Magnetisation dynamics at different timescales: dissipation and thermal processes.

Numerical modelling methodology.

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Objective: large-scale modelling of complex ferromagnetic materials









Patterned FePt magnetic media

Fe elongated nanoparticles prepared by extrusion





CoCrPt magnetic Self-organized Co nanoparticles



FePt nanoparticles

recording media



SmCo for hard magnets

FePt nanoparticles Prepared by laser ablation



Very soft magnetic material: Finemet

Objective: modelling of technological processes





Conventional magnetic recording



Ultra-fast (fs) Kerr dynamics

Heat-assisted magnetic recording



All-optical magnetic recording

Introduction

- Magnetic system is not isolated, the magnetisation change can occur at any timescale.
- Magnetism is a quantum phenomena.
- Ab-initio calculations, although rapidly developing, at the present state of art are not capable to calculate magnetisation dynamics in complex materials at arbitrary timescale and temperature.
- At larger spatial scale, relatively large magnetisation volumes (10nm) can be considered as classical particles.



The exchange term: micromagnetics versus spin models

•Micromagnetics calculates the magnetostatic fields exactly but which is forced to introduce an approximation to the exchange valid only for long-wavelength magnetisation fluctuations.

•The exchange energy is essentially short ranged and involves a summation of the nearest neighbours. Assuming a slowly spatially varying magnetisation the exchange energy can be written

$$E_{exch} = \int W_e dv$$
, with $W_e = A(\nabla m)^2$

with

$$(\nabla \mathbf{m})^2 = (\nabla m_x)^2 + (\nabla m_y)^2 + (\nabla m_z)^2$$

The material constant $A = JS^2/a$ for a simple cubic lattice with lattice constant a. A includes all the atomic level interactions within the micromagnetic formalism.

 $E_i^{exch} = \sum J_{ij} \vec{S}_i \cdot \vec{S}_j$

•Atomistic models are discrete and use the Heisenberg form of exchange

Micromagnetic models of nanostructured materials



Models need nanostructure and micromagnetic parameters from experiment

Different timescales: Electron-spin All-optical Langevin dynar							es: Langevin dynamics	
	10-1-5 1	S	relaxation processes	las ex	er-pulsed periments		on atomistic level	
	10-11s p	S	Magnetisation	Fast-Kerr			Langevin dynamics	
	10 ⁻⁹ s m	IS	precession.	measurements, FMR, synchrotro	s, tron	level		
				radiation studie		ies	all success to the	
	10 -6s μs						acceleration	
	10-3s n	ns	Hysteresis		Convention	al 👘	techniques	
			measurement	S.	(VSM SQUID)			
	10 ⁻⁰ s s	;		(*****,****				
	10 ³ s k	ıs	Magnetic visa	cosi	osity		kinetic Monte	
	10 ⁶ s month						Carlo with energy	
			l ona-time th	ner	ermal stability		calculations	
	10 ⁹ s ye	ears	for magnetic recording.					

Natural magnetisation dynamics: **100 pico- 100** nano-second timescale

Outline for today: 100ps-100ns (natural) dynamics

- Non-thermal dynamics:
- Ferromagnetic resonance
- Basic dynamical equation: the Landau-Lifshitz-Gilbert
- The problem of magnetic damping (α): main processes
- Thermal dynamics:
- Principles of the Langevin dynamics.
- Modelling of thermal spinwaves
- The Landau-Lifshitz-Bloch micromagnetics for dynamics close to Tc

Ferromagnetic resonance(FMR): (Arkadiev, 1911; Kittel, 1947)

A ferromagnetic body under applied field has a maximum absorption in frequencies:



The absorption peak contains information about anisotropy field.



Lorentzian absorption line typical of FMR showing microwave power absorption as a function of swept bias field. Precession and relaxation of **M** in response to an applied field **H**.

Torque on magnetisation

$$\frac{1}{\gamma}\frac{\partial M}{\partial t} = -[M \times H_0]$$



The absorption line width contains Information on damping processes

Ferromagnetic resonance

- The experiment is normally performed in almost saturated conditions.
- The absorption peak contains information about anisotropy field.
- The linewidth contains information about dissipation processes.

FMR tecniques as a probe of **magnetisation dynamics** In-plane anisotropy in square array of Py dots

Py square lattice of closely packed circular dots, $1/1.1 \,\mu m$





Courtesy of G.Kakazei et al

The Landau-Lifshitz (LL) and the Landau-Lifshitz-Gilbert (LLG) equations of motion

(for magnetization vector):

LL equation $\frac{d\vec{M}}{dt} = -\gamma'_{0} \left[\vec{M} \times \vec{H}\right] - \frac{\alpha_{LL}\gamma_{0}}{M_{s}} \left[\vec{M} \times \left[\vec{M} \times \vec{H}\right]\right]$ **Gilbert equation** (physically more reasonable

for large damping) Gilbert damping, 1955

$$\frac{d\vec{M}}{dt} = -\gamma_0 \left[\vec{M} \times \vec{H}\right] + \frac{\alpha_G}{M_s} \left[\vec{M} \times \frac{d\vec{M}}{dt}\right]$$



How the Gilbert equation could be transformed into the LL equation → LLG equation

$$\vec{M} \times \frac{d\vec{M}}{dt} = -\gamma_0 \vec{M} \times \left[\vec{M} \times \vec{H}\right] + \frac{\alpha_G}{M_s} \vec{M} \times \left[M \times \frac{d\vec{M}}{dt}\right]$$
$$\vec{M} \left(\vec{M} \cdot \frac{d\vec{M}}{dt}\right) - \frac{d\vec{M}}{dt} \left(\vec{M} \cdot \vec{M}\right)$$
$$= 0 \qquad = M_s^2$$

The LL eq. is equivalent to G equation with substitutions

$$\gamma'_0 = \frac{\gamma_0}{1 + \alpha_G^2}, \qquad \alpha_{LLG} = \frac{\alpha_G}{1 + \alpha_G^2}$$

Convenient form of the LLG equation:

$$\vec{m} = \frac{M}{M_s}, \quad E' = E/(2KV)$$

$$\tau = \gamma H_K t / \left(1 + \alpha_G^2\right)$$

$$\vec{h} = \frac{\vec{H}_{\text{int}}}{H_K} = \frac{\partial E'}{\partial \vec{m}},$$
$$H_K = \frac{2K}{M_s}$$

Contains all contributions: anisotropy, Exchange, magnetostatic, Zeeman, depends on M

- Anisotropy field



The Bloch-Bloembinger damping:

$$\left(\frac{d\vec{M}}{dt}\right)_{X,Y} = -\gamma_0 \left[\vec{M} \times \vec{H}\right]_{X,Y} - \frac{1}{T_2} M_{X,Y}$$

Transverse relaxation

$$\left(\frac{d\vec{M}}{dt}\right)_{Z} = -\gamma_{0} \left[\vec{M} \times \vec{H}\right]_{Z} + \frac{1}{T_{1}} (M_{s} - M_{Z})$$

Longitudinal relaxation

The problem of damping:

- Different relaxation processes:
- Magnon-magnon scattering
- Magnon-electron interactions (especially in metals)
- Phonon-magnon interactions (magnetostriction)
- Impurities
- Extrinsic factors (grain boundary, surface roughness, etc.)
- Temperature disorder

-Theory of magnetic damping constant (α):





Kittel formula for spinwaves dispersion relation:

Anisotropic single crystal ferromagnet: Angle between M and k $\left(\frac{\omega}{\gamma}\right)^2 = \left(H_0 + H_A + Ak^2\right)\left(H_0 + H_A + Ak^2 + 2\pi M_s \sin^2 \theta_k\right)$ Exchange **Magnetostatic** Applied Anisotropy interaction interaction field



field



Magnons and their interactions:

- Classical spinwaves correspond to quasiparticles called magnons.
- Homogeneous magnetisation (FMR mode) corresponds to magnon with k=0.

magnon scattering

2

• Linear normal modes (magnons) do not interact. Nonlinear processes correspond to magnonmagnon interactions. Magnon decay Two magnon merging

3

3

2

These interactions define kinetic effects (e.x. heat conductivity) and width and shape of the FMR line and magnon lifetime

Nonlinear phenomena: Suhl instabilities.

 For large excitation power - FMR saturation occurs

k

k=0

 If the density of magnons gets higher than critical value – the homogeneous oscillations become unstable

$$\omega_0 = \omega(k) + \omega(-k) = 2\omega(k)$$

The occurrence of the instability depends on the system geometry and is governed by the applied field.

$$2\omega_0 = \omega(k) + \omega(-k) = 2\omega(k)$$
 Second

Second condition, more easy to meet

Inherent relaxation processes (via spin-wave instabilities)

 Even without external dissipation it is possible to reach magnetisation reversal via spin-wave instabilities.



Main non-inherent relaxation processes:

- o Direct spin-lattice relaxation due to nonuniformities
- Heterogeneity of composition
- Polycrystalline structure (grain coundaries, etc.)
- Nonuniform stresses, dislocations
- Geometrical roughness: pores, surfaces etc.
- O Indirect spin-lattice relaxation
 Via ions with strong spin-orbital coupling
 - Via charge carriers

The problem of damping (α)

- Although there exist theories trying to evaluate the damping parameter basing on a particular mechanism, the comparison with experiment remains poor.
- Normally the Gilbert damping α is a phenomenological parameter, taken from the experiment.
- The values from FMR and direct measurement of magnetisation switching (fast Kerr measurements) not always coincide.

Observation of the precessional dynamics:

W.K.Hiebert etal, PRB, PRL, Nature (2002) Scanning optical microscope





Simulation with LLG

Dinamical effects: Precesional switching: Faster and less field. Landau-Lifshitz-Gilbert (LLG):

Experiment with ps field pulses perpendicular to the magnetisatrion (C.Back et al, Science, 1999) Fe/GaAs



150 µm



•H || M –non-precessional switching •Prcessional switching is faster, however, the ringing phenonema occur.

Simulación LLG



Comparison of Patterns



Observed (SEMPA)



- 100 μm

From Ch.Stamm- SLAC overview

Calculated (fit using LLG)

Anisitropies same as FMR Damping α = 0.017 **4x** larger than FMR WHY? Additional angular momentum dissipation? - spin current pumped across interface into paramagnet causes additional damping (SPIN ACCUMULATION)

Thermal effects

Thermal fluctuations play very important role in magnetisation dynamics:

At the microscopic level:

- At the equilibrium they are responsible for <u>thermally</u> <u>excited spinwaves</u>.
- Spinwaves are responsible for thermal magnetisation reversal via the spinwave instabilities and energy transfer to main reversal mode.

At more macroscopic level

- •Thermal fluctuations are responsible for random walk in a complex energy landscape
- •Eventually <u>energy barriers</u> could be overcome with the help of thermal fluctuations leading to magnetisation decay.

*The theory of thermal magnetization fluctuations of single domain, non-interacting particles was introduced by W.F.Brown (*W.F.Brown Phys Rev **130** (1963) 1677)

"We now suppose that in the presence of thermal agitation, "the effective field" describes only statistical (ensemble) average of rapidly fluctuating random forces, and that for individual particle this expression must be augmented by a term h(t) whose statistical average is zero"

 $< h(t) >= 0, \quad < h(t)h(t+\tau) >= \mu \delta \delta(\tau), \quad i,j = x, y, z$

"The random-field components are formal concepts, introduced for convenience, to produce the fluctuations δM " **W.F.Brown outlined two methods:** -Based on the fluctuation-dissipation theorem -Imposing the condition that the equilibrium solution of the $2E_0k_BEr$ -Planck equation is the Boltzman distribution $\mu = \frac{1}{M_s V_i (1 + \alpha^2)}$

As a result of both in a non-interacting system:

Thermal micromagnetics Langevin dynamics approach

$$\frac{d\vec{M}}{dt} = -\frac{\gamma}{1+\alpha^2}\vec{M} \times \vec{H} - \frac{\gamma\alpha}{M_s(1+\alpha^2)}\vec{M} \times (\vec{M} \times \vec{H})$$

$$\vec{H} = \vec{H}_{Zeeman} + \vec{H}_{aniso} + \vec{H}_{exch} + \vec{H}_{magnetost} + \vec{H}_{therm}$$

$$< H_{therm,i}(t) >= 0, \quad < H_{therm,i}(t)H_{therm,j}(t') >= \frac{2ok_BT}{M_sV}\delta_{ij}\delta(t-t')$$

Initially introduced for nanoparticlesThis was brought to micromagnetics.

No correlations Between time and different particles!!

W.F.Brown, Phys Rev 130 (1963) 1677.

Note on the damping and thermal processes.

- In principle, the Gilbert (or other) form of damping is as a result of spin coupling with the oscillator thermal bath, in this sense, the thermal fluctuations are already included into the damping term.
- In some approximations, the undamped LL equation is coupled to a system of oscillators (phenomenological phonon bath) and the resulting LLG damping is derived.

Fokker-Plank equation for isolated nanoparticle :

$$\frac{\partial P}{\partial \tau} = -\frac{\partial}{\partial \vec{m}} \left[-\vec{m} \times \vec{h} - \alpha \, \vec{m} \times \left(\vec{m} \times \vec{h} \right) + D \, \vec{m} \times \left(\vec{m} \times \frac{\partial}{\partial \vec{m}} \right) \right] P$$

Diffusion coefficient (strength of fluctuations)

$$P_{eq}(\vec{m}) \propto \exp\left[-E(\vec{m})/k_BT\right]$$
$$D = \frac{\alpha k_B T}{M_s V}$$

Boltzmann distribution in the equilibrium

The noise can be introduced either to precessional term or to both damping and precessional terms

Problem of numerical scheme

$$\frac{dM}{dt} = -\frac{\gamma}{1+\alpha^2} \vec{M} \times \vec{H} - \frac{\gamma \alpha}{M_s (1+\alpha^2)} \vec{M} \times (\vec{M} \times \vec{H}) \qquad \vec{H} = \vec{H}_{int} + \vec{H}_{thermal}$$

- The noise is multiplicative although for small deviations additive.
- Ito & Stratonovich interpretation of stochastical differential equations- two different interpretations of stochastical integrals:
- The Ito evaluates the integral on the lower point of the integration interval while the Stratonovich – in the middle one.

$$\int_{t_n}^{n+1} B(t,m) \circ dW_t \approx \frac{1}{2} \Big[B(t_{n,m_n}) + B(t_{n+1,m_{n+1}}) \Big] \Delta W_n$$

- The Ito intepretation produces a stochastical drift.
- Stratonovich interpretation should be used, for example the Heun numerical scheme^k.
- However, if after each integration step the magnetisation is renormalized – normal scheme could be used**: $m_i = m_i / \sqrt{m_{i,x}^2 + m_{i,y}^2 + m_{i,z}^2}$

J. Garcia-Palacios et al, Phys Rev B 58 (1998) 14937 D.Berkov et al J.Phys:Cond Mat 14(2002) 281

Generalisation of the Langevin dynamics to many spin problem:

Although the thermal fluctuations properties were derived for only non-interacting particles, the same form of the Langevin-LLG equation is used to calculate the switching properties even in an interacting system.

- The main assumption is that the noise is uncorrelated in time (no memory effects, separation of timescales.
- Around the equilibrium the formalism of the Onsager coefficients can be done for many spin system which shows that for particular damping (LLG) for many spin system no correlation between particles exist.
- In a general case Fokker-Plank equation no solution exists.

O.Chubykalo et al J. Magn.Magn.Mat 226, (2003) 28

Langevin dynamics based on the Landau-Lifshitz-Gilbert equation.

could be formulated for both

•Atomistic spins (localized classical magnetic moments μ in the Heisenberg description with J and on-site anisotr. d), $\alpha(\lambda)$ defines coupling to thermal bath Characteristic timescale is determined by exchange; (fs-ps)

•Micromagnetic units (averaged magnetisation, Ms(T)), A(T), K(T)

The temperature in this case is included twice:

The damping α contains already thermal averaging: α(T)
 Langevin dynamics defines different trajectories
 Characteristic timescale is determined by anisotropy;
 (ps-ns)

Modelling of thermal spinwaves

• Langevin dynamics calculations have been carried out for approximately 10 precessional periods • Fourier transform in both space and time has been performed $\vec{m}_x(\vec{r},t) = \sum_{\vec{k},\omega} f(\omega_k,k) \exp[i(\vec{k}\vec{r}-\omega_k t)]$



Thermal Langevin dynamics: micromagnetics versus atomistic spin (Heisenberg) model

Atomistic (classical) Heisenberg model for FePt (parametrised through ab-initio)



Langevin dynamics based on the micromagnetic Landau-Lifshitz-Gilbert equation.

Scaling approaches – correctly scale M(T), K(T), A(T)

with discretization size within micromagnetics.

 $\begin{aligned} \frac{d\vec{M}_{i}}{d\tau} &= -\vec{M}_{i} \times \vec{H}_{i} - \alpha \ \vec{M}_{i} \times \left(\vec{M}_{i} \times \vec{H}_{i}\right) \\ \tau &= \gamma_{0} t / (1 + \alpha^{2}), \quad \vec{H}_{i} = (-1 / M_{s} V_{i}) (\delta E / \delta \vec{M}_{i}) \\ \vec{H} &= \vec{H}_{int} + \vec{h}_{therm} \\ < h_{i} >= 0, \qquad < h_{i}(t) h_{j}(t + \tau) >= \frac{2\alpha k_{B} T}{M V (1 + \alpha^{2})} \delta(\tau) \delta_{ij} \end{aligned}$

Langevin dynamics for the micromagnetics does not correctly describe spinwaves: . The spectrum is cut

The spectrum is cut and Tc is not correct
density of states is not correct.



FIG. 1. M vs \tilde{T} curves for the model Permalloy cube, from LLG Eq. (6), with parameters as given in text, and unrenormalized (square symbols) and renormalized (oval symbols) values of the exchange constant.

G.Grinstein and R.H.Koch, PRL 90 (2003) 207201.

Atomistic modelling of magnetisation reversal

Field applied at 30°

Field applied at 135°



64³ magnetic moments on cubic lattice



•Magnetisation magninute is not conserved

•Damping is enhanced at high T

Temperature-dependent magnetisation dynamics cannot be described within standard LLG approach.

O.Chubykalo-Fesenko et al, Phys Rev B 74 (2006) 094436

Longitudinal and transverse relaxation at high T





- longitudinal relaxation time shows critical slowing down
- transverse relaxation time breaks down close to Curie temperature
- magnitude of magnetisation not constant in time (and space)

LLB equation

Transverse (LLG) term

Longitudinal term introduces fluctuations of M

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

- $\bullet\,\mathbf{macro-spin}\,\,\mathbf{polarization}$ is $\mathbf{m}=\langle\mathbf{S}\rangle$
- longitudinal (α_{\parallel}) and transverse (α_{\perp}) damping parameters are given by $\alpha_{\parallel} = \alpha_{\overline{3T_c}}^{2T}$, $\alpha_{\perp} = \alpha \left[1 \frac{T}{3T_c}\right]$
- effective field:

$$\mathbf{H}_{\text{eff}} = \mathbf{H} - \frac{m_x \mathbf{e}_x + m_y \mathbf{e}_y}{\tilde{\chi}_{\perp}} + \begin{cases} \frac{1}{2\tilde{\chi}_{\parallel}} \left(1 - \frac{m^2}{m_e^2}\right) \mathbf{m}, \ T \lesssim T_c \\ \frac{J_0}{\mu_s} \left(\epsilon - \frac{3}{5}m^2\right) \mathbf{m}, \ T \gtrsim T_c \end{cases}$$

here H is applied field and m_e is zero-field equilibrium spin polarization the second term is an expression for the anisotropy field

D.Garanin Phys Rev B, 55 (1997) 3050.

LLB versus LLG equation:

- Magnetisation length is not conserved
- Temperature dependent micromagnetic parameters
- Two relaxations: transverse and longitudinal
- Damping parameters dependence on temperature
- Valid both below and above Tc

Langevin dynamics based on the Landau-Lifshitz-Bloch equation.

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

$$\left\langle \xi_{\parallel}^{i}(t) \xi_{\parallel}^{j}(t') \right\rangle = \frac{2k_B T}{\gamma \alpha_{\parallel} M_s (T=0) V_i} \delta_{ij} \delta(t-t')$$

$$\left\langle \xi_{\perp}^{i}(t) \xi_{\perp}^{j}(t') \right\rangle = \frac{2k_B T}{\gamma \alpha_{\perp} M_s (T=0) V_i} \delta_{ij} \delta(t-t')$$

$$\int_{\text{upper definition of the set o$$

D.Garanin, O.Chubykalo-Fesenko, Phys.Rev B 70 (2004) 212409.

COMPARISON BETWEEN ATOMISTIC AND ONE-SPIN LLB SIMULATIONS





time $(\gamma J/\mu_0)$

time



Multscale approach



Multiscale modelling: all the parameters were evaluated from atomistic modelling for FePt with ab-initio input parameters (Tc= 650K)



solid line - one spin LLB



- The usual formalism for large-scale calculations of magnetic properties is Micromagnetics.
- Although different theories of magnetic damping parameters exist, due to a complexity of the problem, the damping parameter remains phenomenological.
- Thermal effects can be introduced, but the limitation of long-wavelength fluctuations means that the standard micromagnetics cannot reproduce phase transitions.
- The Landau-Lifshitz-Bloch equation is a valid micromagnetic formalism for high temperatures.