Magnetisation dynamics at different timescales: dissipation and thermal processes part II.

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Different timescales:

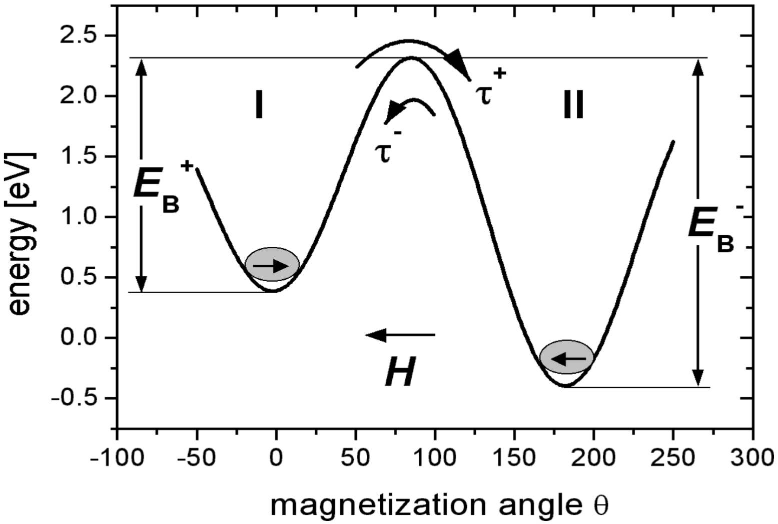
10^{-14}s fs	Electron-spin relaxation processes.	All-optical laser-pulsed experiments	Langevin dynamics on atomistic level
10^{-11}s ps	Magnetisation precession.	Fast-Kerr measurements	Langevin dynamics on micromagnetic level
10^{-9}s ns			
10^{-6}s μs			dynamics acceleration techniques
10^{-3}s ms	Hysteresis measurements.	Conventional magnetometers (VSM, SQUID)	
10^{-0}s s			
10^{3}s hs	Magnetic viscosity experiments.		kinetic Monte Carlo with energy barriers calculations
10^{6}s month			
10^{9}s years	Long-time thermal stability for magnetic recording.		

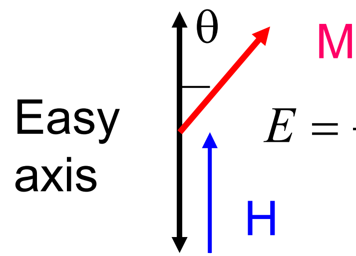
Outline of the talk

- Long-time (>μs) magnetisation dynamics
  - The concept of energy barriers and switching for an individual nanoparticle
  - Calculation of energy barriers for more complicated systems
  - Energy barriers for systems of interacting nanoparticles
  - Kinetic Monte Carlo for thermal decay evaluation in a completely interacting system.
- Ultra-short timescale magnetisation dynamics (fs-ps)
  - The ultra-fast pump-probe experiment
  - Modelling
  - Introduction of the correlated noise approach.

Long timescale (up to years)

The Stoner-Wolfarth particle





$$E = -KV\cos^2\theta - MH\cos\theta$$

Energy barrier:

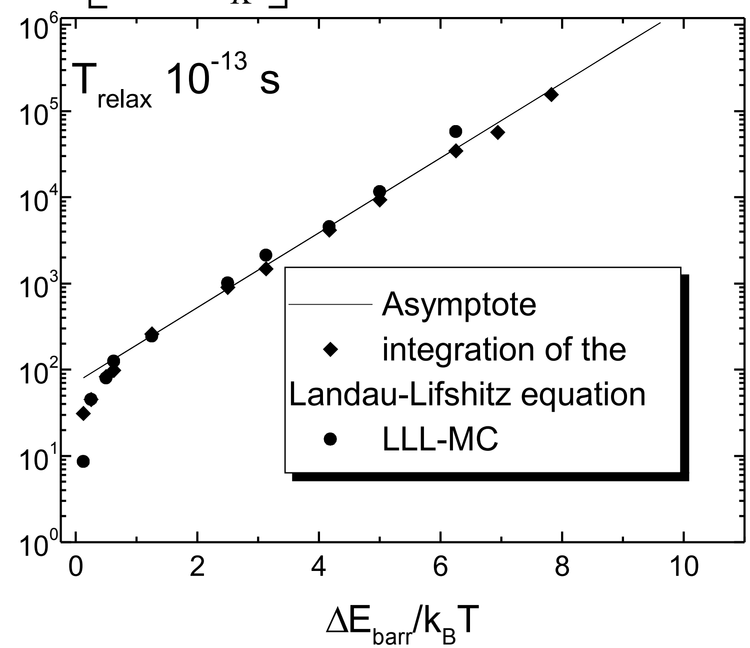
$$\Delta E = KV\left[1 - \frac{H}{H_K}\right]^2, \quad H_K = 2K/M_s$$

The Arrhenius-Neél law

$$\tau_{\pm}^{-1} = f_0 \exp(-\Delta E_{\pm}/kT) = f_{1,2}$$

$$dm_1/dt = -f_1 m_1 + f_2 m_2$$ - Master equation

$$f_2 << f_1 \quad m(t) \propto \exp(-t/\tau)$$



Brown's asymptote:

For an independent small particle with applied field parallel to anisotropy
The reversal probability is obtained from the solution of the
Fokker-Planck equation

$$\frac{1}{\tau} = \frac{2K\alpha\gamma}{M_s(1+\alpha^2)\sqrt{\pi}}\sqrt{\frac{KV}{k_B T}}(1-h^2)(1+h)\exp\left[-KV(1+h)^2/k_B T\right]$$

•In general, $f_0=1/\tau_0=F(H, \alpha, K, \theta, T)$ but in most of cases the approximation

$f_0=10^9\text{-}10^{11}\ s^{-1}$ is sufficient

The general problem does not have solution.

Relaxation in complex systems

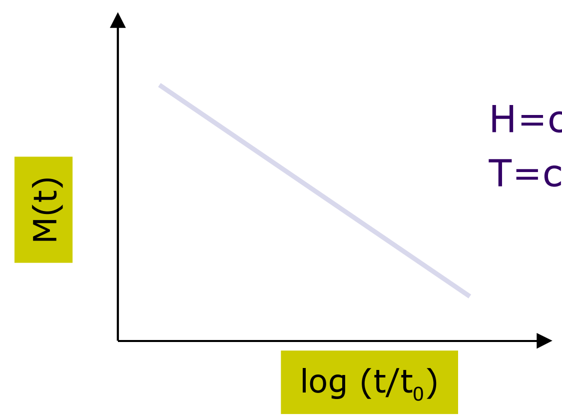
$$\tau_{\pm}^{-1} = f_0 \exp(-\Delta E_{\pm} / kT) \qquad m(t) \propto \int \exp[- -t/\tau(\Delta E)]\rho(\Delta E)\,d\Delta E$$

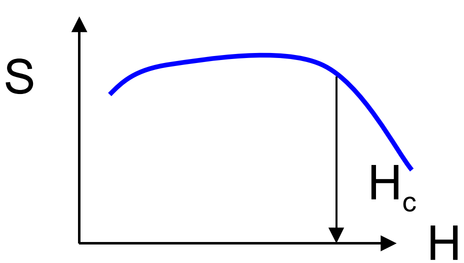
If in some interval $[\Delta E, \Delta E + \delta\Delta E]$ $\quad \rho(\Delta E) \approx const$

$m(t) \propto \Delta E \propto M_0 - S\log(t)$ -widely observed behavior

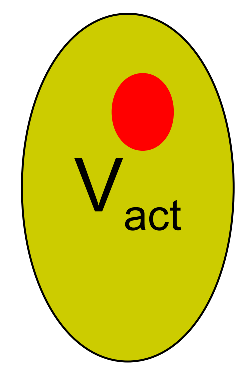
•Based on the assumption of Homogeneous energy barrier distribution which is constant in time

$S = dM / d(\log t)$ -magnetic viscosity

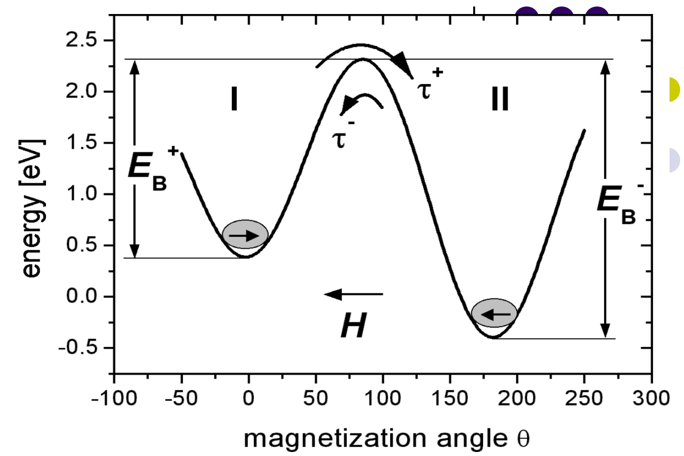




$V_{act} = k_B T / (\mu_0 H M_s)$ Activation volume



Superparamagnetism



- The relaxation time of a grain is given by the Arrhenius-Neel law

$$\tau_{\pm}^{-1} = f_0 \exp(-\Delta E_{\pm} / kT)$$

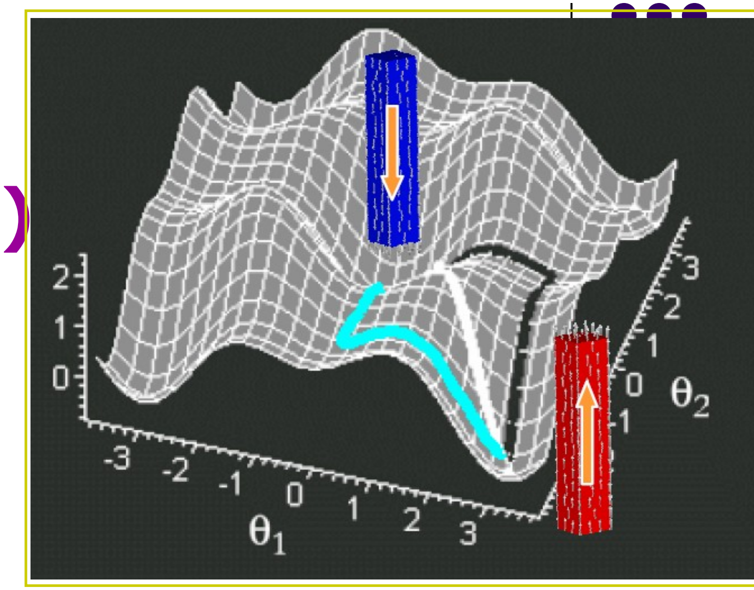
- where $f_0 = 10^9 s^{-1}$. and ΔE is the energy barrier
- This leads to a critical energy barrier for superparamagnetic (SPM) behaviour $\Delta E_c = KV_c = k_B T ln(t_m f_0)$

- where t_m is the 'measurement time'
  - Nanoparticles with $\Delta E < \Delta E_c$ exhibit thermal equilibrium (SPM) behaviour - no hysteresis

$KV > 25k_BT$ – for stability at room temperature, $KV > 60k_BT$ – for magnetic recording

Slow processes:

$k_B T \ll \Delta E$ (Energy barrier)



$$t_i = t_0 \exp(\Delta E / k_B T)$$

- Energy barrier calculation is essential part for determination of long-time thermal stability and slow thermal relaxation

- This is important from the point of view of magnetic recording applications.

- Evalulation of energy barriers should be done in a multidimensional space and is a difficult problem

- in an interacting system energy barriers are dynamical and should be constantly recalculated.

Slow processes:

$k_B T << \Delta E$ (Energy barrier)

Energy barriers should satisfy conditions:

grad E =0

Only one (lowest) eigenvalue of the Hessian matrix $\dfrac{\partial^2 E}{\partial m_i \partial m_j}$

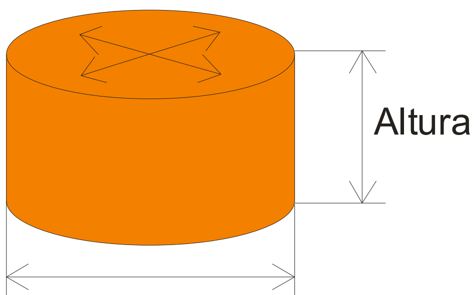
$\varepsilon_1 < 0$

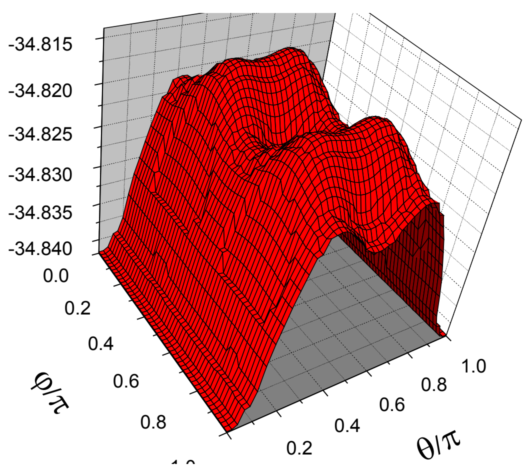
Constrained (Lagrangian multiplier) method:

(for simple cases only)

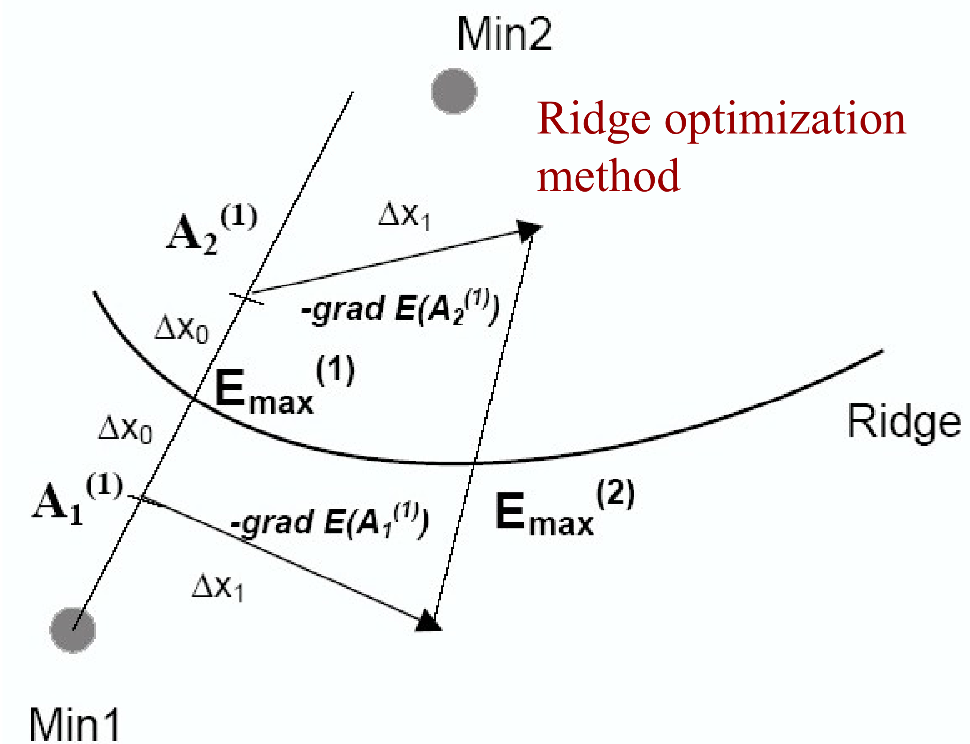
$$\mathcal{F} = \mathcal{H} - \mathcal{N}\boldsymbol{\lambda}(\boldsymbol{\nu} - \boldsymbol{\nu}_0), \boldsymbol{\nu} \equiv \frac{\sum_i \boldsymbol{m}_i}{\left|\sum_i \boldsymbol{m}_i\right|}$$

Fe dot





Ridge optimisation method

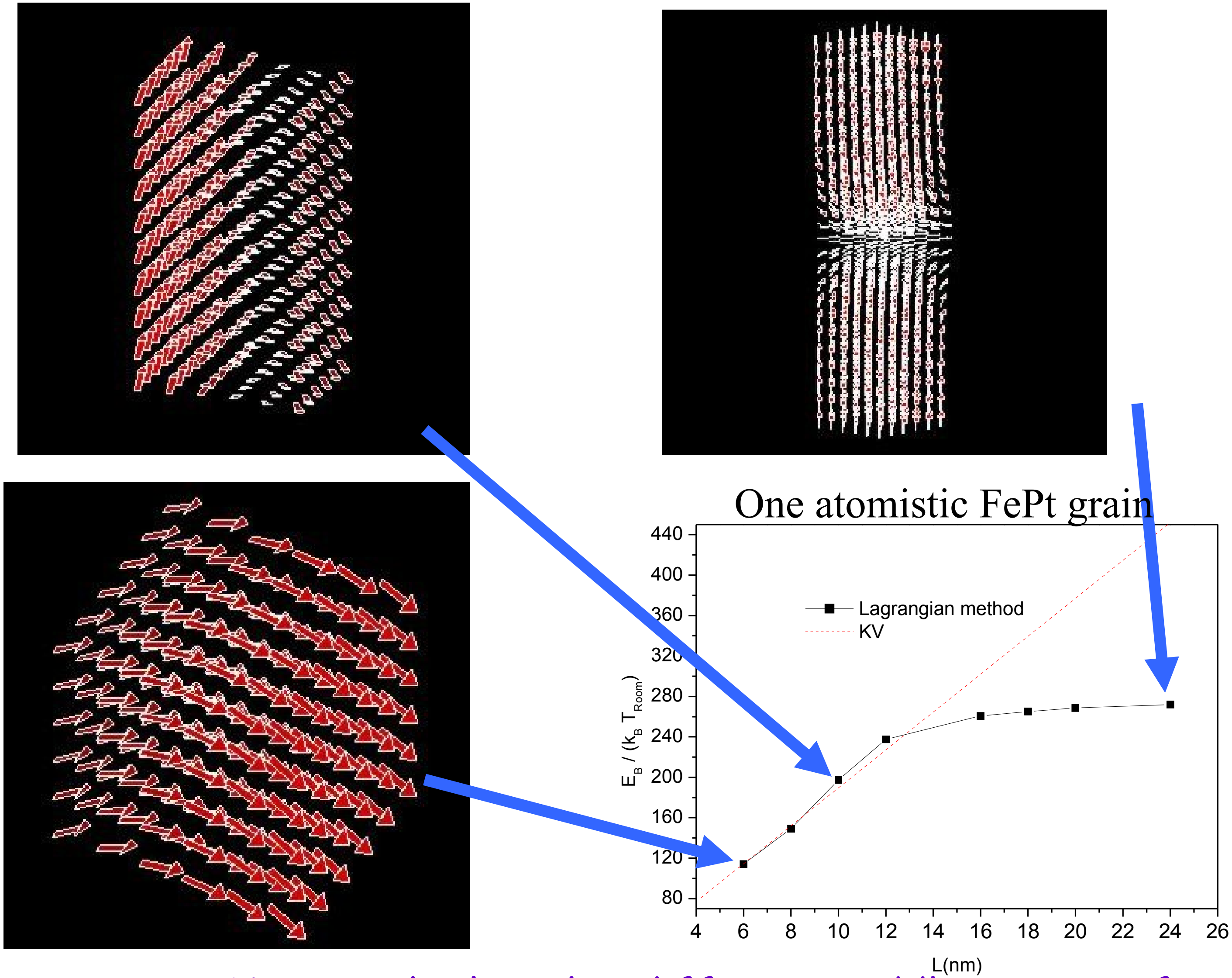


The obtained point is checked:

- To have a unique negative eigenvalue
- To separate the basins of attractions of the two minima from which one is initial

Similar method –elastic band

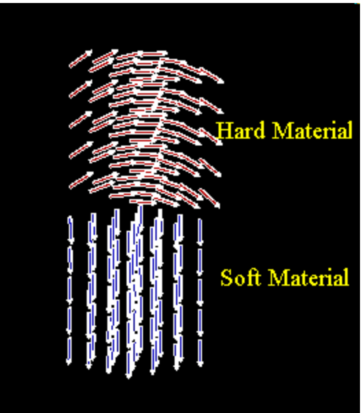
Energy barriers in a single FePt grain



Varying the length-> different saddle point configurations corresponding to different reversal mechanism

Energy barriers as a function of Js: soft/hard grain

saddle point configuration



For Js>0.1J energy barriers are collective and larger than individual energy barrier of the hard material

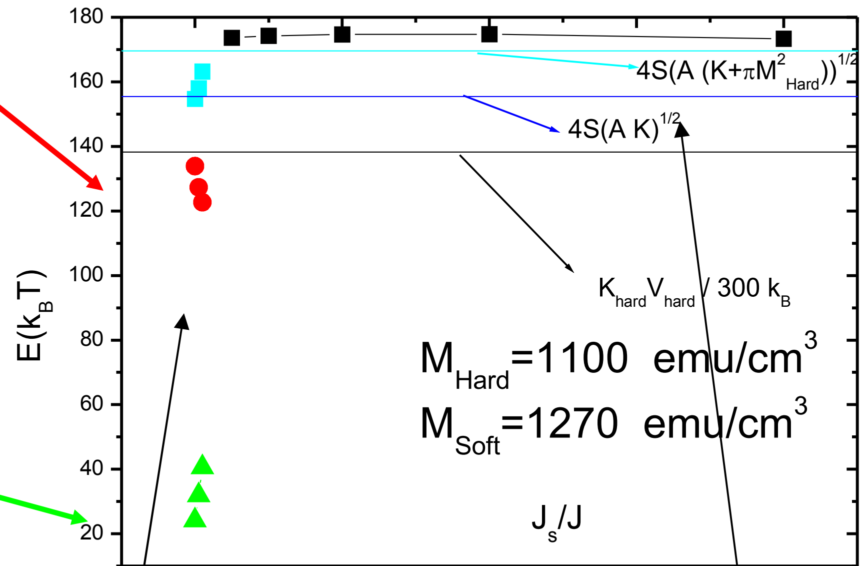
Collective energy barrier

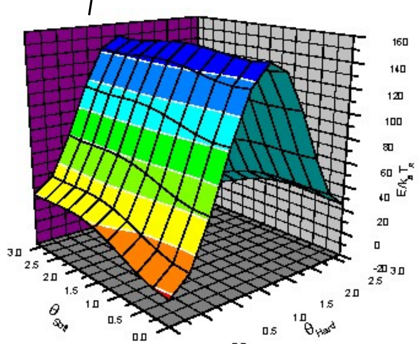
is defined by the energy of the domain wall in the hard layer

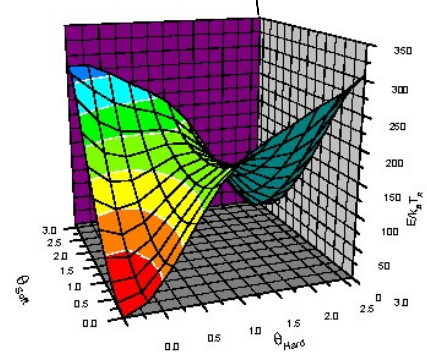
hard layer follows

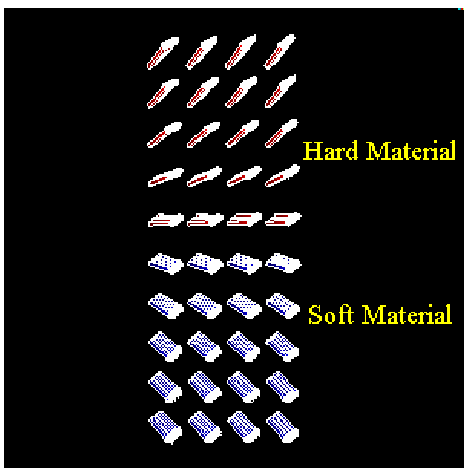
Individual energy barriers

soft layer goes first

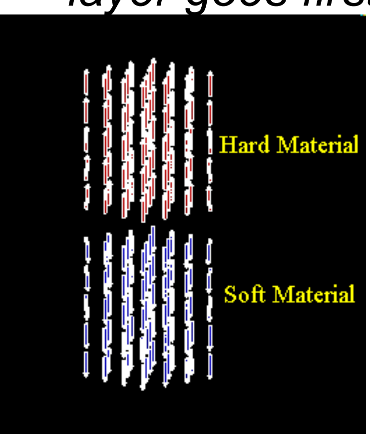








saddle point configuration



Energy barriers for systems of nanoparticles

The Pfeiffer approximation

H.Pfeiffer Phys Status Solidi A 118, 295 (1990):

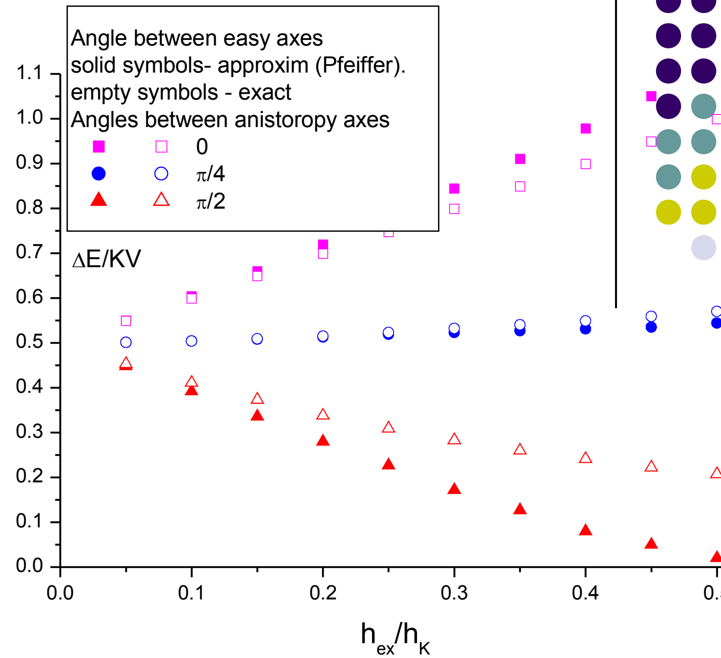
$$\Delta E_{Pf} = KV[1 - h_{\mathrm{int}} / g(\phi)]^{k(\phi)}$$

$$g(\phi) = [\cos^{2/3}(\phi) + \sin^{2/3}(\phi)\ ]^{-3/2}$$

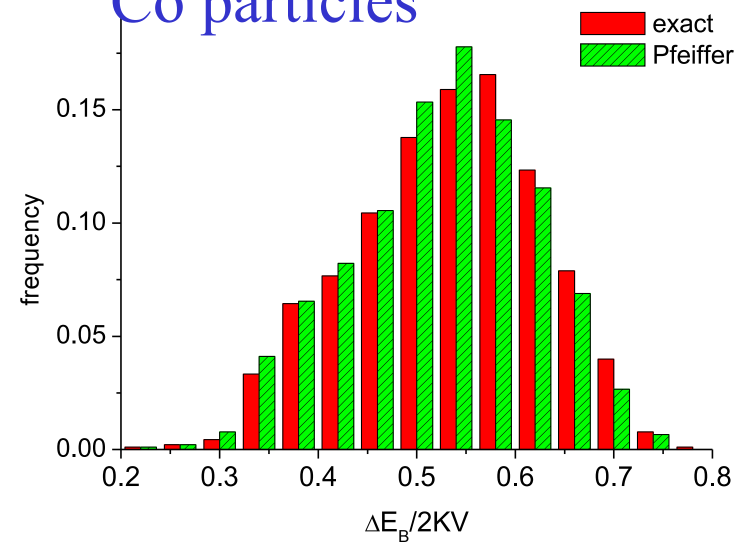
$$k(\phi) = 0.86 + 1.14 g(\phi)$$

$$\Delta E_B = \begin{cases} \Delta E_{Pf} & (\vec{M}, \vec{h}_{\mathrm{int}}) < 0 \\ \Delta E_{Pf} + E^2_{\min} - E^1_{\min} & (\vec{M}, \vec{h}_{\mathrm{int}}) > 0 \end{cases}$$

Φ is the angle between anisotropy and interaction field

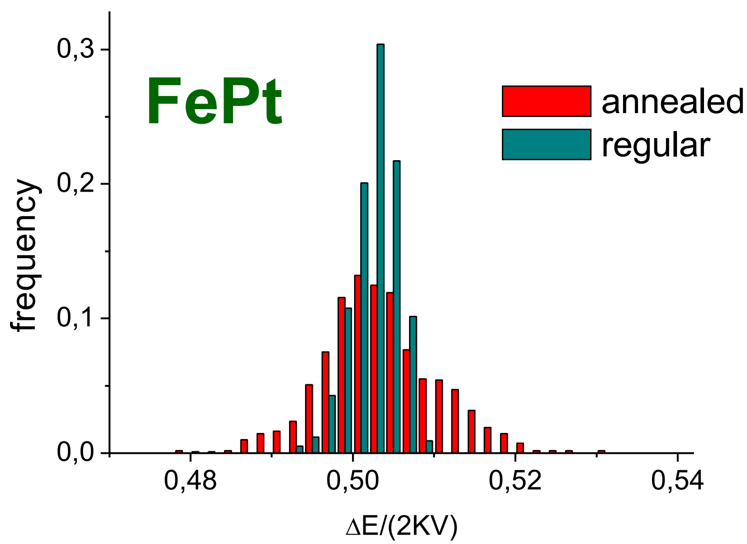


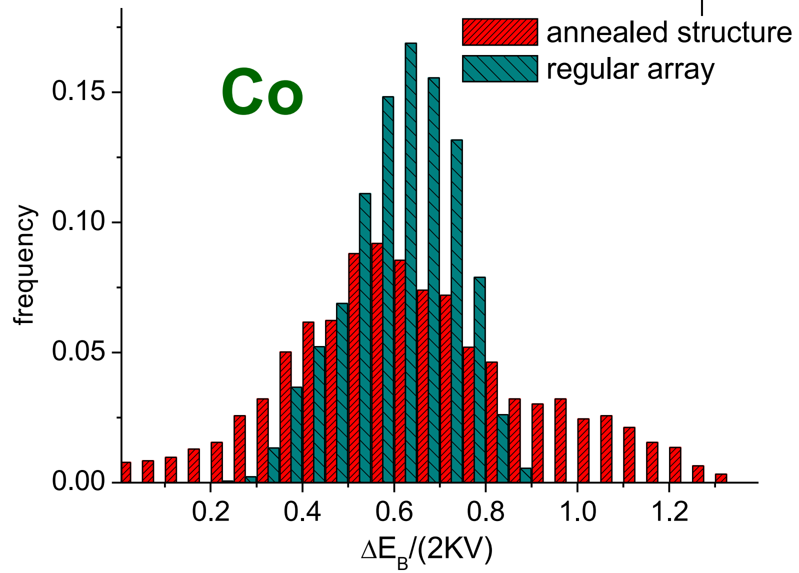
Co particles



The Pfeiffer approximation is slightly displaced to smaller values

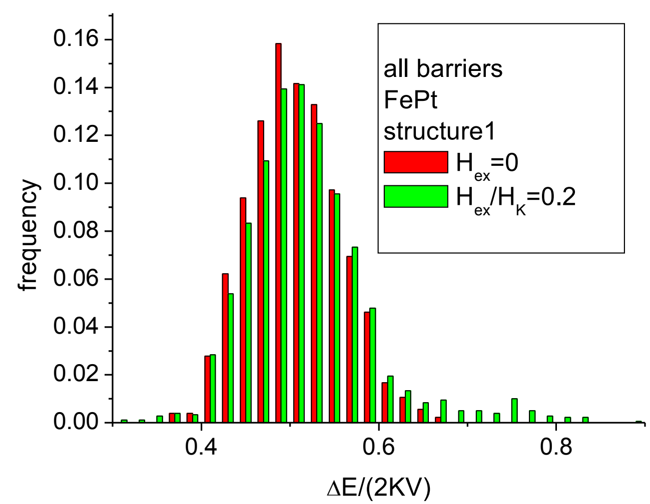
Multidimensional energy barrier distribution for Co and FePt particles
(only magnetostatic interactions)



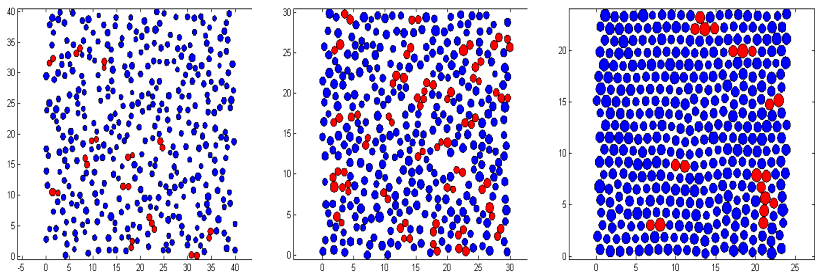


Multidimensional energy barrier distribution

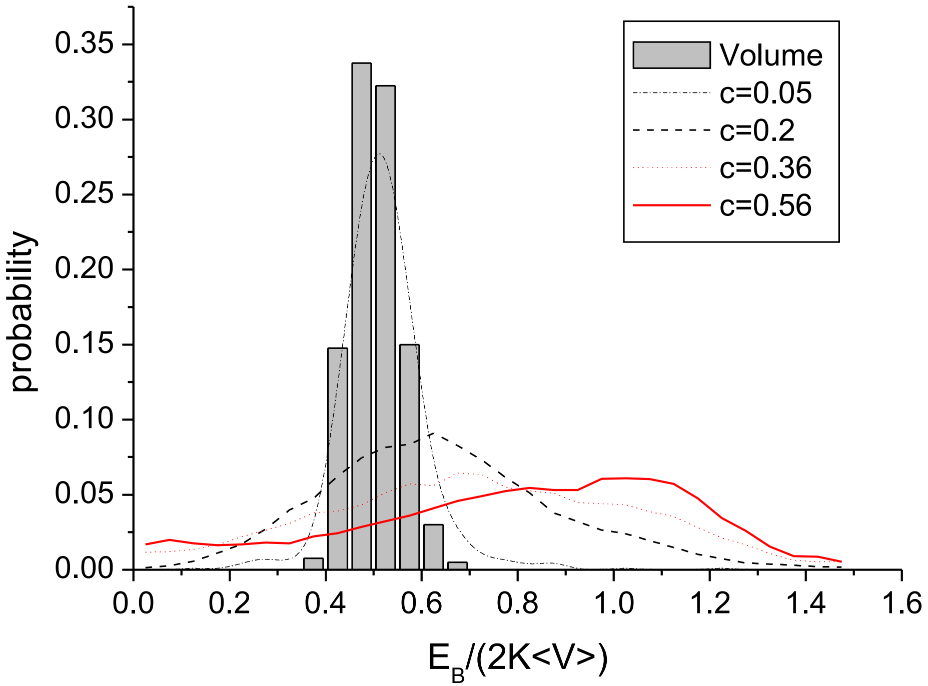
for annealed FePt particles array in the presence of exchange



Multidimensional energy barrier distributions evaluated at the remanence



c=0.2 c=0.36 c=0.56



Magnetostatic interactions:
- **Broaden distributions**
- **Displace the center to larger values**

This is consistent with experimental observations that with the increase of the strength of interactions:

- **Magnetisation decay starts earlier**
- **The blocking temperature increases**

Kinetic Monte Carlo

- **Evaluate all energy barriers in multidimensional space**
- **Evaluate all transition rates, according to the Arrhenius law**

$$f_i = f_0 \exp(-\Delta E / k_B T) \qquad f = \sum f_i$$

- **Choose a particle (cluster) with the probability proportional to its transition rate and invert it**
- **Approximate the waiting time from the exponential distribution**
- **Recalculate all the energy barriers**

$$D(t)dt = f \exp(-ft)dt$$

in practice is possible for only small interaction

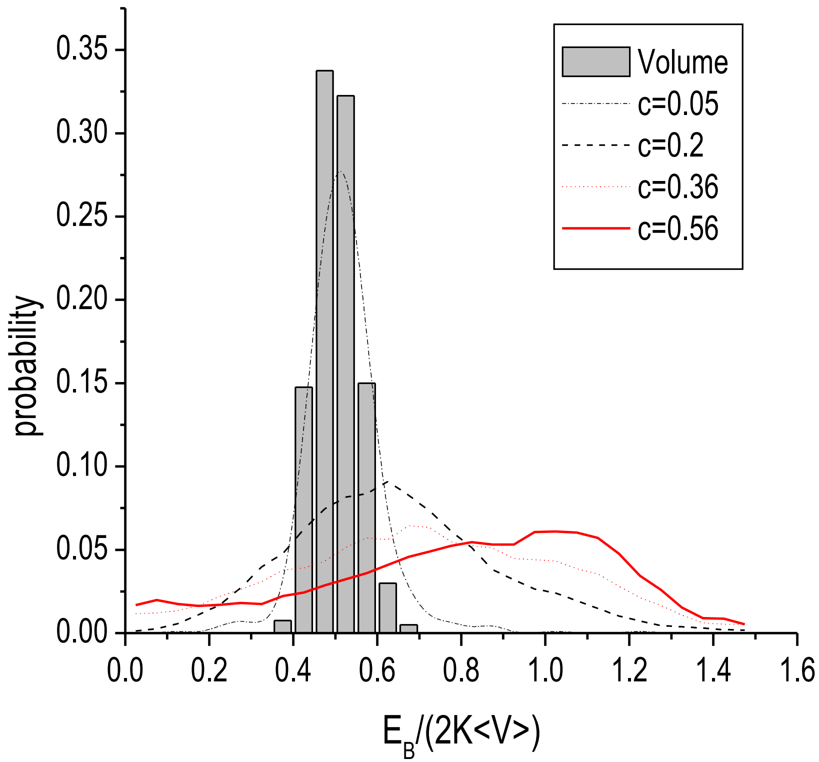
- **Only plausible, if a good initial guess for all the clusters is known**
- **Energy barrier distribution is a dynamical property and requires a large computational effort.**

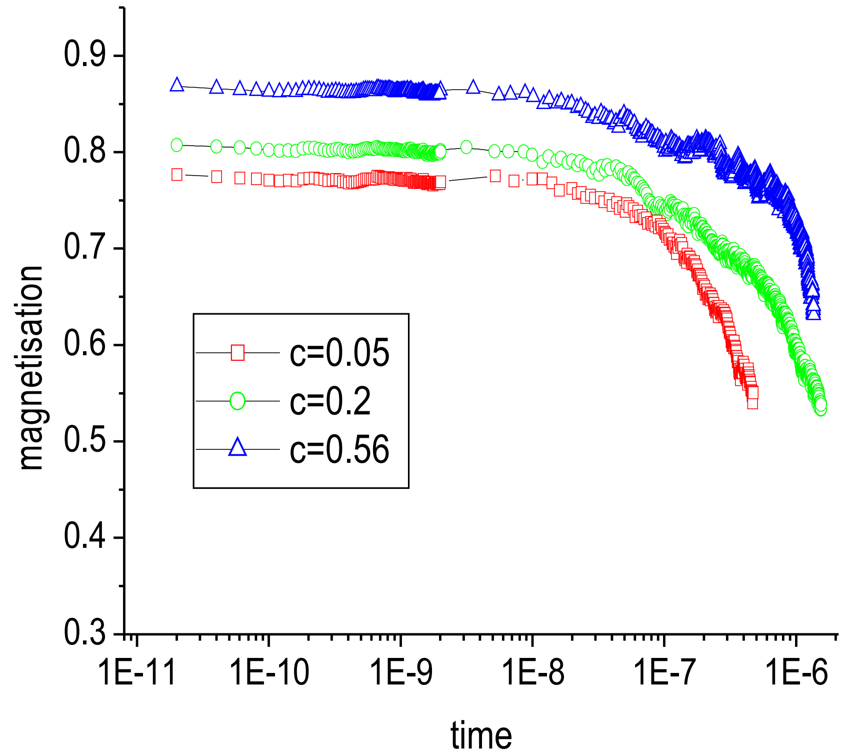
For initial guess, we use the Metropolis MC with simulated annealing

Thermal decay for an emsemble of 2D Co particles:

(starting from the remanent state, in-plane field 2D easy random easy axes distribution)

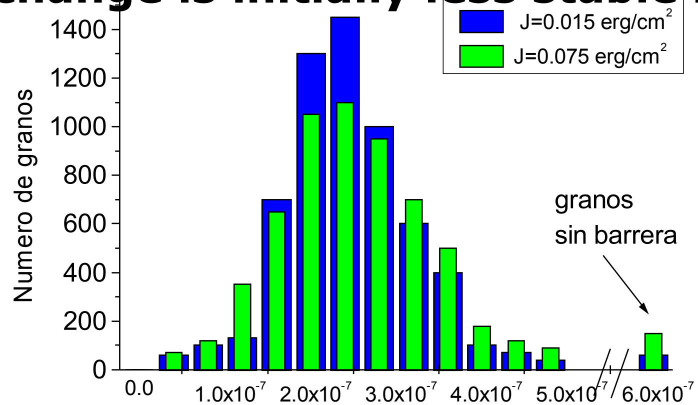
Initial energy barrier distributions

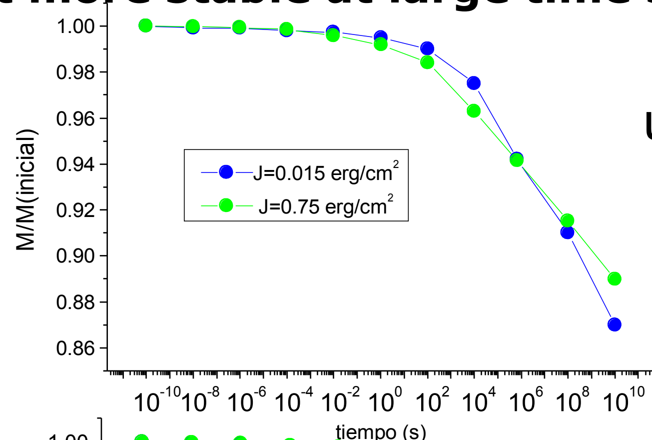




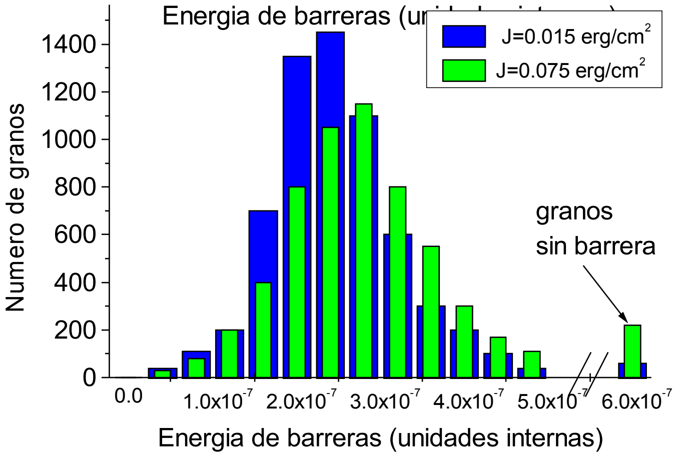
Energy barrier and magnetic relaxation calculated for conventional Co longitudinal recording medium

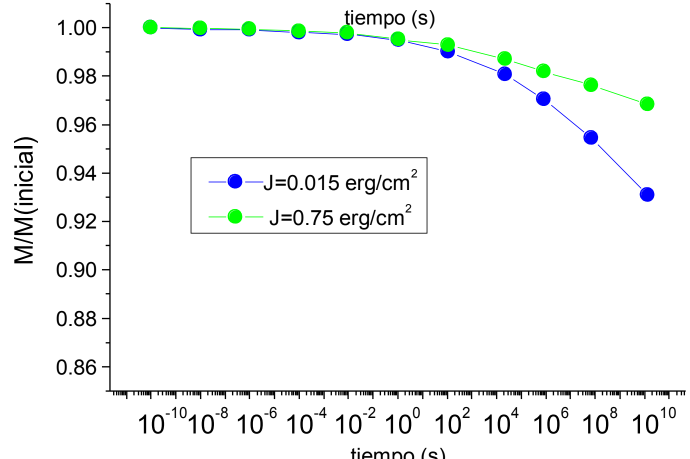
Important conclusions: Untextured medium with small amount of exchange is initially less stable but more stable at large time scale





untextured



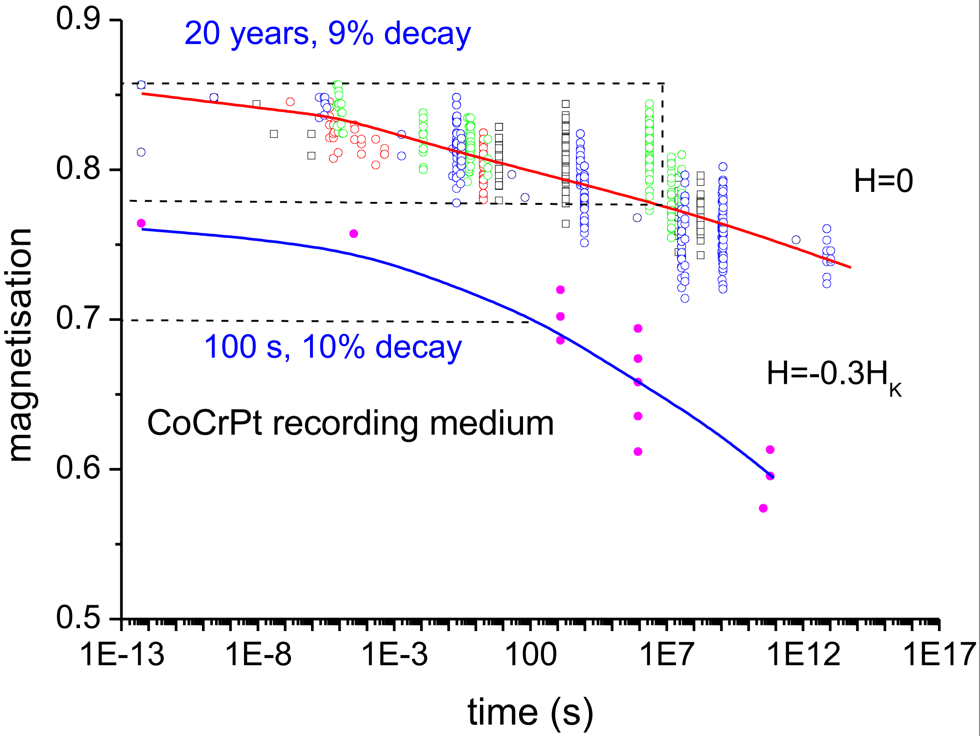


textured

Textured medium is more stable in the presence of small amount of exchange

Perpendicular CoCrPt recording medium

(100 grains, full micromagnetic contributions)



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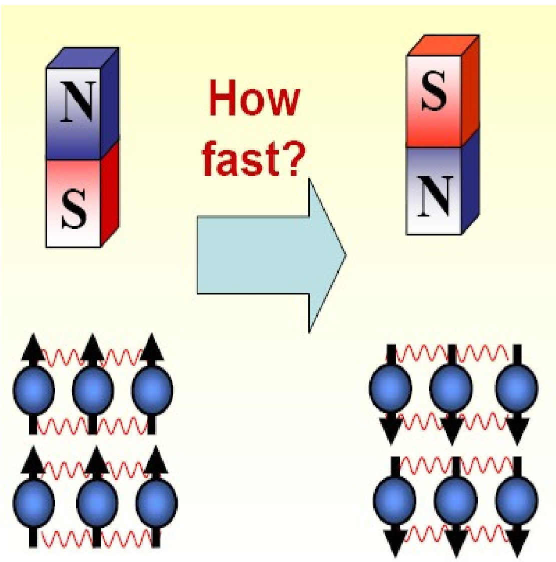
Textured, random easy axes, <V>= 6.5nm, K=2.4 10 erg/cm , M = 442 emu/cm

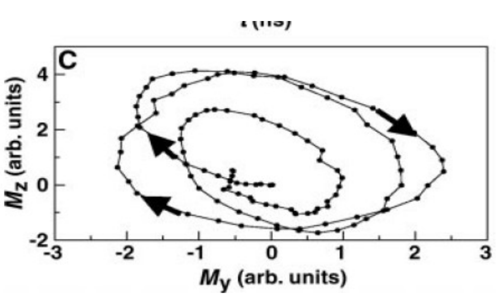
Ultrafast timescale: femto-pico seconds

"Spin-flip" as a fundamental problem



Imaging Precessional Motion of the Magnetization Vector

Y. Acremann,[1] C. H. Back,[1*] M. Buess,[1] O. Portmann,[1] A. Vaterlaus,[1] D. Pescia,[1] H. Melchior[2]



Applied physics

Speed limit ahead

C. H. Back and D. Pescia

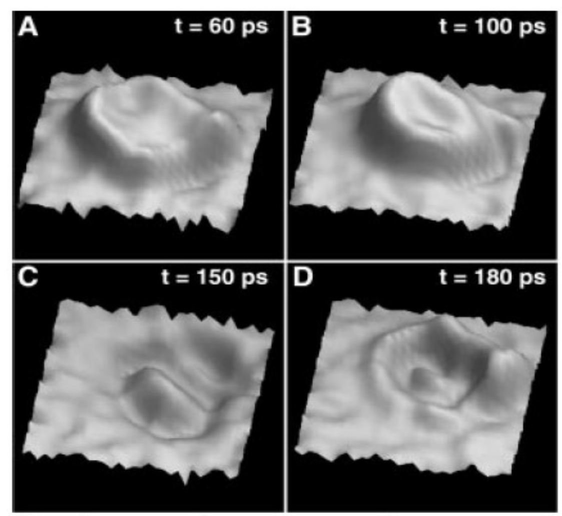
Are there any limits to what science and technology can achieve? When it comes to recording data in magnetic media, the answer is yes: there is a natural limit to the speed at which data can be encoded.

Via field pulses:

The ultimate speed of magnetic switching in granular recording media

I. Tudosa[1], C. Stamm[1], A. B. Kashuba[2], F. King[3], H. C. Siegmann[1], J. Stöhr[1], G. Ju[4], B. Lu[4] & D. Weller[4]

2000



Motivation

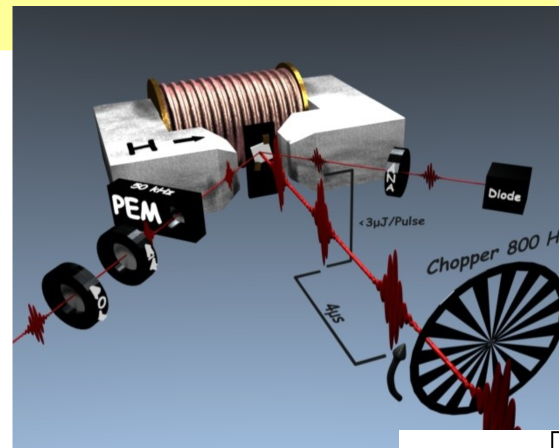
- Model the 3 regions of ultrafast spin dynamics experiments:

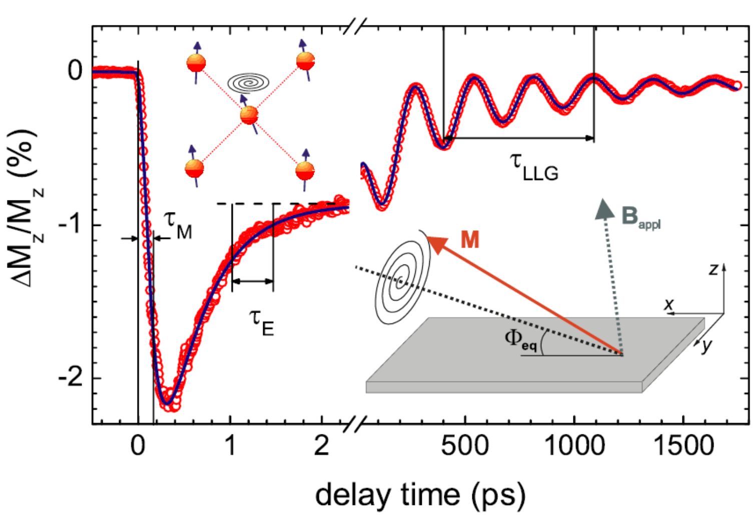
M. Van Kampen et *al, Phys. Rev. Lett*. **95**, 267207 (2005)

Fast demagnetisation $\tau \approx$ femtoseconds (10^{-15}s)

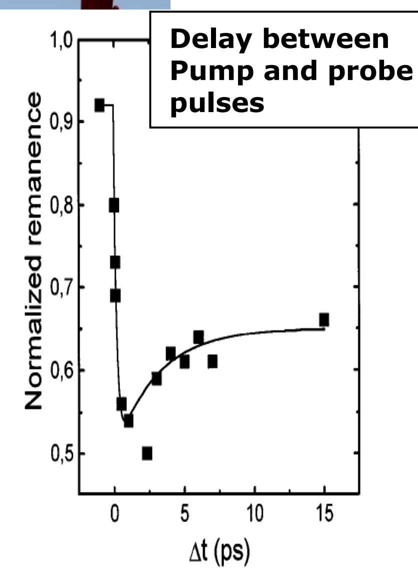
II. Magnetisation recovery $\tau \approx$ picoseconds

III. Damped precession $\tau \approx$ nanoseconds

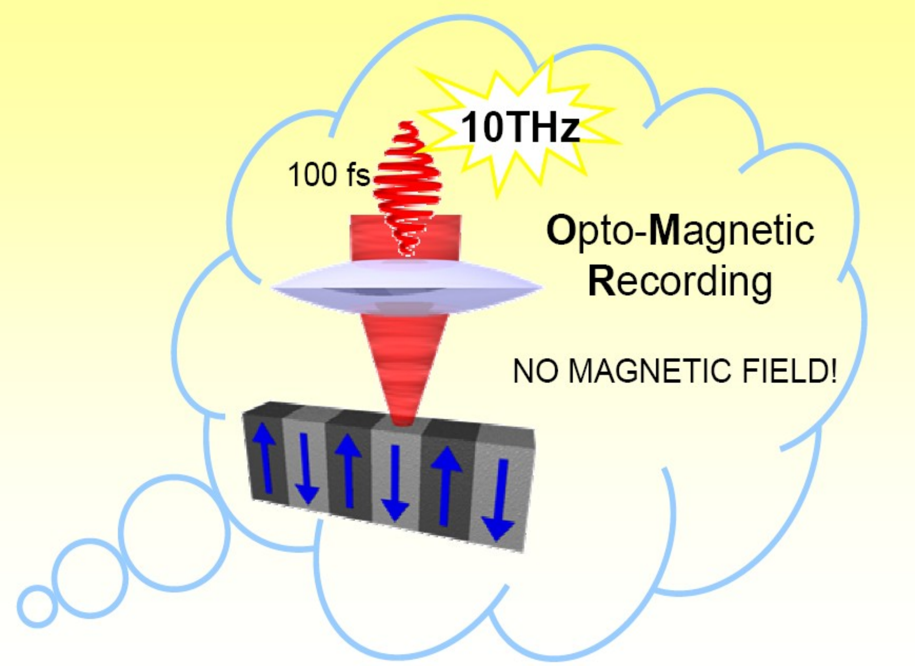




E.Beaurepaire et *al*.
Phys. Rev. Lett. **76**, 4250 (1996)

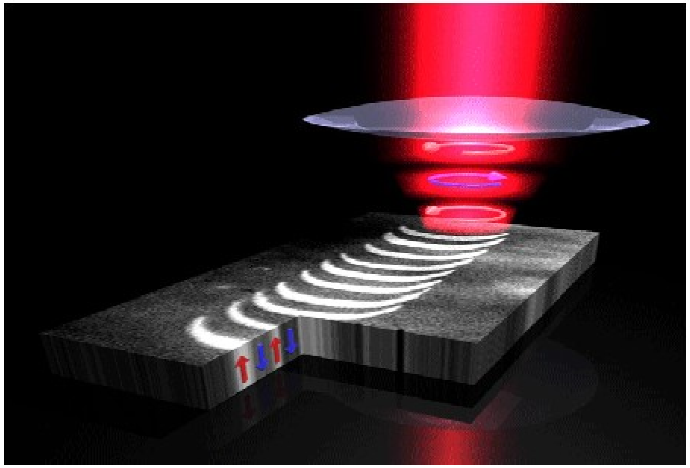


Can light reverse M?



Magnetization reversal induced by a single 40 fs laser pulse.

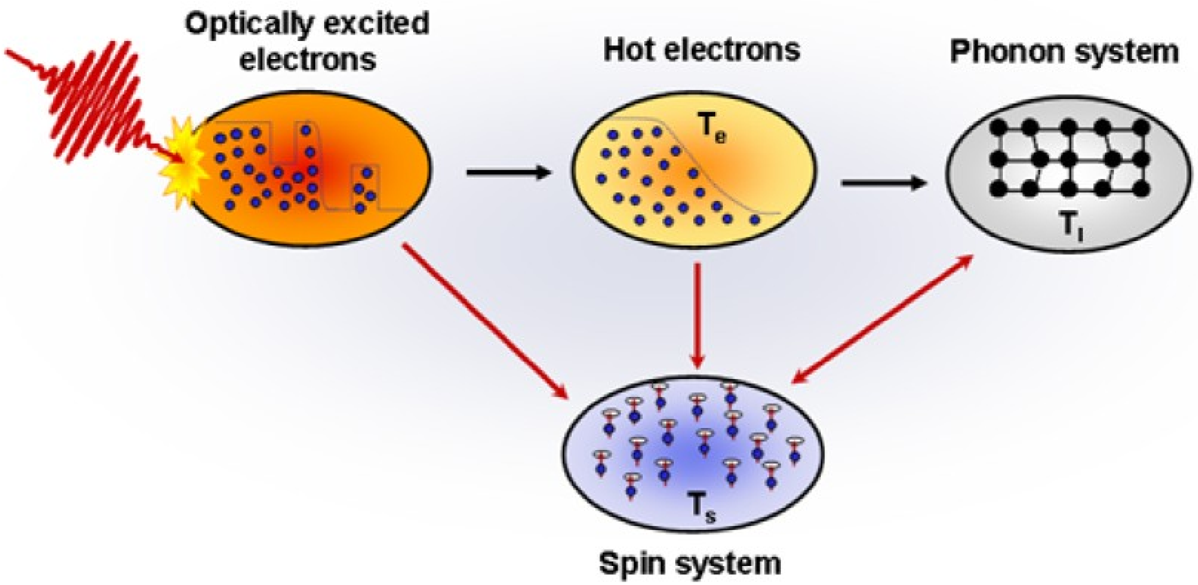
.Each domain is written with a single 40 fs laser pulse.
.Magnetization reversal must occur within 1 ps.
.Femtosecond (THz) opto-magnetic switch is faisible.



C.D. Stanciu et al.:

First demostration of all-optical
Magnetic recorsing – 5microns bits

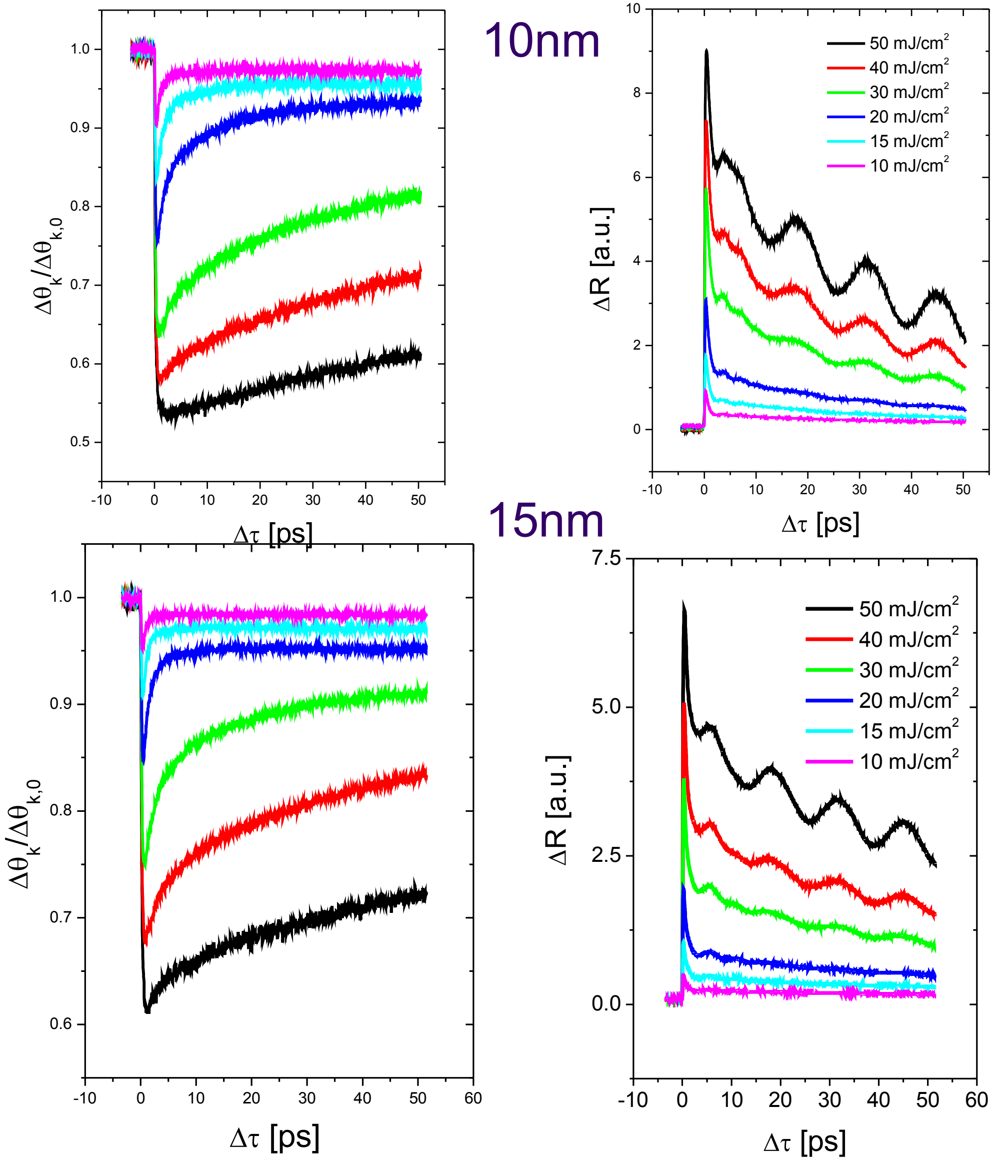
Femtosecond dynamics chalenges:



Femtosecond pump-probe experiment allows to investigate the dynamics of electron, phonon and spin systems in non-equilibrium conditions

- The physics of the dynamics even in simple 3d metal such as Ni is not understood.
  - Direct spin-momentum transfer to electron system is discarded
  - Excitation of non-magnetic states mediated by enhanced spin-orbit coupling
  - No inverse Faray effect
  - Thermal mechanism ?!

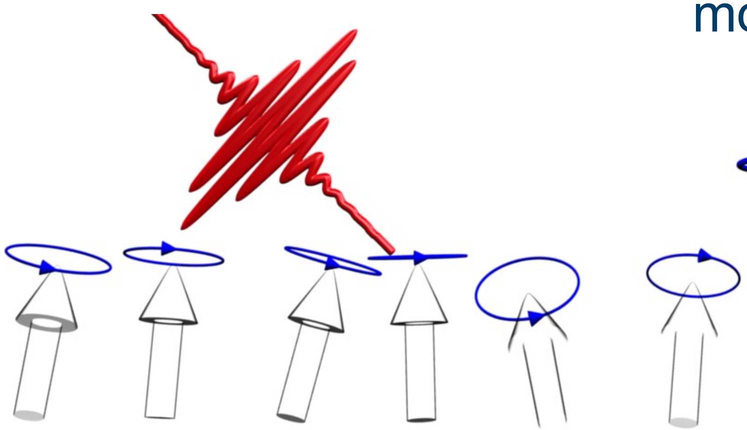
Experimental measurements: Kerr signal and Reflectivity dynamics in Ni thin films

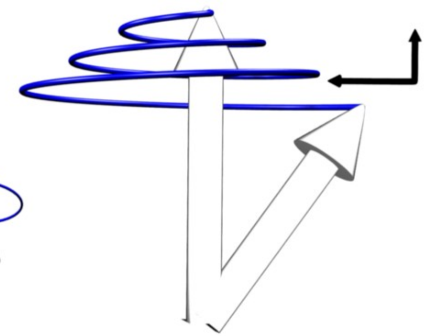


- ➢Fs demagnetisation + ps recovery
- ➢Slowing down of the magnetisation rates as a function of pump fluency
- ➢Excitation of incoherent stress Waves at high pump fluency

Schematics of the model

Schematics: Laser excitation … in a thermal macrospin model

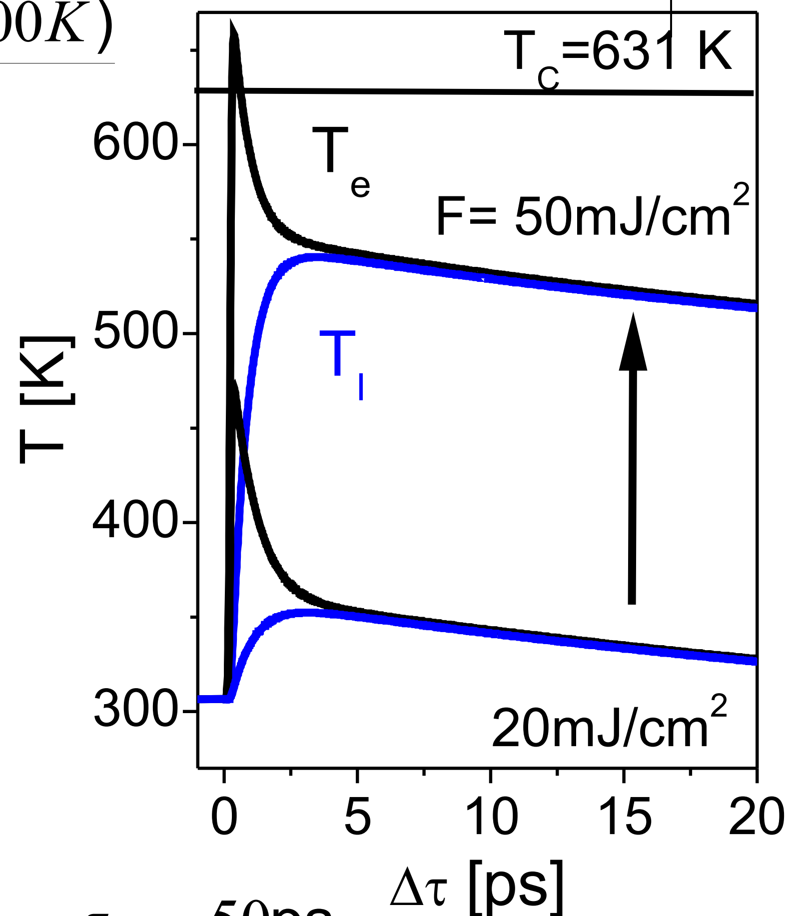




Theoretical model: 2T model

$$C_e \frac{\mathrm{d}T_e}{\mathrm{d}t} = -G_{el}(T_e - T_l) + P(t) - \frac{(T_l - 300K)}{\tau_{th}}$$

$$C_l \frac{\mathrm{d}T_l}{\mathrm{d}t} = G_{el}(T_e - T_l)$$



- Assumptions (simplification):

$$C_e = \gamma T_e \ , \quad \gamma = 3 \cdot 10^2 \ \frac{\mathrm{J}}{\mathrm{m^3K}}$$

- Fitting results:

$$G_{el} \approx 10 \cdot 10^7 \ \frac{\mathrm{W}}{\mathrm{m^3K}}, \quad C_l = 3.1 \cdot 10^6 \ \frac{\mathrm{J}}{\mathrm{m^3K}}, \ \tau_{th} = 50\mathrm{ps}$$

Three main approaches:
(all based on the Langevin dynamics)

- Atomistic model based on the LLG (Langevin) equation
- Atomistic model based on the Landau-Lifshitz-Miyasaki-Seki equation

-> to take into account

electron-electron corelations

- Micromagnetic approach based on the Landau-Lifshitz-Bloch (Langevin) equation

-> to extend modelling size

Atomistic model

- Uses the Heisenberg form of exchange

$$E_i^{exch} = \sum_{j \neq i} J_{ij} \vec{S}_i . \vec{S}_j$$

- Dynamics governed by the Landau-Lifshitz-Gilbert (LLG) equation.
- Random field term introduces the temperature (Langevin Dynamics).
- Variance of the random field determined by the electron temperature Tel.

$$\dot{\vec{S}}_i = -\frac{\gamma}{1+\alpha^2} \vec{S}_i \times H_i(t) - \frac{\lambda\gamma}{1+\lambda^2} \vec{S}_i \times (\vec{S}_i \times \vec{H}_i(t))$$

$$< h_j(t) >= 0 \qquad < h_i(0) h_j(t) >= \delta(t) \delta_{ij} 2 \lambda k_b T / \gamma$$

The Landau-Lifshitz-Bloch equation [D.Garanin,PRB 55 (1997) 3050] :

$$\dot{\mathbf{m}} = \gamma\left[\mathbf{m}\times\mathbf{H}_{eff}\right] + \frac{\gamma\alpha_{||}}{m^2}\left(\mathbf{m}\cdot(\mathbf{H}_{eff})\mathbf{m}\right) - \frac{\gamma\alpha_{\perp}}{m^2}\left[\mathbf{m}\times\left[\mathbf{m}\times\left(\mathbf{H}_{eff}\right)\right]\right]$$

$$T_C = 631\text{K}$$

$$M_S = 500\frac{\text{emu}}{\text{cm}^2}$$

$$\mathbf{H}_{app} = 1500\text{Oe}$$

- Using two relaxations parallel and perpendicular
- Magnetisation magnitude is no conserved
- Entropy correction

•LLB is coupled to 2T model

•Temperature-dependent parameters from MFA

$$\alpha_{||} = 2\lambda T / 3T_C$$

$$\alpha_{\perp} = \lambda[1 - T / 3T_C]$$

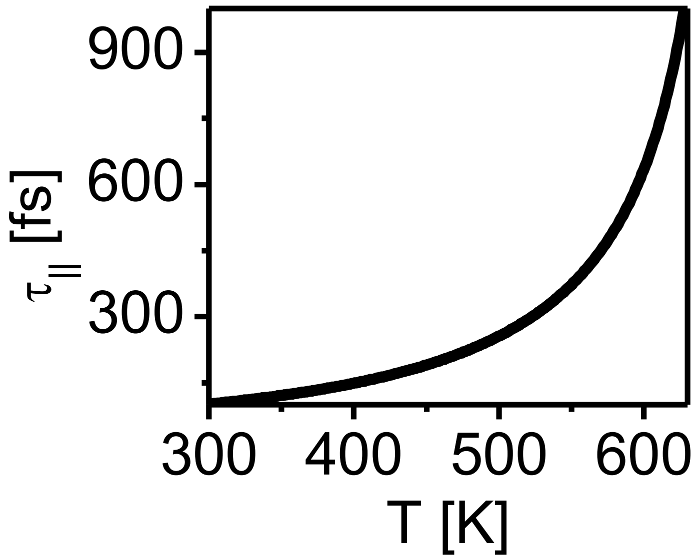
$$\alpha_{\perp}(300\text{K}) = 0.04 \Rightarrow \lambda = 0.045$$

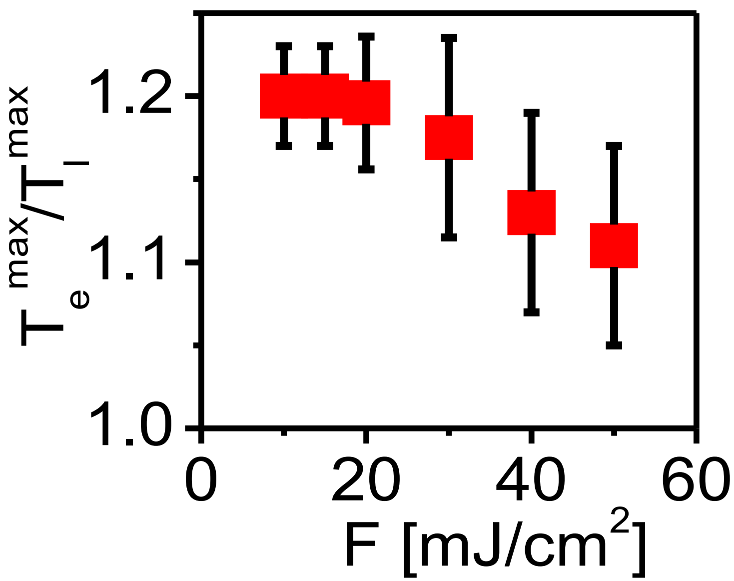
Slowing down of the electron temperature increase

- Nonlinear electron specific heat dependence on temperature?
- Phonon contribution to reflectivity?
- Excitation of coherent stress waves?

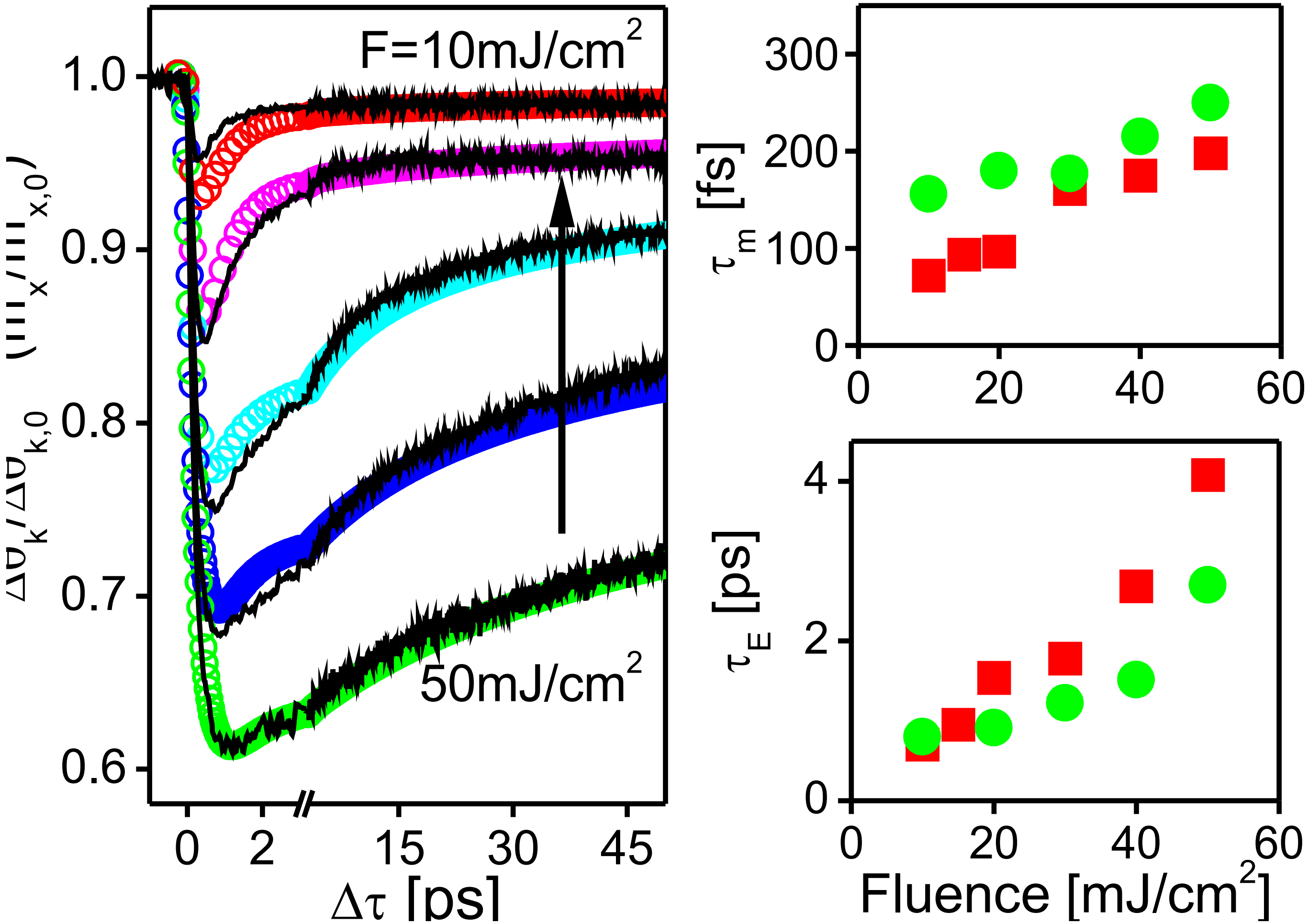
Critical slowing down for strong demagnetization

$$\tau_{\|} = \frac{\chi_{\|}(H,T)}{\gamma \alpha_{\|}(T)}$$





Simulation and experimental results for Ni 15nm thin film

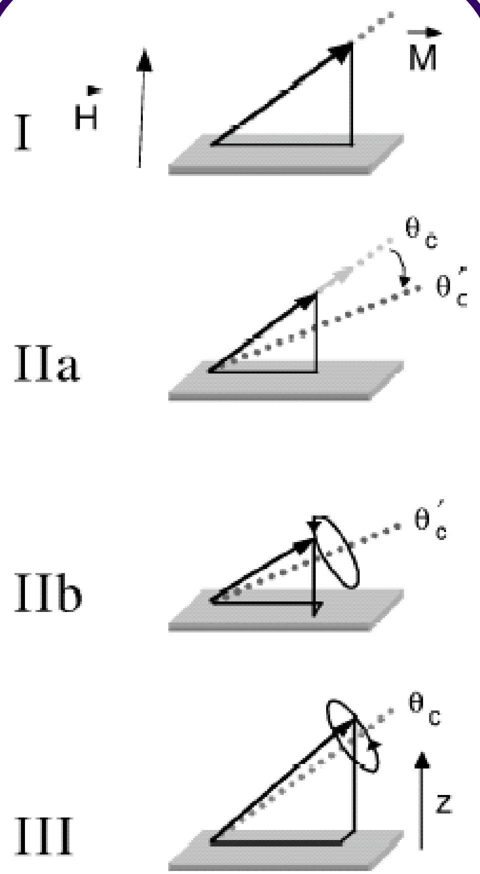


slowing down

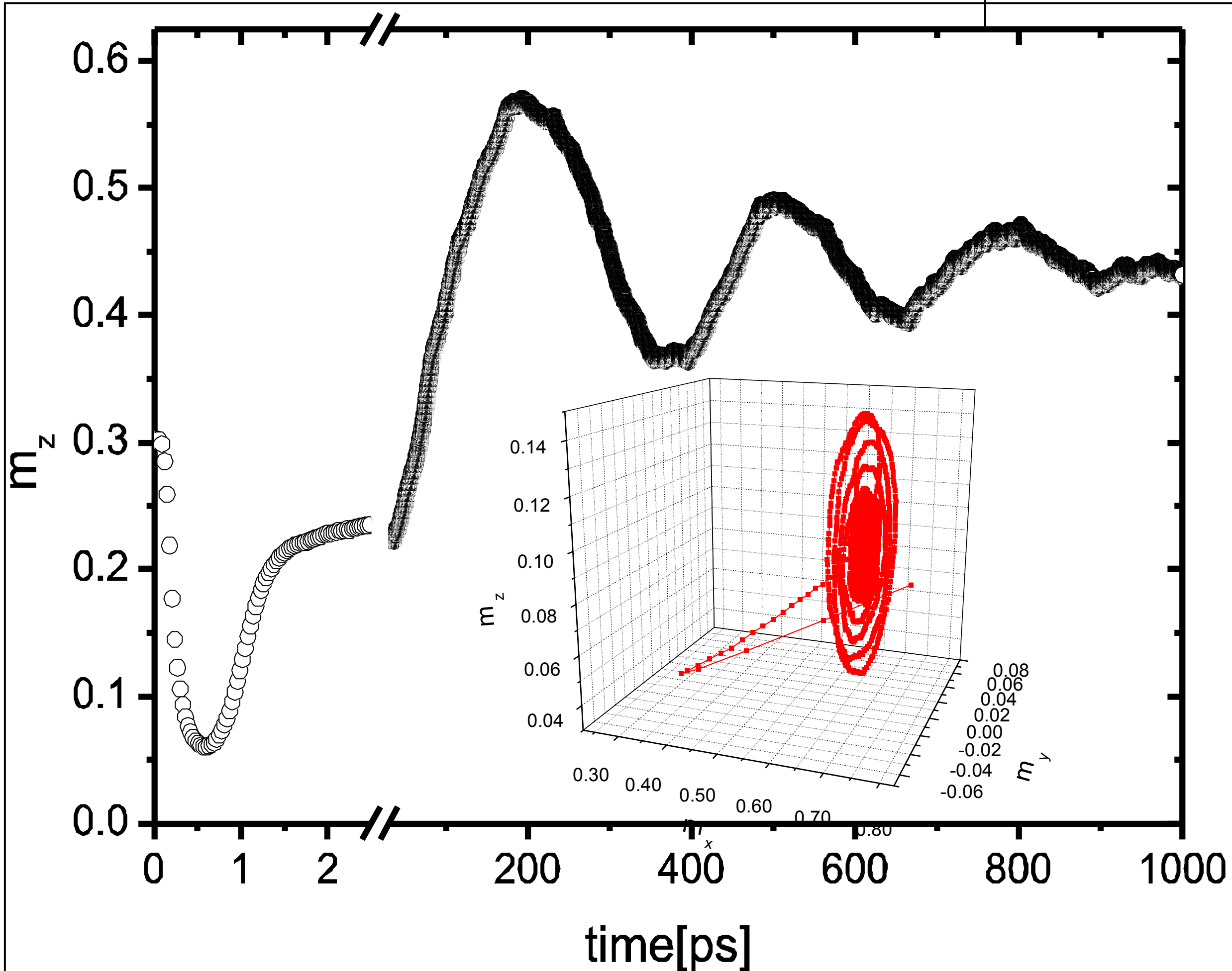
slowing down

Thermal character of demagnetization mechanism revealed

Modelling of optically-induced precession.



M.van Kampen et al, PRL **88** (2002)227201
(experiment)



Precessional frequency decreases with laser pulse fluency

Langevin dynamics based on the LLG: the white noise approximation is not always valid

- The electron-electron correlation time in metals

is of the of 10 fs

- The electron-phonon correlation time is of the order of 1ps
- Strong fields (including exchange field) have characteristic frequencies of the order of inverse correlation time
- The spin and phonon systems are not at equilibrium, the fluctuation-dissipation theorem should be avoided.

It is necessary to introduce correlated noise!

The Ornstein-Uhlenbeck process

Equilibrium angle distributions:

$$\varsigma = \mu_0 H / k_B T, \quad \tau_c = 10\, fs$$

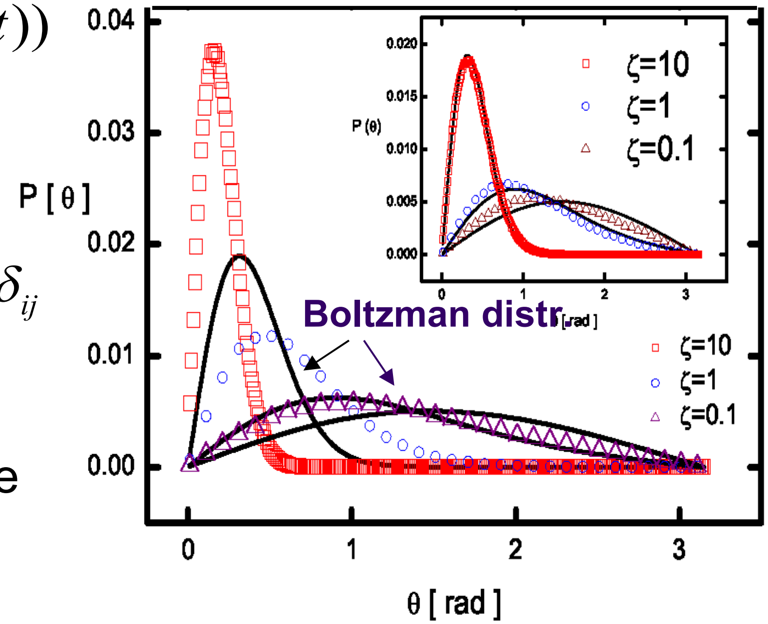
$$\dot{\vec{S}}_i = -\frac{\gamma}{1+\alpha^2}\vec{S}_i \times H_i(t) - \frac{\alpha\gamma}{1+\alpha^2}\vec{S}_i \times (\vec{S}_i \times \vec{H}_i(t))$$

$$\vec{H}_i = \vec{H}_i + \varepsilon_i$$

$$< \varepsilon_i(t) >= 0, \quad < \varepsilon_i(t)\varepsilon_j(t') > = \frac{D}{\tau_c}\exp(-|t-t'|/\tau_c)\delta_{ij}$$

Diffusion coefficient Correlation time

$$D = \frac{\alpha k_B T}{\gamma \mu_0}$$



- For small temperatures (additive noise), the diffusion coefficient is re-normalised
- For large temperatures (multiplicative noise), the distribution is not Bolzman

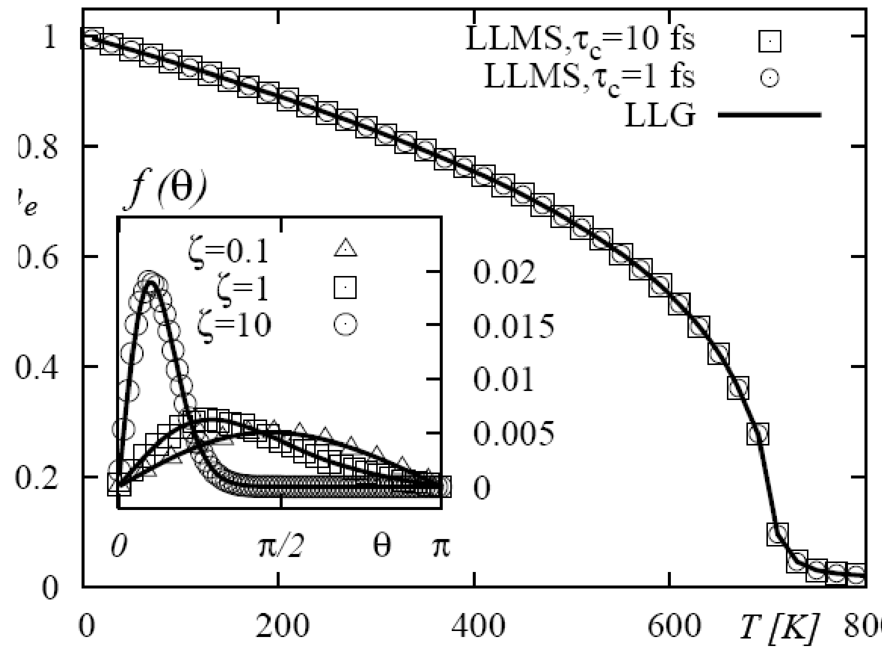
Landau-Lifshitz-Miyazaki-Seki (LLMS) equations

$$\dot{\mathbf{s}}_i = \gamma[\mathbf{s}_i \times (\mathbf{H}_i + \boldsymbol{\eta}_i)],$$

$$\dot{\boldsymbol{\eta}}_i = -\frac{1}{\tau_c}(\boldsymbol{\eta}_i - \chi \mathbf{s}_i) + \mathbf{R}_i$$

$$\langle \mathbf{R}_i(t) \rangle = 0$$

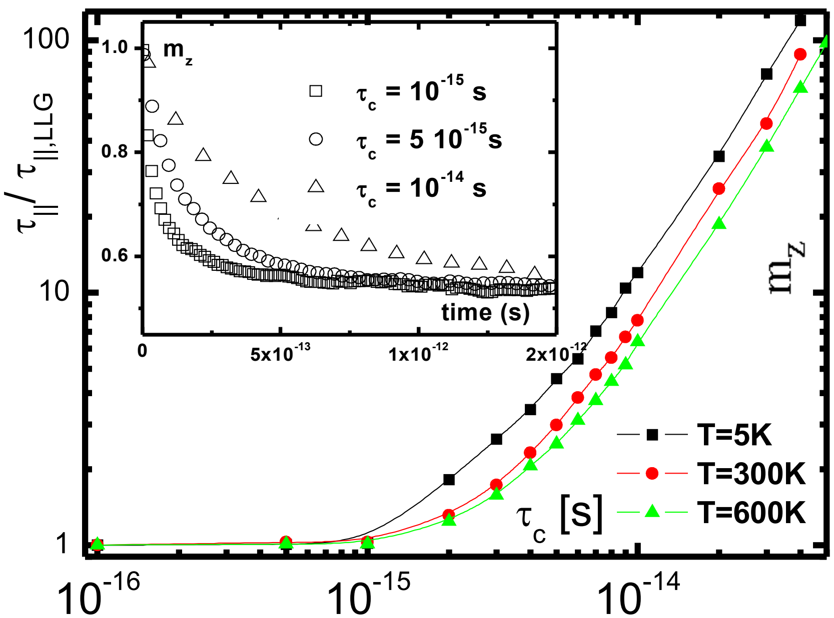
$$\langle \mathbf{R}_i(t)\mathbf{R}_j(t') \rangle = (2\chi k_B T/\tau_c)\delta_{ij}\delta(t - t')$$



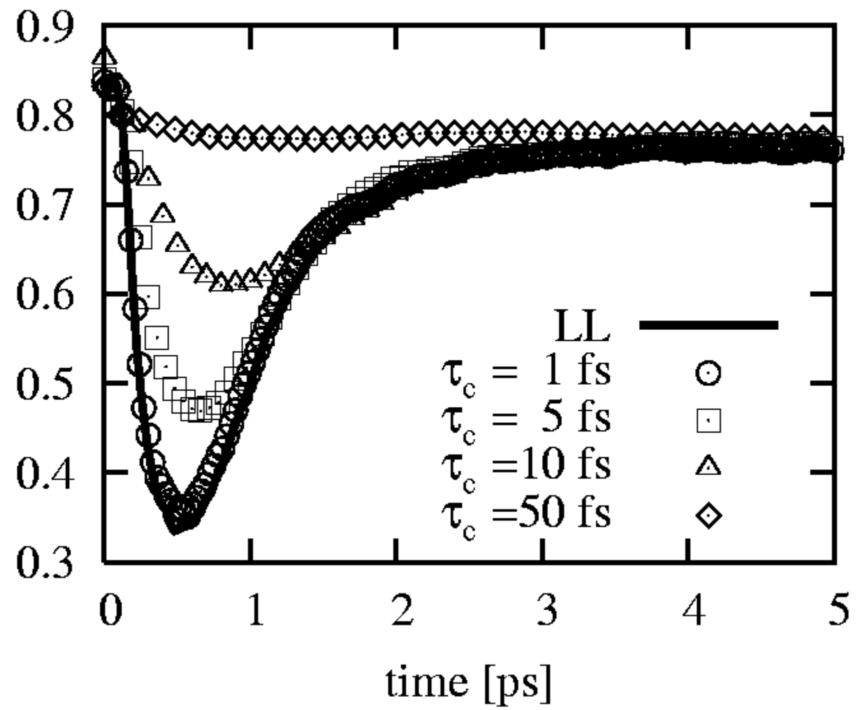
- The coupling term χ describes the adjustment of the noise to the spin direction
- When τ_c ->0, the LLG equation is recovered
- We generalize the LLMS equations to many spin case.

$$\boxed{\alpha = \gamma\chi\tau_c}$$

Longitudinal relaxation as a function of correlation time



Modelling of the ultra-fast pump-probe experiment for different correlation times



64x64x64 spins (Ni)

Main features of this new approach and conclusions

- The fluctuation-dissipation theorem is applied to the electron system only
- The spin and electron systems do not need to be

in the equilibrium with each other

- It is thermodynamically consistent
- The exact values of τcorr and χ are subject of ab-initio calculations

- If τcorr ~10 fs the longitudinal relaxation is affected

by correlations but the perpendicular relaxation (αLLG) is not.

- The slowing down of demagnetisation rates as a function of correlation time

(different materials have different demagnetisation rates,

Should be possible to control with dopping, observed in half metals)

Reference: U Atxitia et al Phys. Rev. Lett. 102 (2009) 055013

CONCLUSIONS

- For the long-timescale:

  - Energy barrier determination is essential for the long-time magnetisation decay
  - Energy barriers of nanoparticles which are not circular or elliptical are not KV
  - Energy barriers are changing in time due to magnetic interactions
  - The kinetic Monte Carlo combination with simulated annealing is capable to determine magnetisation decay for arbitrary timescale.

CONCLUSIONS

- For ultra-short timescale:

- We have shown that the Langevin dynamics approach adequately describes all stages of laser-induced dynamics:

- **Femtosecond linear demagnetisation.**
- **Picosecond magnetisation recovery.**
- **Laser-induced precession**.

- The main contribution to the slowing down of ultrafast demagnetisation rate comes from the slowing down of the longitudinal relaxation approaching Tc

- In some extreme conditions with characteristic timescale of the order of electron-electron correlation time a coorelated noise approach is necessary

- We have introduced an approach based on the Landau-Lifshitz-Miyazaki-Seki equation.