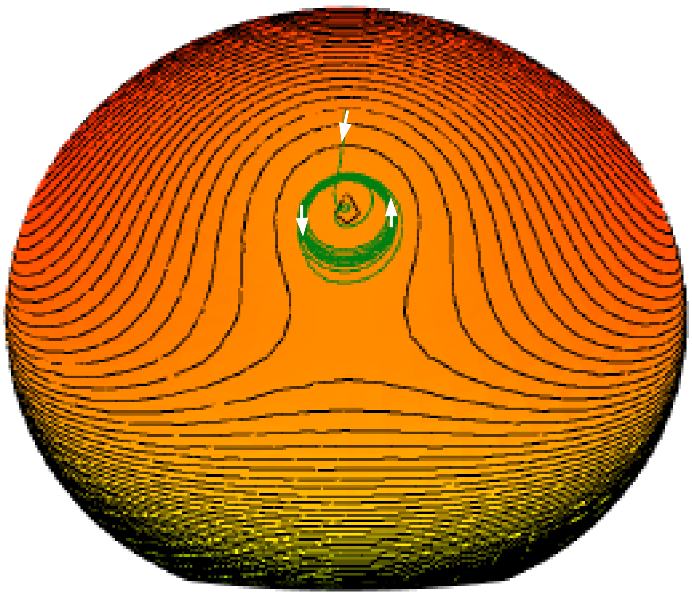


Magnetization reversal via precession

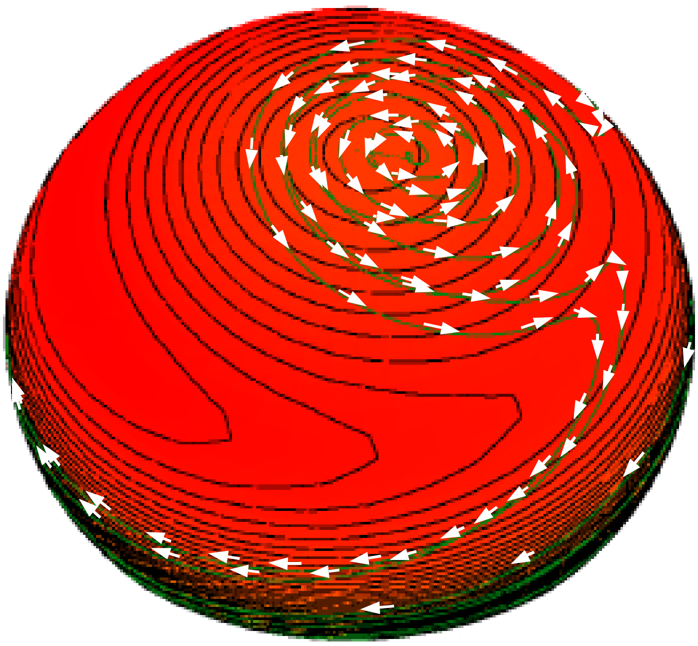


H = 0.615H_a, ΔH = 0.02H_a, θ = 10.8° α = 0.05, f = 3 GHz

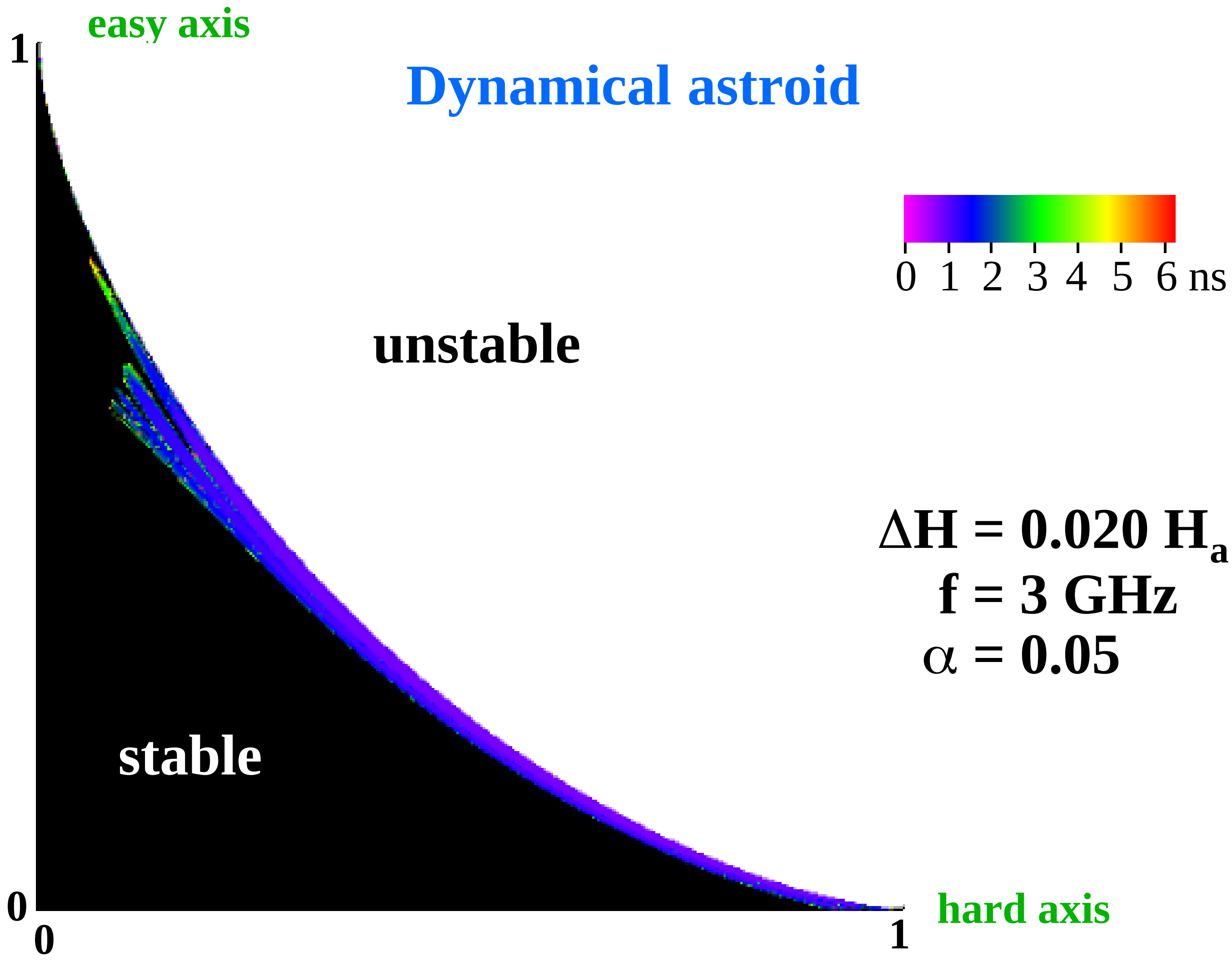
Magnetization reversal via precession



$H = 0.625H_a$, $\Delta H = 0.02H_a$,
$\theta = 11.4°$ $\alpha = 0.05$, $f = 3$ GHz

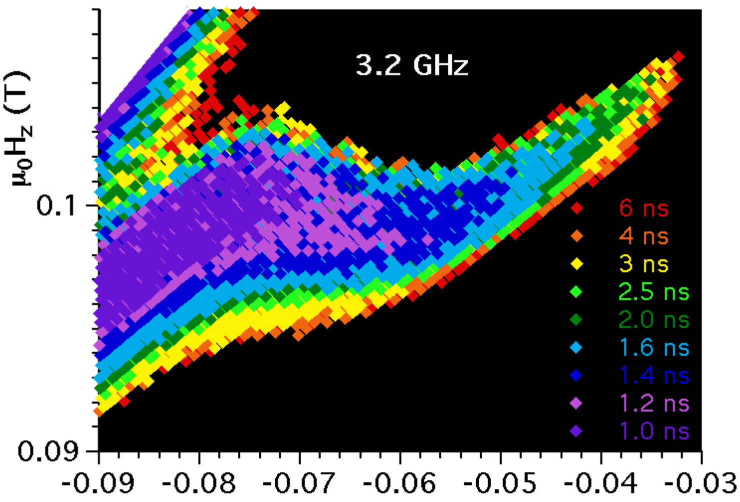


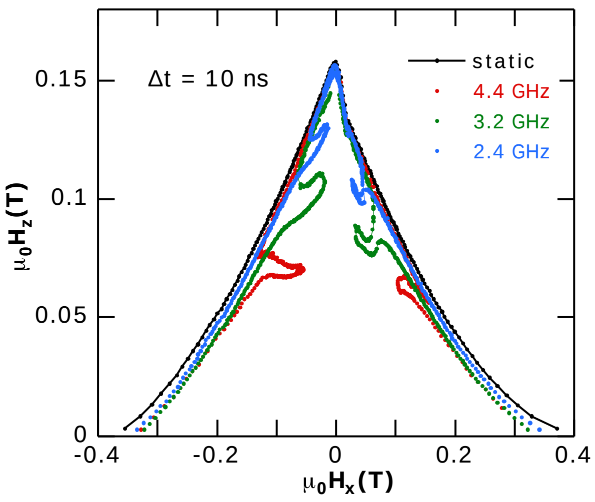
$H = 0.64H_a$, $\Delta H = 0.02H_a$,
$\theta = 1.55°$ $\alpha = 0.01$, $f = 3$ GHz

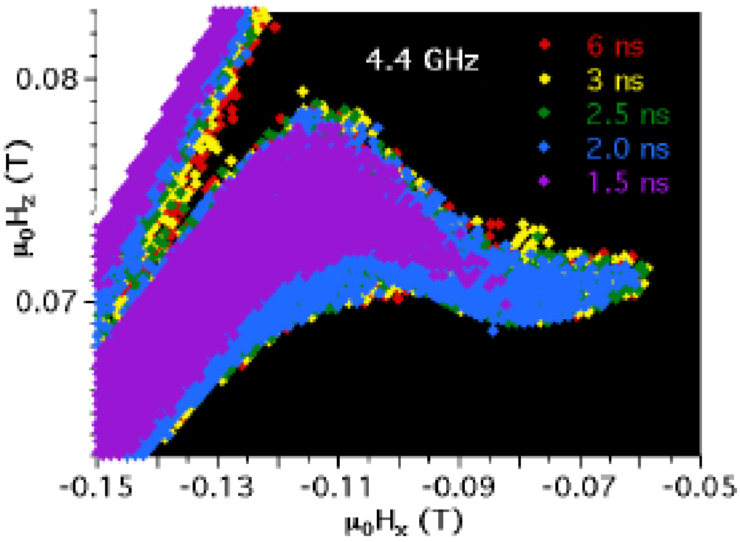


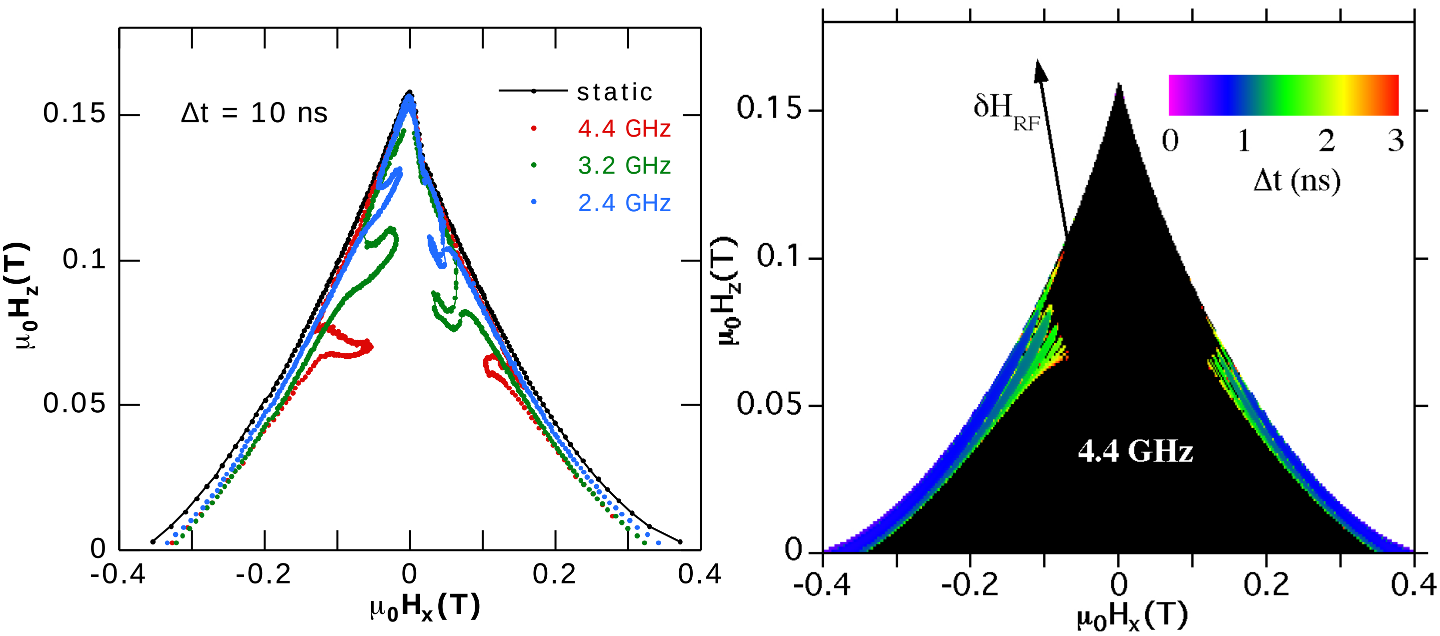
easy axis
1
Dynamical astroid
0 1 2 3 4 5 6 ns
unstable
ΔH = 0.020 H_a
f = 3 GHz
α = 0.05
stable
0
0
1
hard axis

Pulse length dependance



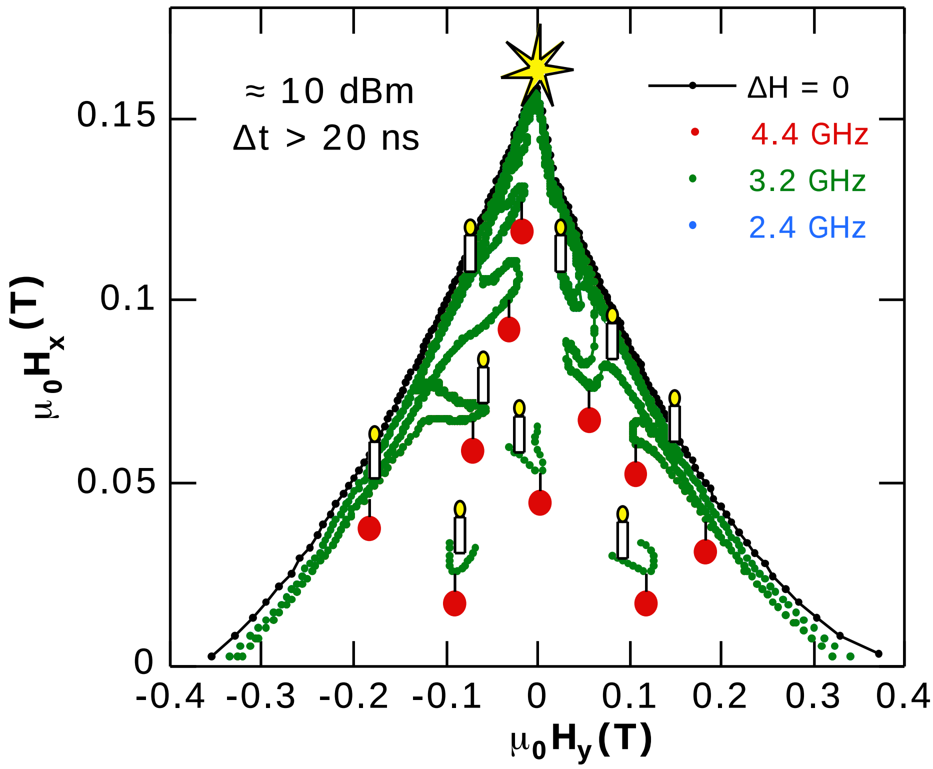






Δt = 10 ns
static
4.4 GHz
3.2 GHz
2.4 GHz
0.15
0.1
0.05
0
-0.4
-0.2
0
0.2
0.4
μ0Hz(T)
μ0Hx(T)
δHRF
0
1
2
3
Δt (ns)
4.4 GHz

Switching field measurement with microwaves
Magnetization reversal via precession



Conclusion

Measurements of magnetization reversal of nanoparticles containing about a thousand atoms

- The magnetic anisotropy is governed by surface anisotropy
- Switching field as a function of temperature are in agreement with the Néel Brown model
- Measurements probing nanosecond timescales (pulse fields, microwaves)

Coming soon (or later)

- Studying smaller particles (molecules)

Outline

Part I: *classical magnetism*

1. Magnetization reversal by uniform rotation
   (Stoner-Wohlfarth model)
   - theory
   - experiment (3 nm Co clusters)
2. Influence of the temperature on the magnetization reversal
   (Néel-Brown model)
3. Magnetization reversal dynamics
   (Landau-Lifshitz-Gilbert)
   - magnetization reversal via precession
   - dynamical astroid