Magnetisation dynamics at different timescales: dissipation and thermal processes.

Numerical modelling methodology.

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Objective: large-scale modelling of complex ferromagnetic materials

Fe elongated nanoparticles prepared by extrusion

Self-organized Co nanoparticles

FePt nanoparticles

Lithographed Fe antidots

Patterned FePt magnetic media

CoCrPt magnetic recording media

FePt nanoparticles Prepared by laser ablation

SmCo for hard magnets

Very soft magnetic material: Finemet
Objective: modelling of technological processes

Conventional magnetic recording
- All-optical pump-probe technique
- 100 fs pulses
- 20 μJ/pulse
- 1 kHz rep. rate

Ultra-fast (fs) Kerr dynamics

Heat-assisted magnetic recording
- Ti:Sapphire laser & amplifier system

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

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

with

\[ (\nabla m)^2 = (\nabla m_x)^2 + (\nabla m_y)^2 + (\nabla m_z)^2 \]

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.

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

\[ E^{\text{exch}}_i = \sum_{j \neq i} J_{ij} \vec{S}_i . \vec{S}_j \]
Micromagnetic models of nanostructured materials

Models need nanostructure and micromagnetic parameters from experiment
Different timescales:

- $10^{-14}$ s fs: Electron-spin relaxation processes.
- $10^{-11}$ s ps: Magnetisation precession.
- $10^{-9}$ s ns: Fast-Kerr measurements, FMR, synchrotron radiation studies.
- $10^{-6}$ s μs: Conventional magnetometers (VSM, SQUID).
- $10^{-3}$ s ms: Hysteresis measurements.
- $10^{-0}$ s s: Magnetic viscosity experiments.
- $10^{3}$ s hs: Long-time thermal stability for magnetic recording.
- $10^{6}$ s month: Langevin dynamics on atomistic level.
- $10^{9}$ s years: Langevin dynamics on micromagnetic level.
- $10^{0}$ s s: Dynamics acceleration techniques.
- $10^{3}$ s hs: Kinetic Monte Carlo with energy barriers calculations.
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 ($\alpha$): main processes

- Thermal dynamics:
  - Principles of the Langevin dynamics.
  - Modelling of thermal spinwaves
  - The Landau-Lifshitz-Bloch micromagnetics for dynamics close to $T_c$
Ferromagnetic resonance (FMR): (Arkadiev, 1911; Kittel, 1947)

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

\[ \omega = \gamma \sqrt{H + (N_x - N_z)M[H + (N_y - N_z)M]} \]

The absorption peak contains information about anisotropy field.

Torque on magnetisation

Precession and relaxation of \( \mathbf{M} \) in response to an applied field \( \mathbf{H} \).

Lorentzian absorption line typical of FMR showing microwave power absorption as a function of swept bias field.

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 techniques 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 µm

Resonance field (Oe)

Azimuthal angle φ (degrees)

Amplitude (arb. units)

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)

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

**Landau-Lifshitz damping, 1935**

**Gilbert damping, 1955**

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

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

The LL eq. is equivalent to G equation with substitutions

\[ \gamma'_0 = \frac{\gamma_0}{1 + \alpha_G^2}, \quad \alpha_{LLG} = \frac{\alpha_G}{1 + \alpha_G^2} \]
Convenient form of the LLG equation:

\[ \dot{m} = \frac{\dot{M}}{M_s}, \quad E' = E / (2KV) \]

\[ \tau = \gamma H_K t / (1 + \alpha_G^2) \]

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

- Anisotropy field
The Bloch-Bloembinger damping:

\[
\begin{align*}
\left( \frac{d\vec{M}}{dt} \right)_{x,y} &= -\gamma_0 \left[ \vec{M} \times \vec{H} \right]_{x,y} - \frac{1}{T_2} M_{x,y} \\
\left( \frac{d\vec{M}}{dt} \right)_{z} &= -\gamma_0 \left[ \vec{M} \times \vec{H} \right]_{z} + \frac{1}{T_1} (M_s - M_z)
\end{align*}
\]

Transverse relaxation

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 ($\alpha$):
Ferromagnets and their spin excitations

\[ H = -J \sum_{i,j} S_i \cdot S_j - g\mu_B \sum_i S_i \cdot H \]

**1. Uniform precession (ellipsoid)**

\[ \frac{\partial \mathbf{M}}{\partial t} = -\gamma (\mathbf{M} \times \mathbf{H}_{\text{eff}}); \]

\[ \omega_0 = \gamma H_{\text{eff}}. \]

More generally:

\[ H_{\text{eff}} = -\delta E_{\text{tot}} / \delta \mathbf{M}. \]

**2. Spin Waves**

\[ \omega(k) = \omega(0) + A k^2 \]

Courtesy of K. Guslienko
Kittel formula for spinwaves dispersion relation:

Anisotropic single crystal ferromagnet:

\[
\left( \frac{\omega}{\gamma} \right)^2 = \left( H_0 + H_A + A k^2 \right) \left( H_0 + H_A + A k^2 + 2\pi M_s \sin^2 \theta_k \right)
\]

- Applied field
- Anisotropy field
- Exchange interaction
- Magnetostatic interaction

Angle between M and k

\[\om = \om_0\]

\[\hbar \om_0\]

\[k\]
Magnons and their interactions:

- Classical spinwaves correspond to quasiparticles called magnons.
- Homogeneous magnetisation (FMR mode) corresponds to magnon with $k=0$.
- Linear normal modes (magnons) do not interact. Nonlinear processes correspond to magnon-magnon interactions.

These interactions define kinetic effects (e.g., heat conductivity) and width and shape of the FMR line and magnon lifetime.
Nonlinear phenomena: Suhl instabilities.

- For large excitation power - FMR saturation occurs
- 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 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:

- **Direct spin-lattice relaxation due to nonuniformities**
  - Heterogeneity of composition
  - Polycrystalline structure (grain boundaries, etc.)
  - Nonuniform stresses, dislocations
  - Geometrical roughness: pores, surfaces etc.

- **Indirect spin-lattice relaxation**
  - Via ions with strong spin-orbital coupling
  - Via charge carriers
The problem of damping ($\alpha$)

- 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 $\alpha$ 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:


Scanning optical microscope

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<td>5.0</td>
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<tr>
<td>14.7</td>
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Simulation with LLG

\( H_i = 0 \text{ kA/m} \)
\( H_i = 5.2 \text{ kA/m} \)
Dinamical effects:
Precesional switching:
* Faster and less field. 

Experiment with ps field pulses perpendicular to the magnetisation (C. Back et al, Science, 1999)

Fe/GaAs

\[ \dot{\mathbf{M}} = [\mathbf{M} \times \mathbf{H}_\text{eff}] - \alpha [\mathbf{M} \times [\mathbf{M} \times \mathbf{H}_\text{eff}]] \]

- \( \mathbf{H} || \mathbf{M} \) —non-precessional switching
- Precessional switching is faster, however, the ringing phenomena occur.
Comparison of Patterns

Observed (SEMPA)

Calculated (fit using LLG)
- Anisotropies same as FMR
- Damping $\alpha = 0.017$
- 4x larger than FMR
- WHY?
  - Additional angular momentum dissipation? - spin current pumped across interface into paramagnet causes additional damping
  - SPIN ACCUMULATION

From Ch. Stamm - SLAC overview
Thermal effects
Thermal fluctuations play very important role in magnetisation dynamics:

At the microscopic level:

- At the equilibrium they are responsible for thermally excited spinwaves.
- 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 energy barriers 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”

\[
<i_i(t)>_0, \quad <h_i(t)h_j(t+\tau)> = \mu \delta(\tau), \quad i,j = x,y,z
\]

“The random-field components are formal concepts, introduced for convenience, to produce the fluctuations \( \delta M \)”

W.F. Brown outlined two methods:
- Based on the fluctuation-dissipation theorem
- Imposing the condition that the equilibrium solution of the Fokker-Planck equation is the Boltzman distribution

\[
\mu = \frac{2\alpha k_B T}{M_s V_i (1 + \alpha^2)}
\]

As a result of both in a non-interacting system:
Thermal micromagnetics
Langevin dynamics approach

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

\[
\vec{H} = \vec{H}_{\text{Zeeman}} + \vec{H}_{\text{aniso}} + \vec{H}_{\text{exch}} + \vec{H}_{\text{magnetost}} + \vec{H}_{\text{therm}}
\]

\[
< H_{\text{therm},i}(t) > = 0, \quad < H_{\text{therm},i}(t)H_{\text{therm},j}(t') >= \frac{2\alpha k_B T}{\gamma M_s V} \delta_{ij} \delta(t-t')
\]

- Initially introduced for nanoparticles
- This was brought to micromagnetics.

No correlations between time and different particles!!

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:

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

Diffusion coefficient (strength of fluctuations)

\[
P_{eq}(\vec{m}) \propto \exp\left[ -\frac{E(\vec{m})}{k_B T} \right]
\]

Boltzmann distribution in the equilibrium

\[
D = \frac{\alpha k_B T}{M_s V}
\]

The noise can be introduced either to precessional term or to both damping and precessional terms.
Problem of numerical scheme

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

\[ \vec{H} = \vec{H}_{\text{int}} + \vec{H}_{\text{thermal}} \]

- 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.
  - The Ito interpretation produces a stochastical drift.
  - Stratonovich interpretation should be used, for example the Heun numerical scheme*.
  - However, if after each integration step the magnetisation is renormalized – normal scheme could be used**.

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

J. Garcia-Palacios et al, Phys Rev B 58 (1998) 14937

\[ m_i = \sqrt{m_{i,x}^2 + m_{i,y}^2 + m_{i,z}^2} \]
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.
Langevin dynamics based on the Landau-Lifshitz-Gilbert equation.

could be formulated for both

• Atomistic spins (localized classical magnetic moments $\mu$ in the Heisenberg description with $J$ and on-site anisotropy $d$), $\alpha(\lambda)$ defines coupling to thermal bath

Characteristic timescale is determined by exchange;
(fs-ps)

• Micromagnetic units (averaged magnetisation, $M_s(T)$, $A(T)$, $K(T)$)

The temperature in this case is included twice:

 The damping $\alpha$ contains already thermal averaging: $\alpha(T)$
 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

\[
\bar{m}_x(\vec{r}, t) = \sum_{k, \omega} f(\omega, k) \exp \left[ i(\vec{k} \cdot \vec{r} - \omega_k t) \right]
\]

\[
\omega(\text{s}^{-1}) = \gamma \left( H + H_\perp + 2J/\mu_s \sin^2(ka/2) \right)
\]

<table>
<thead>
<tr>
<th>( f(\omega, k) )</th>
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<td>((0,0,5))</td>
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\[
\omega(T=0\text{K})
\]

\[
\omega(T=10\text{ K})
\]

\[
\omega(T=200\text{ K})
\]

\[
\omega(T=350\text{ K})
\]

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

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

Atomistic Heisenberg model gives correct $T_c$

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

\[
\frac{d\vec{M}_i}{d\tau} = -\vec{M}_i \times \vec{H}_i - \alpha \vec{M}_i \times (\vec{M}_i \times \vec{H}_i)
\]

\[
\tau = \gamma_0 t / (1 + \alpha^2), \quad \vec{H}_i = (-1 / M_s V_i)(\delta E / \delta \vec{M}_i)
\]

\[
\vec{H} = \vec{H}_{\text{int}} + \vec{h}_{\text{therm}}
\]

\[
\langle h_i \rangle = 0, \quad \langle h_i(t) h_j(t+\tau) \rangle = \frac{2\alpha k_B T}{M_s V_i (1 + \alpha^2)} \delta(\tau) \delta_{ij}
\]

Langevin dynamics for the micromagnetics does not correctly describe spinwaves:
- 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.

Atomistic modelling of magnetisation reversal

Field applied at 30°

64³ magnetic moments on cubic lattice

- Magnetisation magnitude is not conserved
- Damping is enhanced at high T

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

Field applied at 135°

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

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

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

\[ H_{\text{eff}} = H - \frac{m_x e_x + m_y e_y}{\tilde{\chi}_\perp} + \begin{cases} \frac{1}{2\tilde{\chi}_\parallel} \left( 1 - \frac{m^2}{m^2_e} \right) m, & T \lesssim T_c \\ \frac{J_0}{\mu_s} \left( \epsilon - \frac{3}{5}m^2 \right) m, & T \gtrsim T_c \end{cases} \]

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

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.

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

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

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

COMPARISON BETWEEN ATOMISTIC AND ONE-SPIN LLB SIMULATIONS

atomistic

one-spin LLB

\[ M(t=0) = \text{atomistic} \]

\[ M(t=0) = \text{one-spin LLB} \]
Multiscale approach
Multiscale modelling: all the parameters were evaluated from atomistic modelling for FePt with ab-initio input parameters (Tc= 650K)

Longitudinal relaxation

Transverse relaxation

solid line - one spin LLB
CONCLUSIONS

- 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.