THE UNIVERSITY of York

Computational Magnetism Group

# Multiscale approaches to sub-picosecond heat driven magnetisation reversal

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# Summary

- Lecture 1:
  - The timescale and lengthscale problem
  - Stocastic single spin models
  - Multispin models; micromagnetics and atomistic theories
  - Spin excitations in ferromagnets and antiferromagnets

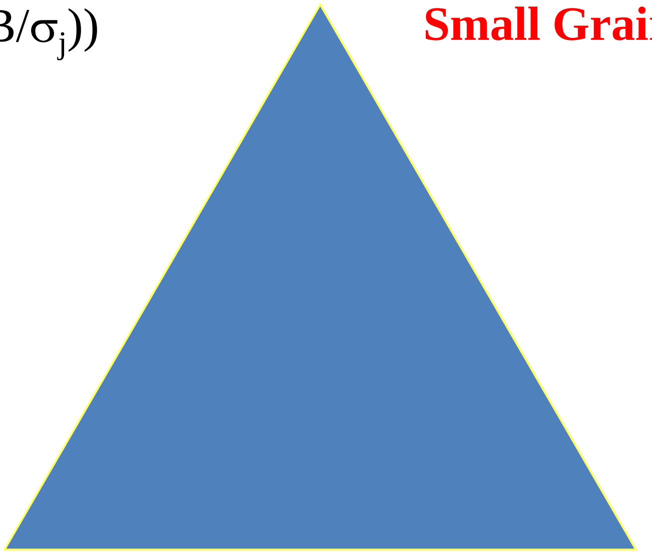
- Lecture 2:
  - Introduction to pulsed laser processes
  - New (linear) magnetisation reversal mechanism
  - Linear reversal is calculated to give reversal times as fast as 300fs !
  - Dynamics and the Landau-Lifshitz- Bloch (LLB) equation of motion
  - LLB-micromagnetics and dynamic properties for large-scale simulations at elevated temperatures
  - Heat Assisted Magnetic Recording (HAMR); experiments and LLB-micromagnetic model
  - Opto-magnetic reversal – the ultimate speed record?

# Media Design Constraints - "Trilemma"

**Media SNR**

SNR~10×log$_{10}$(B/$\sigma_j$))

**Small Grains (V)**



**Thermal Stability**

$$E_B \cong K_u V \cdot \left[1 - \frac{|H_d|}{H_0}\right]^{3/2}$$

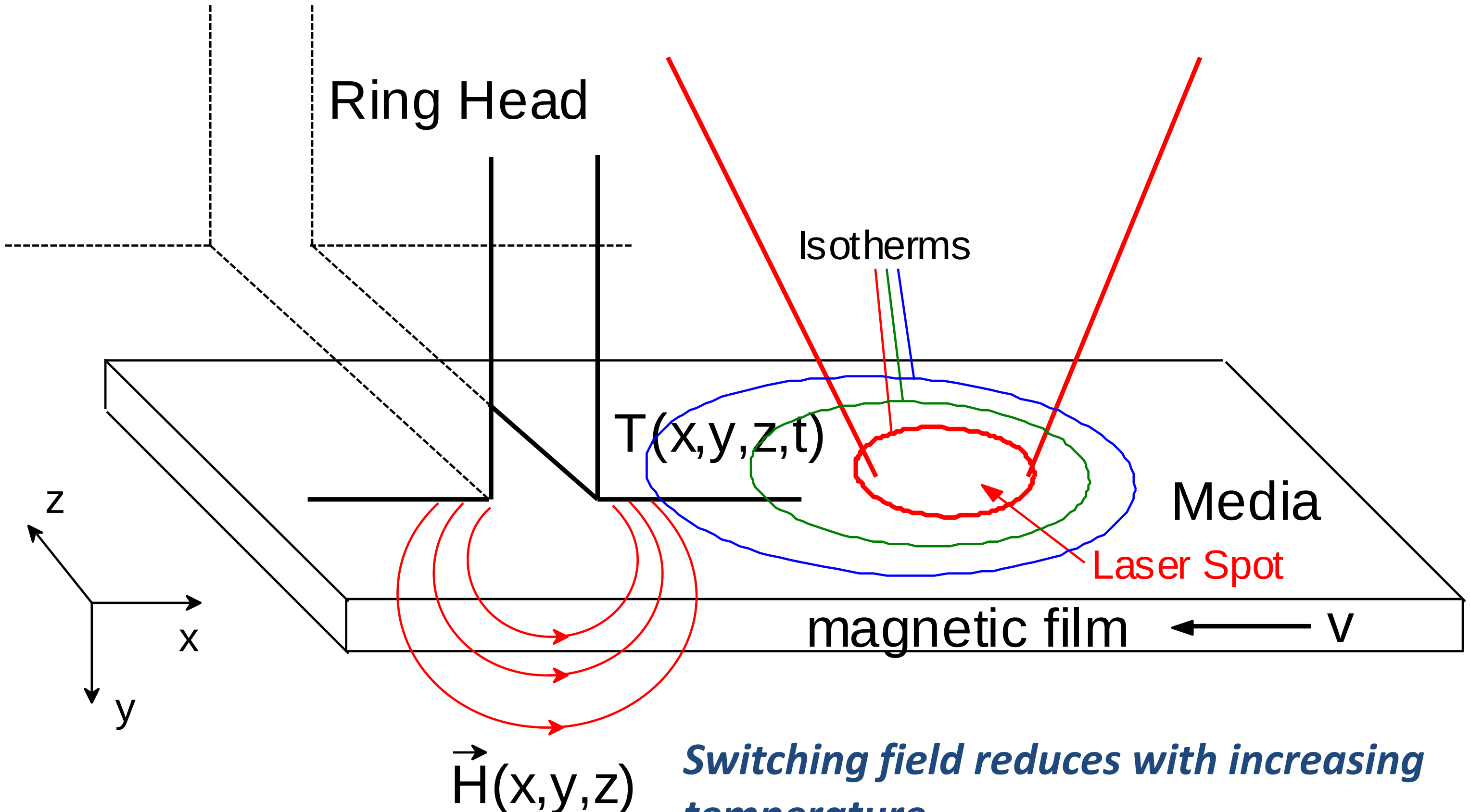
**$K_u V = r_k(T,t,\sigma, H_d) \times k_B T$**

**Writability**

$$H_0 = \alpha \cdot \frac{2 \cdot K_u}{M_S} - N_{eff} \cdot M_S$$

**$H_0$ < Head Field**

# Hybrid Recording Using Light




***Switching field reduces with increasing temperature***

M. Kryder , et al. TMRC 2002

# The need for atomistic/multiscale approaches

- Micromagnetics is based on a continuum formalism which calculates the magnetostatic field exactly but which is forced to introduce an approximation to the exchange valid only for long-wavelength magnetisation fluctuations.
- Thermal effects can be introduced, but the limitation of long-wavelength fluctuations means that micromagnetics cannot reproduce phase transitions.
- The atomistic approach developed here is based on the construction of a physically reasonable classical spin Hamiltonian based on ab-initio information.

# Atomistic model

- Uses the Heisenberg form of exchange

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

- Spin magnitudes and J values can be obtained from ab-initio calculations.
- We also have to deal with the magnetostatic term.
- 3 lengthscales – electronic, atomic and micromagnetic – Multiscale modelling.

# Model outline

**Ab-initio information (spin, exchange, etc)**

↓

**Classical spin Hamiltonian** → **Dynamic response solved using Langevin Dynamics (LLG + random thermal field term)**

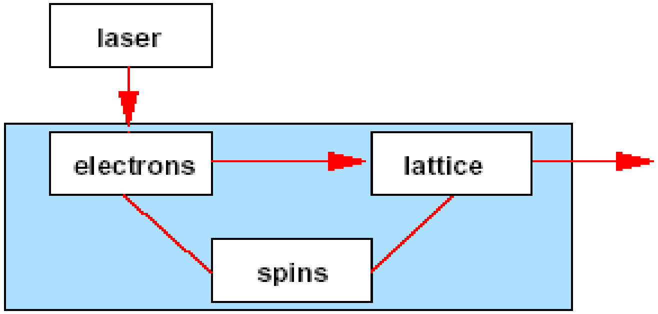
↑

**Magnetostatics**

# Laser Pump-probe experiments

- High energy laser beam (pump) causes rapid heating of a magnetic film
- Part of the beam is split off and used to measure the magnetisation of the film using the Magneto-Optic Kerr Effect (MOKE)
- Magnetisation changes on the sub-picosecond timescale can be demonstrated experimentally
- Very important physics
- Also, potentially important because of the possible use of Heat Assisted Magnetic Recording (HAMR)

# 2 temperature model




**1.** photon energy is transferred to electrons

**2.** energy is exchanged between electrons and phonons

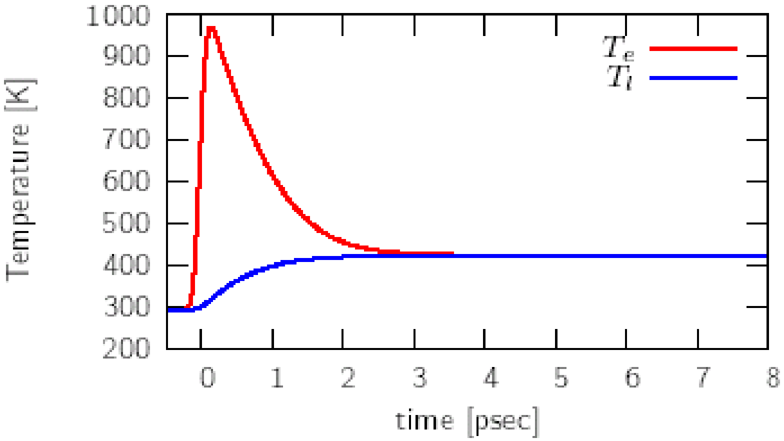
**3.** energy dissipates into environment

**Two temperature model:**

$$\text{electrons:}\quad C_e \frac{\mathrm{d}T_e}{\mathrm{d}t} = -G_{el}(T_e - T_l) + P(t)$$

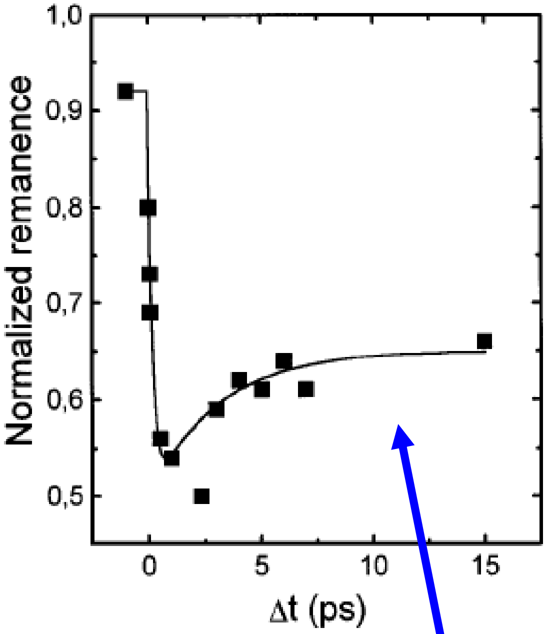
$$\text{lattice:}\quad C_l \frac{\mathrm{d}T_l}{\mathrm{d}t} = G_{el}(T_e - T_l)$$

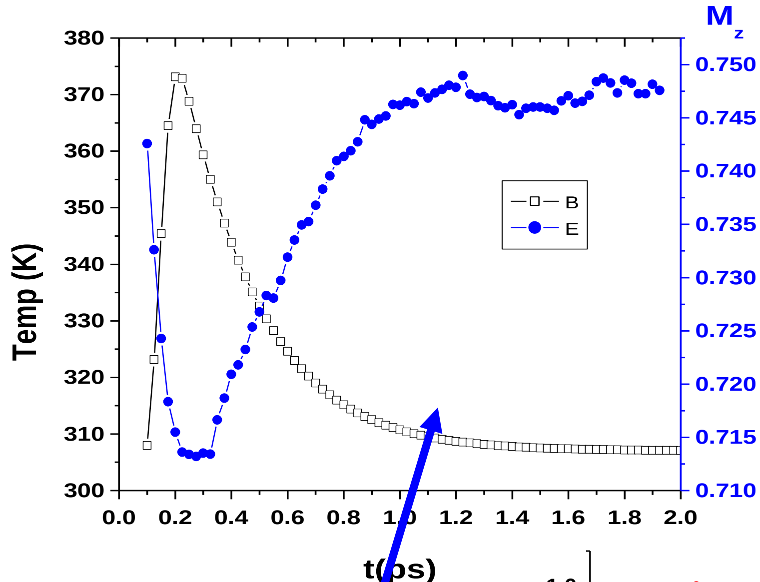
(M. I. Kaganov et al., Sov. Phys. JETP **4**, 173 (1957))

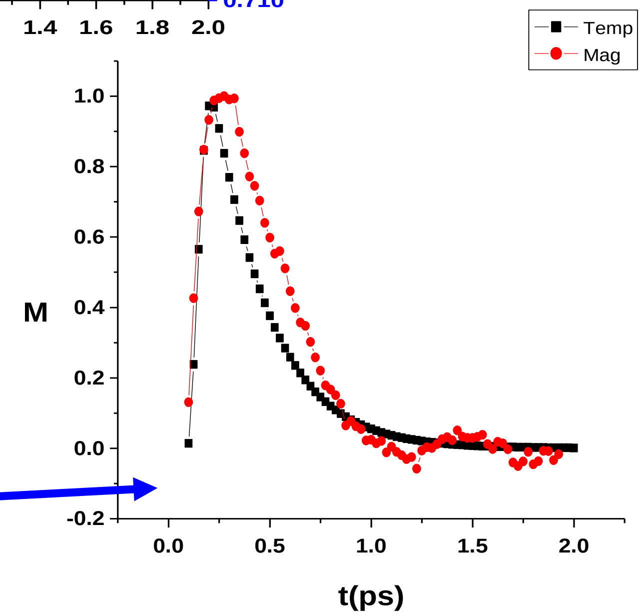



$\Rightarrow$ perform Langevin dynamics simulation with $T_e$ as temperature of the heat bath

# Ultrafast demagnetisation




- Experiments on Ni (Beaurepaire et al PRL 76 4250 (1996)
- Calculations for peak temperature of 375K
- Normalised M and T. During demagnetisation M essentially follows T

# Excitation modes during ultrafast heating

- Approach:
  - Spatial and temporal Fourier transforms
  - Gives the dispersion relation
  - Can also calculate a 'mode occupancy' from the power/mode normalised by the total power
- We have studied the excitations in both ferromagnets and antiferromagnets

# Analytical dispersion relations

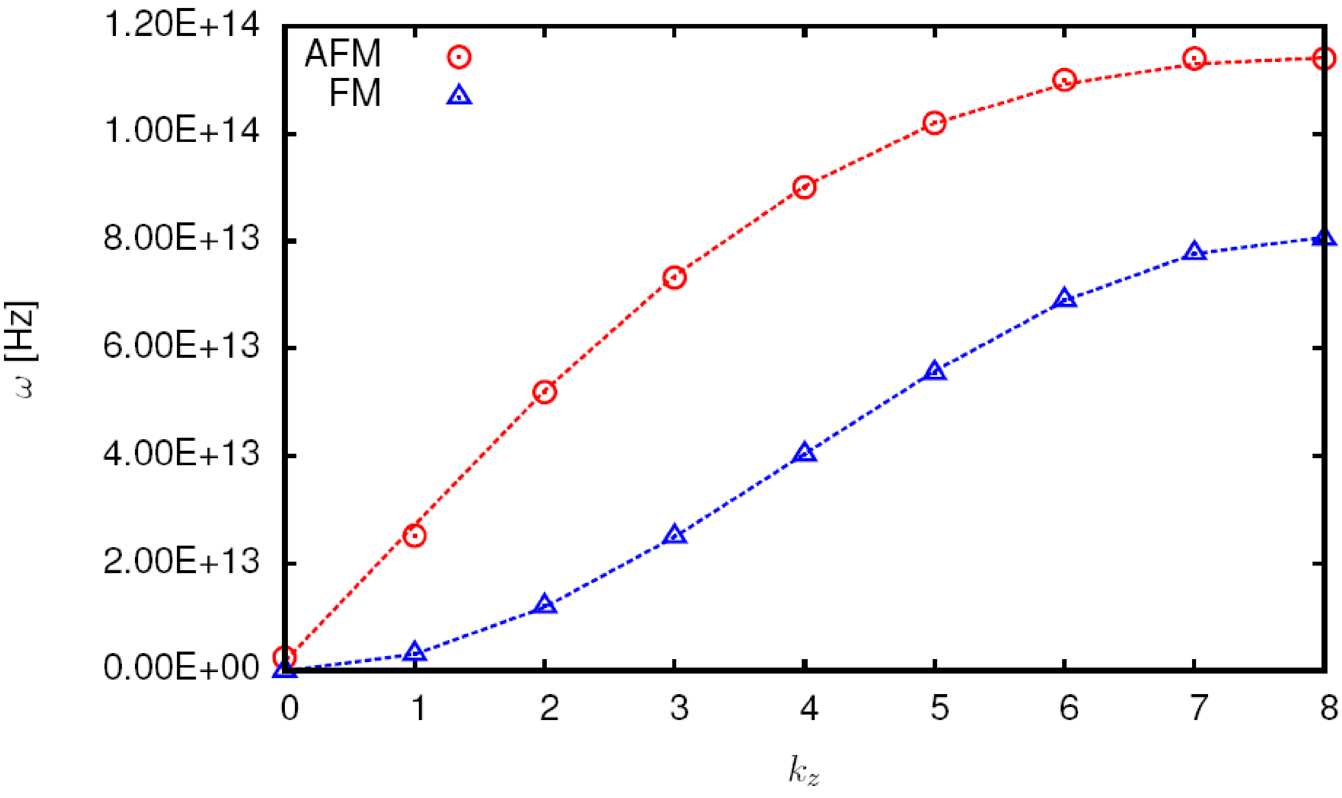
Ferromagnet

$$\omega(k_z) = \gamma \left[B_{ani} + B_{app} + 2SJ(1 - \cos k_z a)\right]$$

Antiferromagnet

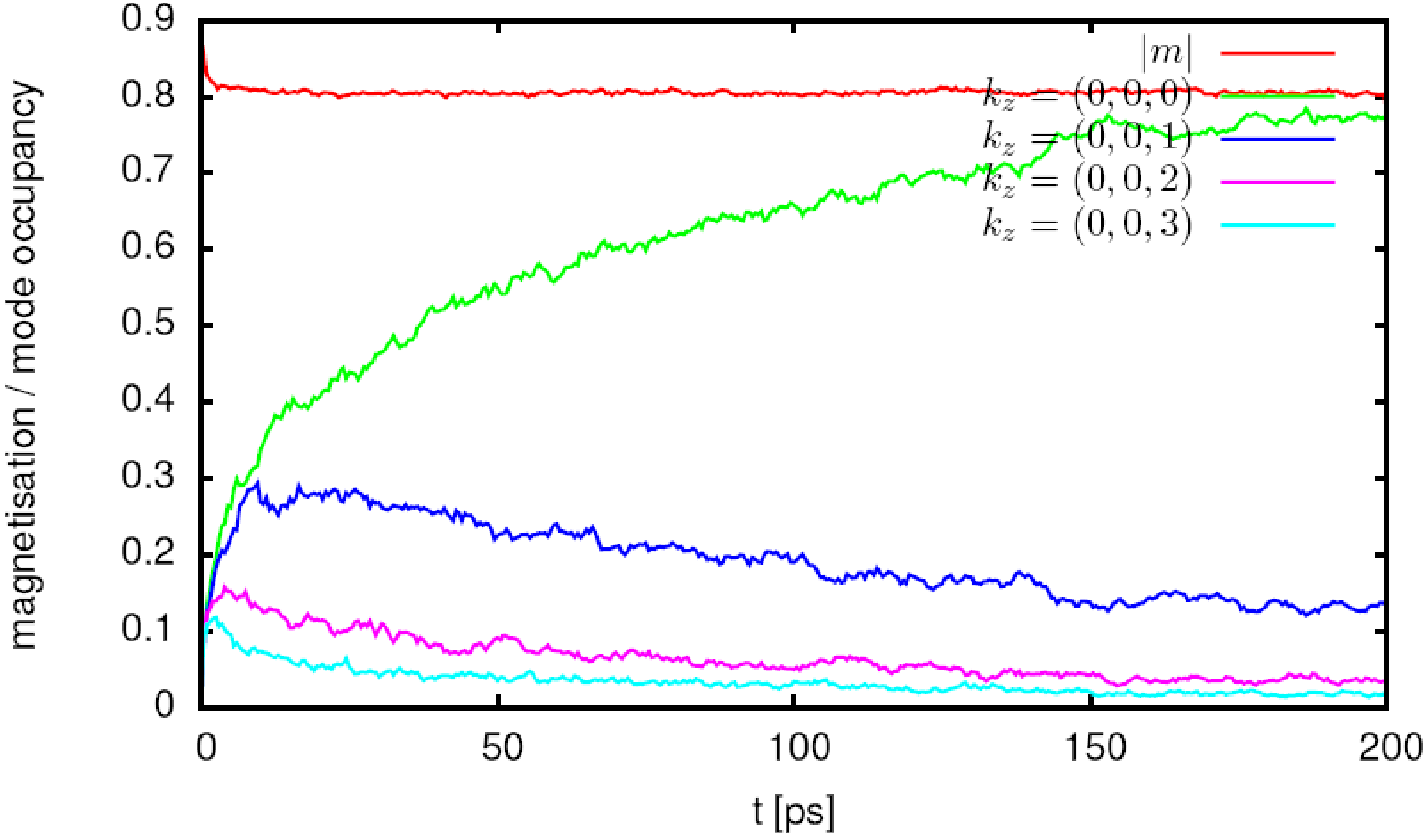
$$\omega(k_z) = \gamma \left[B_{app} \pm \sqrt{(B_{ani} + 6SJ)^2 - (2SJ(1 \cos k_z a + 2))^2}\right]$$

# Analytical vs numerical

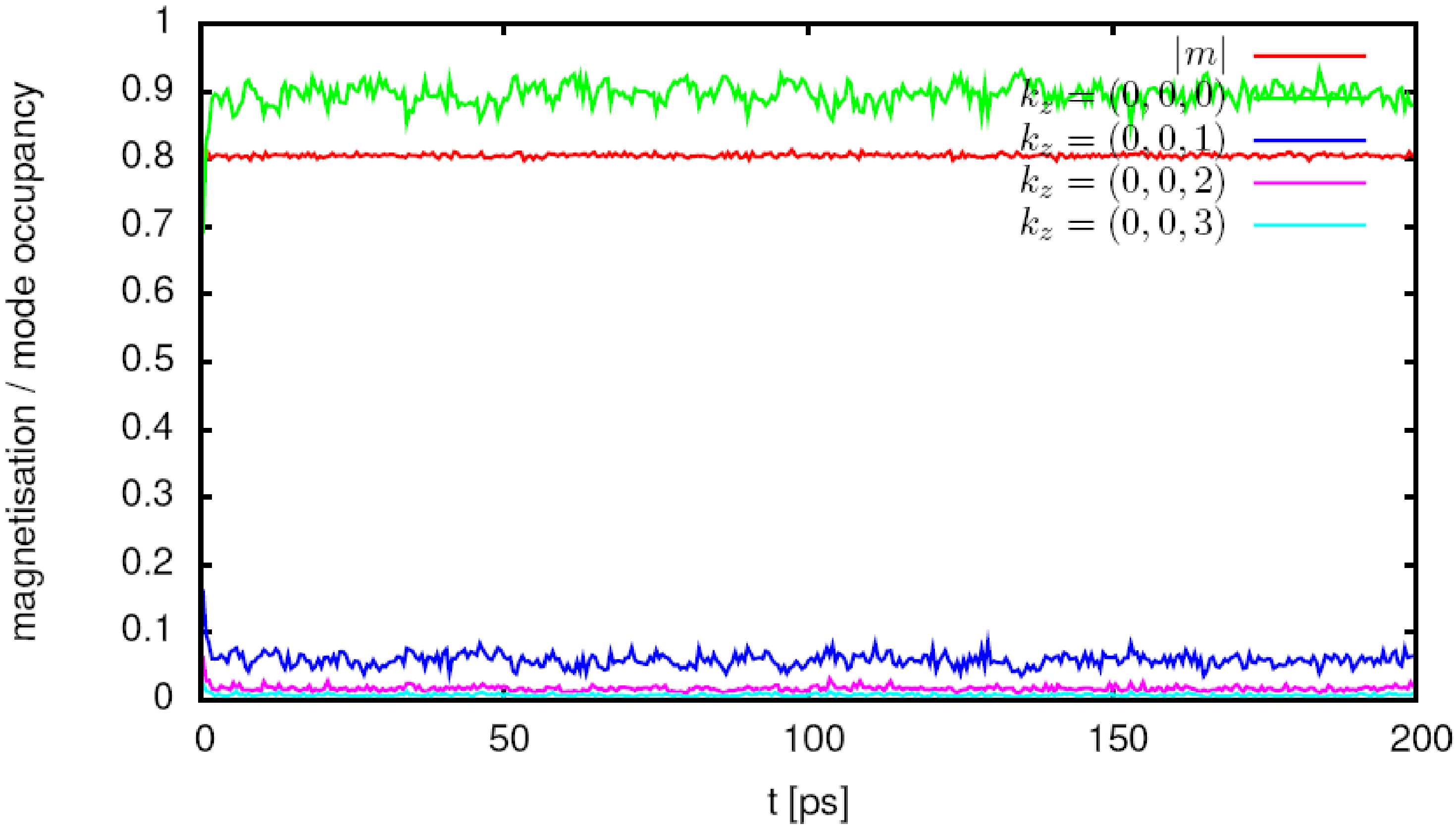



- Solid lines; analytical, symbols; numerical

# Response to step temperature change: ferromagnet

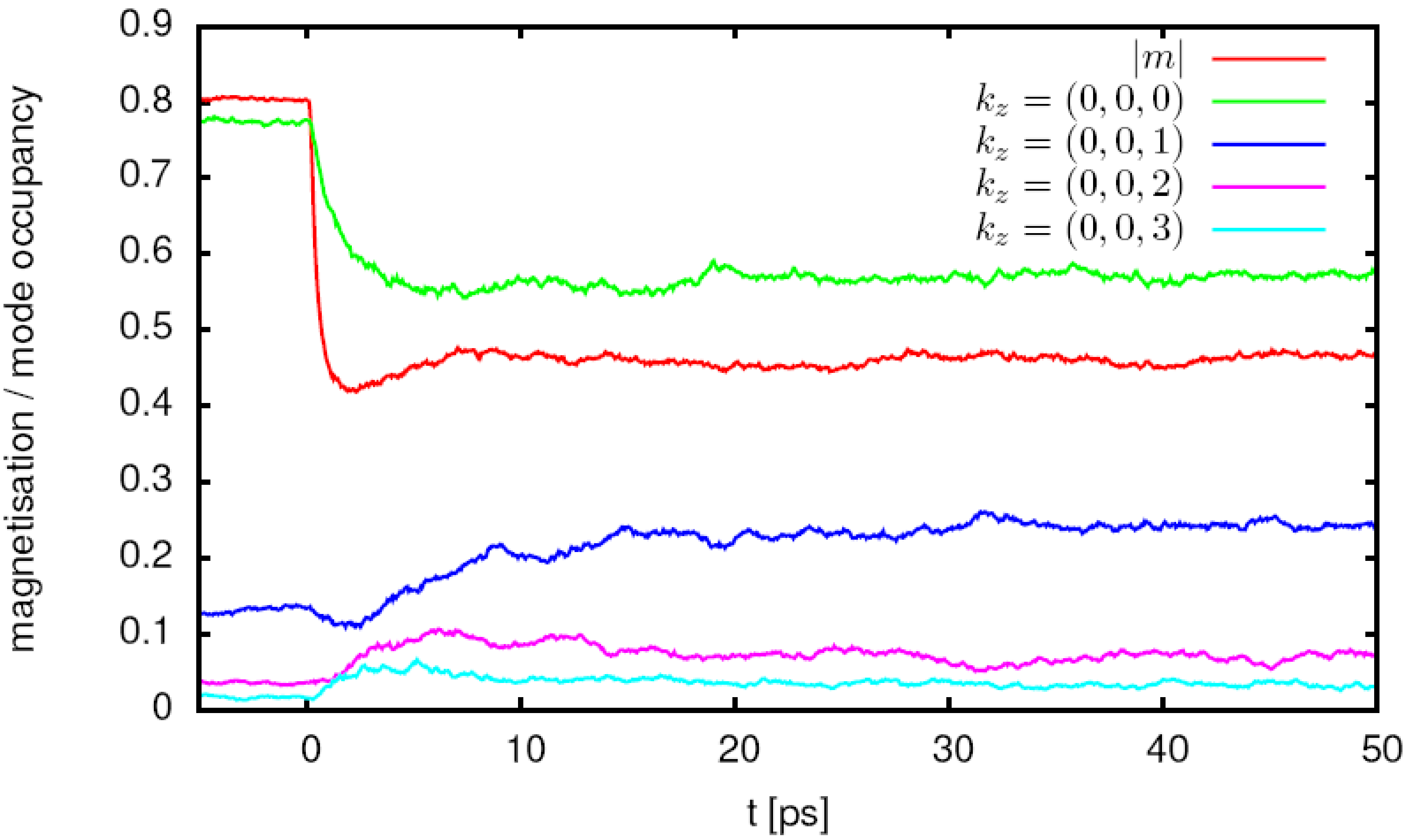


# Antiferromagnet

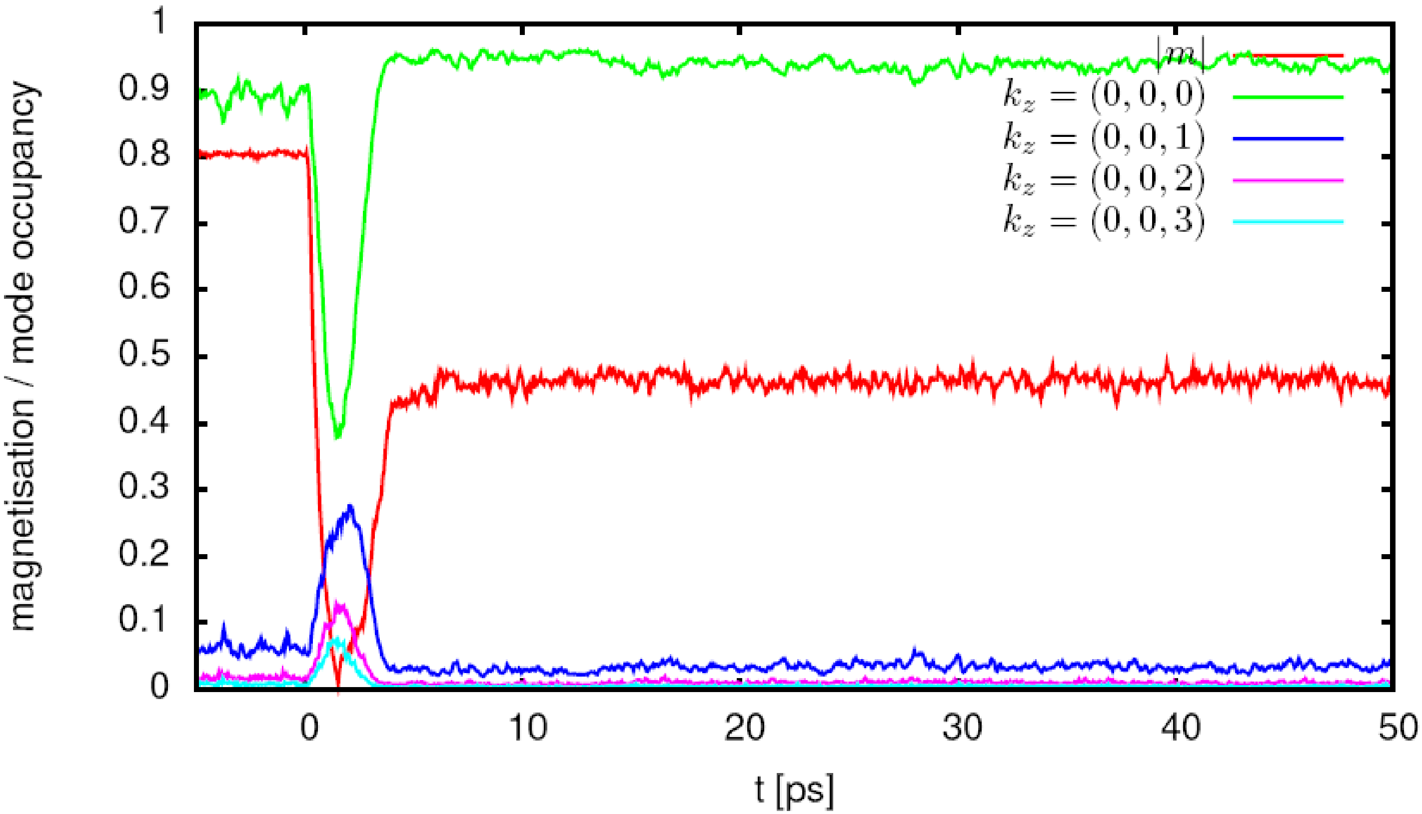



- (Staggered) magnetisation equilibrates more rapidly than FM

# Response to pulse temperature change: ferromagnet

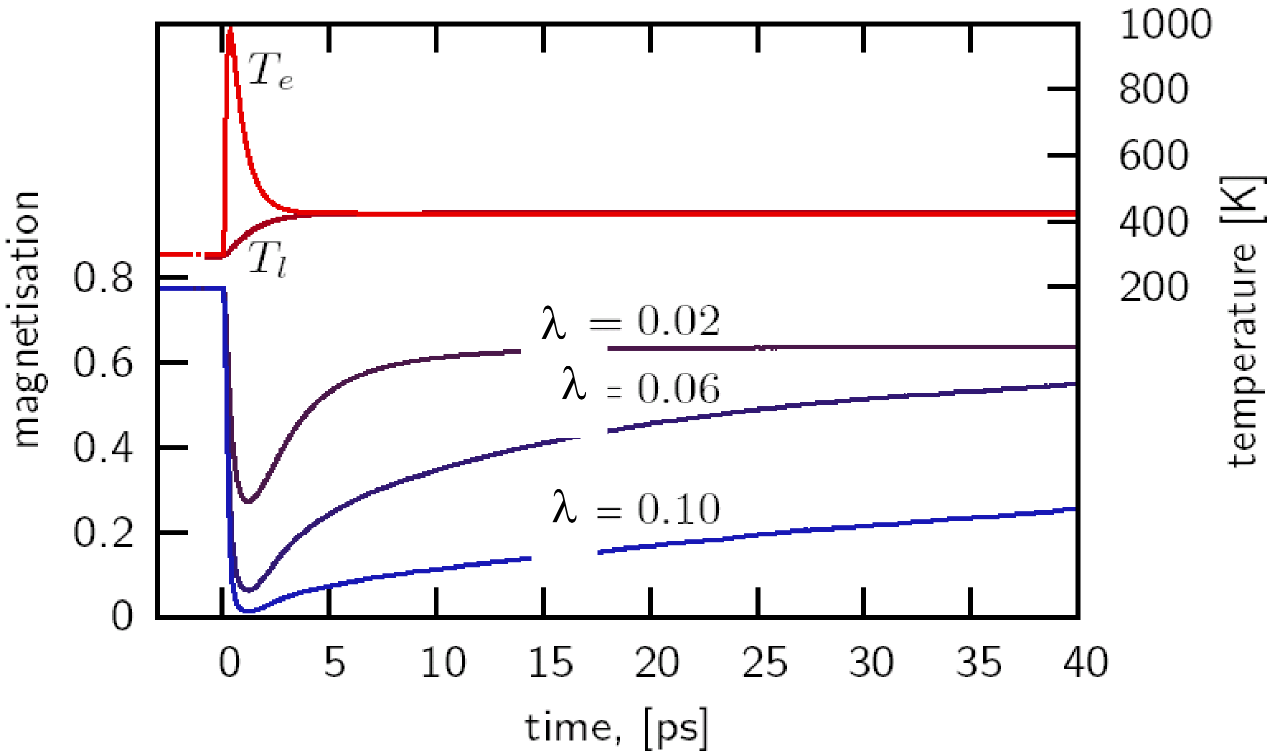


# Antiferromagnet



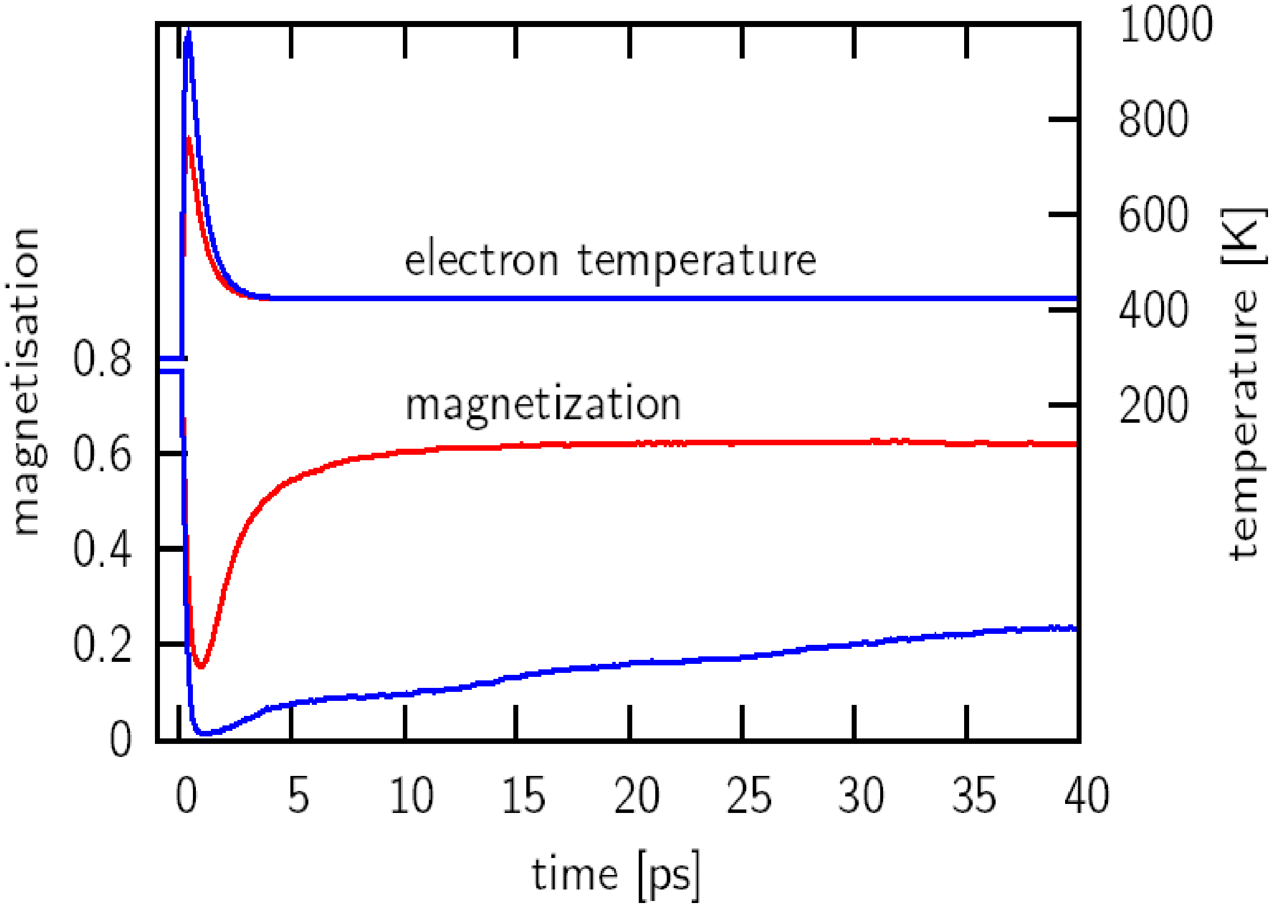

- Much faster response
- Consistent with faster demagnetisation of the AF

# Pump-probe simulations – continuous thin film



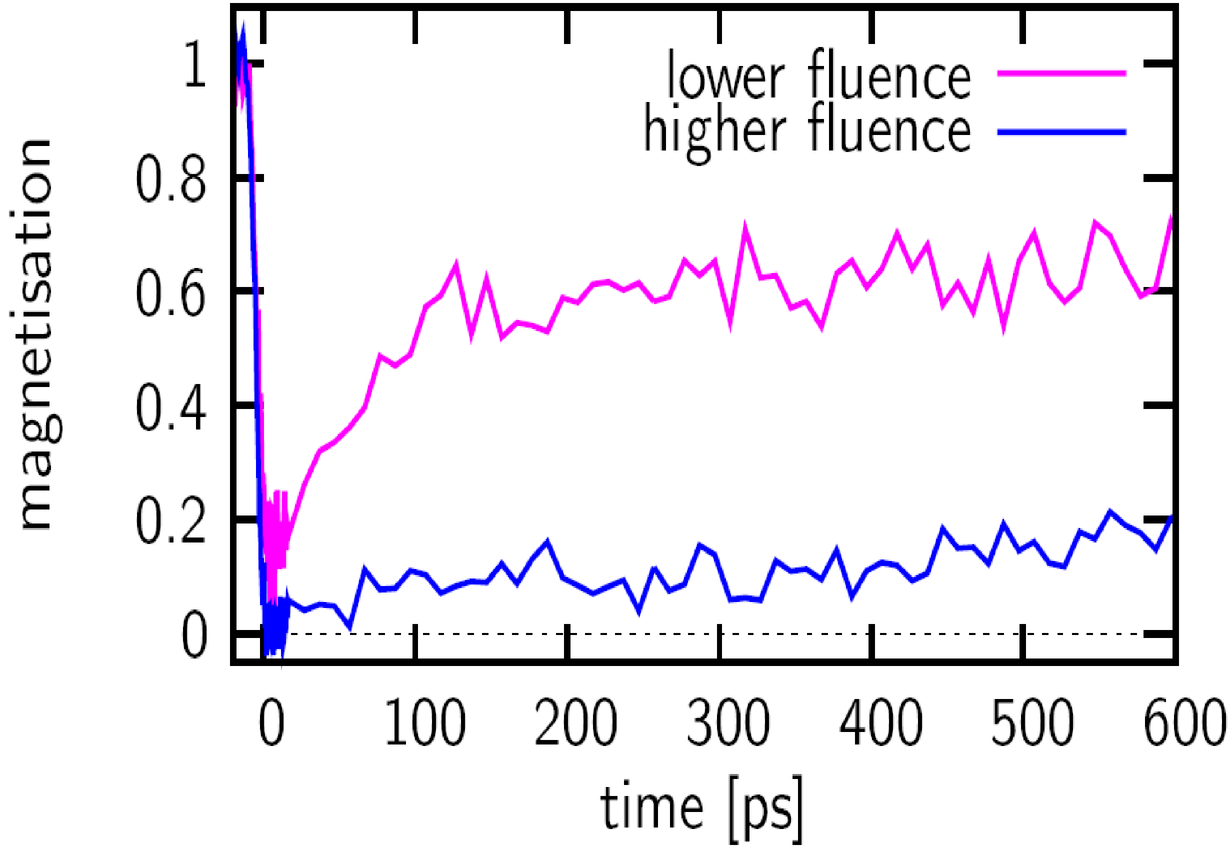

- Rapid disappearance of the magnetisation
- Reduction depends on l (coupling constant)

# Dependence on the pump fluence

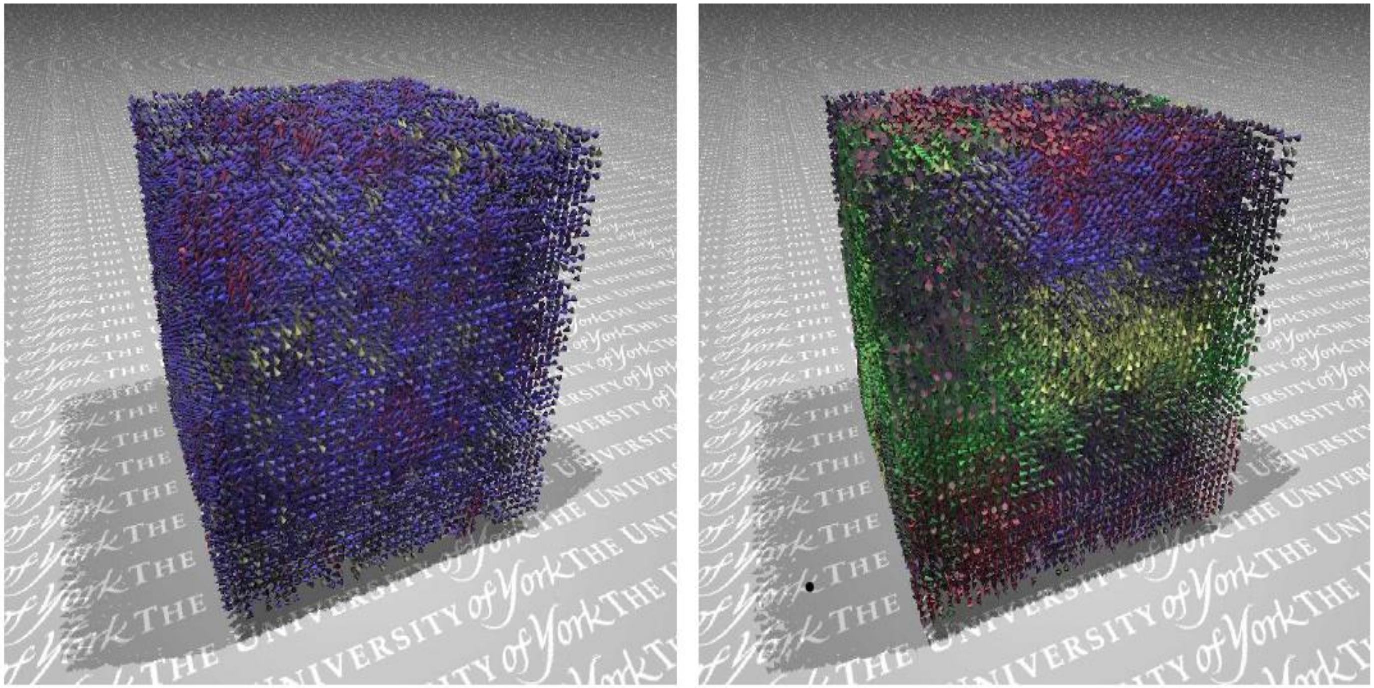



- Note the slow recovery of the magnetisation for the higher pump fluence

# Experiment (J. Hohlfeld)



# Slow recovery due to disordered magnetic state



- Snapshots of the magnetisation distribution after 19ps for l = 0:02 (left) and l = 0:2 (right).
- Fast recovery if there is some ‘memory’ of the initial magnetic state.
- For the fully demagnetised state the recovery is frustrated by many nuclei having random magnetisation directions.

# Comment on the use of the LLG equation and Langevin Dynamics

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

**Spin system** ⇔ **Conduction electrons** ⇔ **Lattice** ⇔ **Heat bath**

- What is α? Transfer of energy via complicated channels
- Very challenging. But interesting physics and important applications
- Are thermal fluctuations really uncorrelated? Next we look at the effects of correlated noise

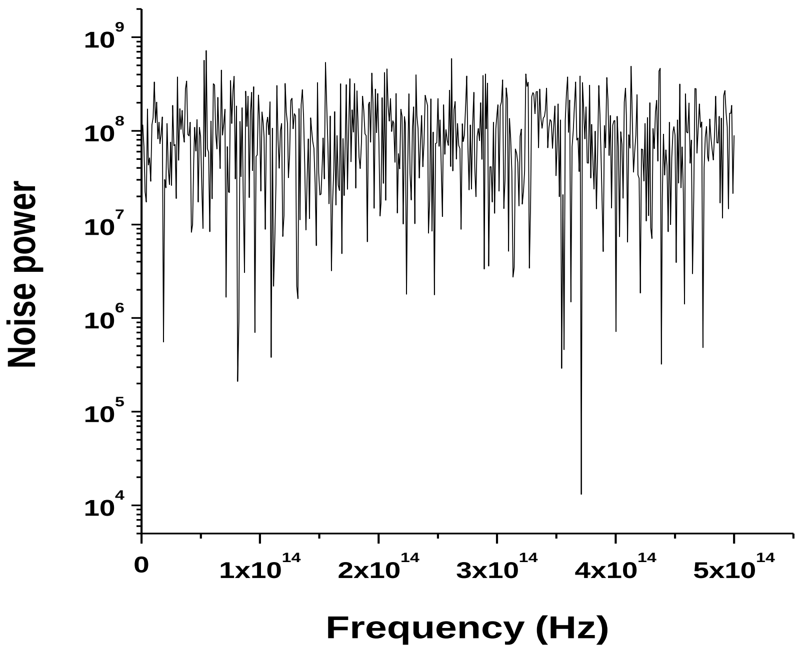
# Coloured noise

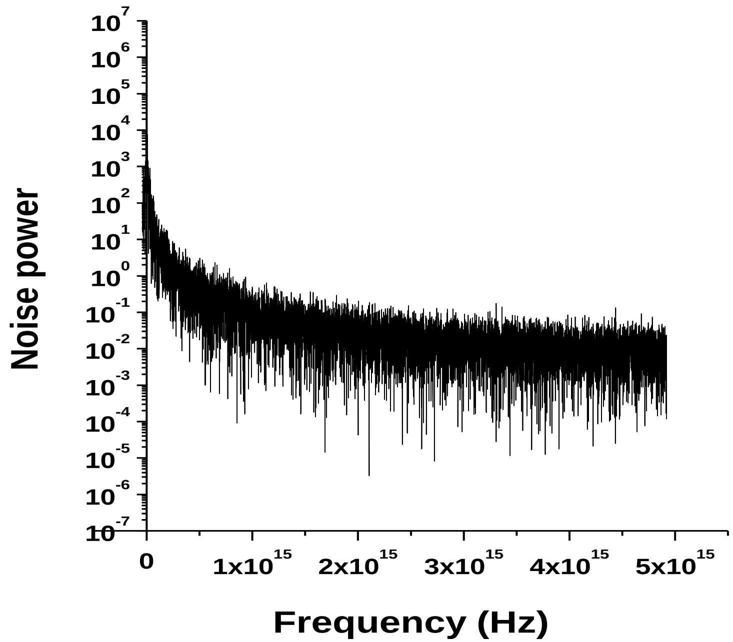
- Simulations so far used white noise
- Assumes uncorrelated noise source
- Here we introduce exponentially correlated noise and investigate the effect on the relaxation time of the magnetisation.

# Exponentially correlated (Ornstein-Uhlenbeck) noise

$$\langle \varepsilon(t) \rangle = 0 \qquad \text{and} \qquad \langle \varepsilon(t)\varepsilon(t') \rangle = \frac{D}{\tau} \exp\left( -\frac{|t-t'|}{\tau} \right)$$

- Gaussian noise with zero mean and exponential correlation function.
- Correlation time $\tau$
- Variance $\sigma^2 = \langle \varepsilon^2 \rangle = D/\tau$




Correlations shift the noise to low frequencies (right)

# Basis of the simulation – Miyazaki- Seki equation
# Atxitia et al Phys. Rev. Lett. 102, 057203 (2009)

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times (\mathbf{H}_0 + \mathbf{H}_{th})$$

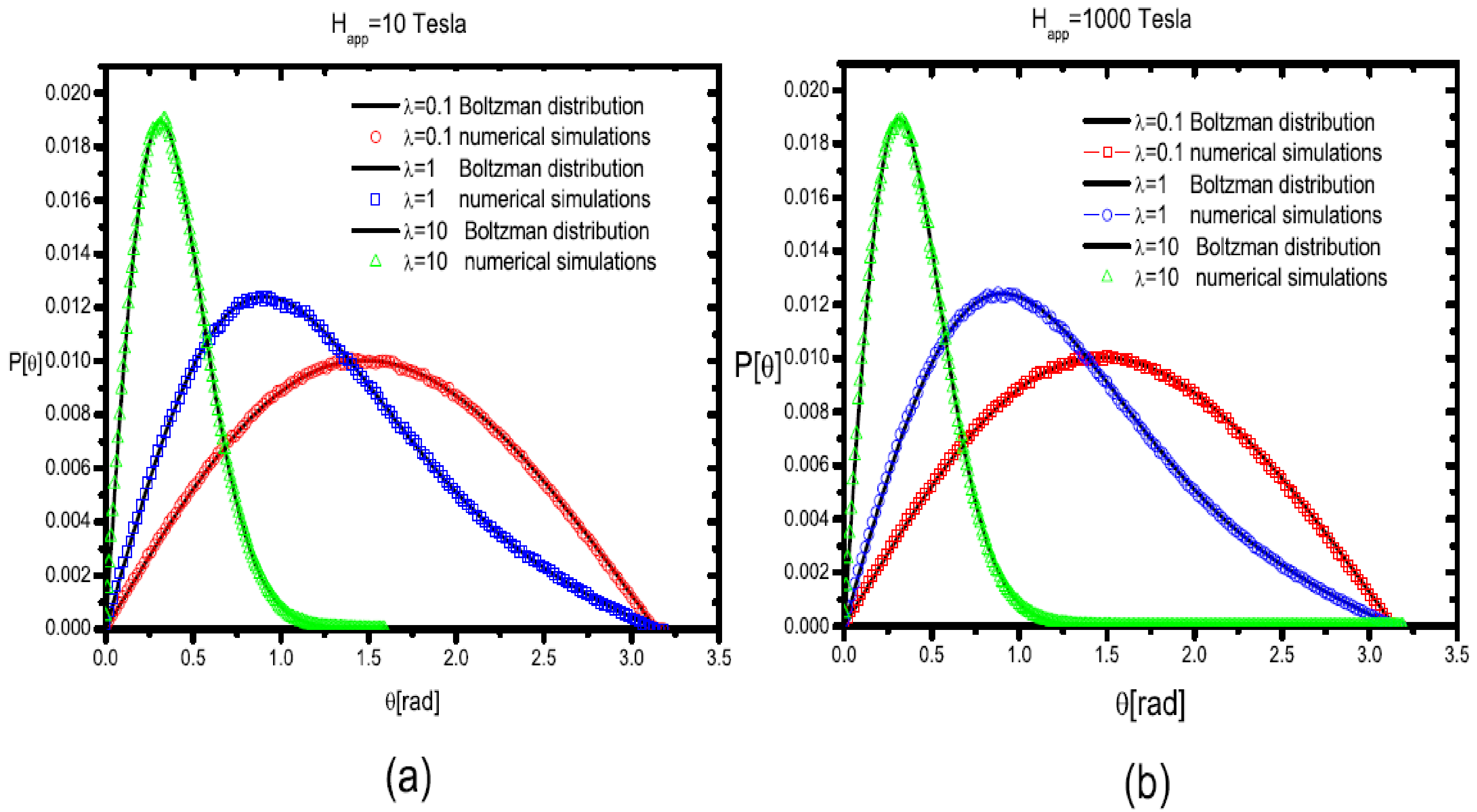
$$\frac{d\mathbf{H}_{th}}{dt} = -\frac{1}{\tau_c}(\mathbf{H}_{th} - \chi \mathbf{M}) + \mathbf{R}$$

$$\langle R_i(t) R_j(t') \rangle = \frac{2}{\tau_c} \chi k_B T \delta_{ij} \delta(t - t')$$

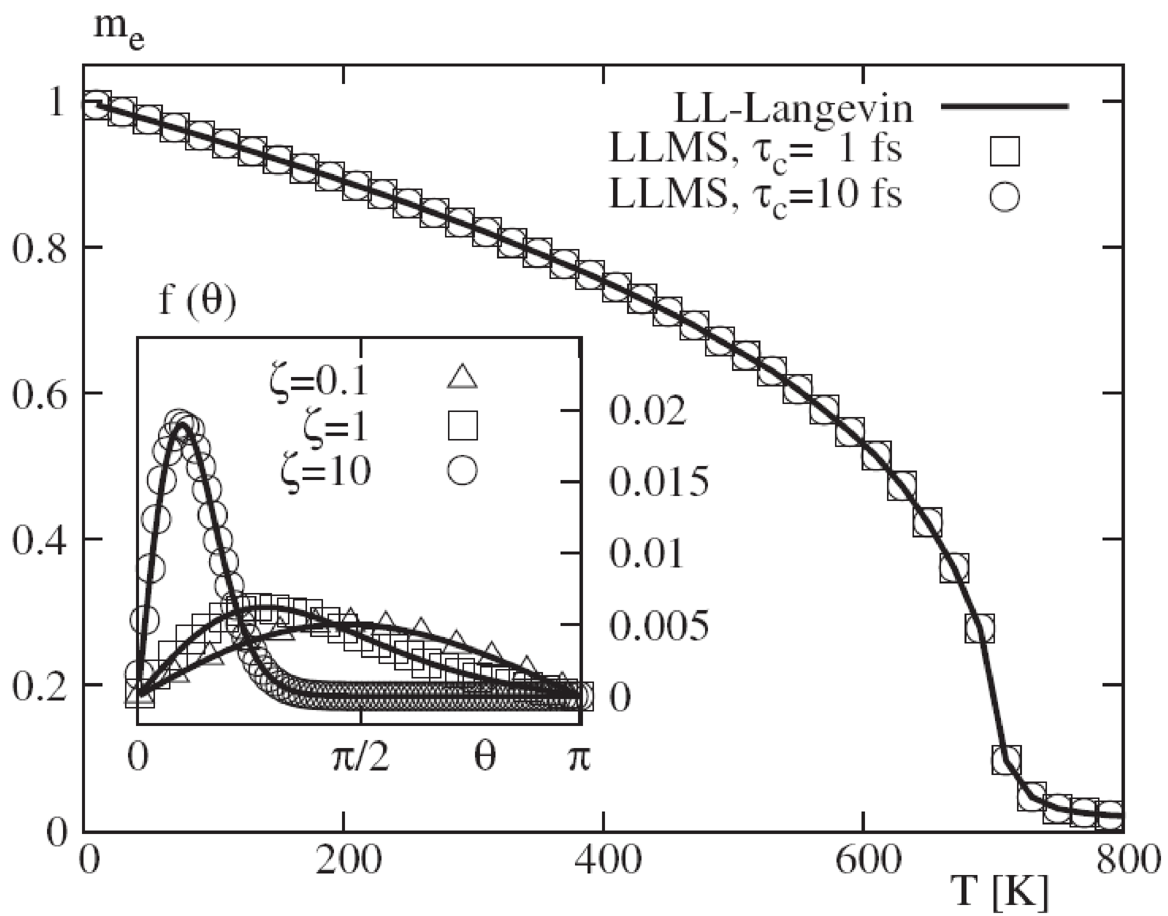
$\chi=\alpha/(2\gamma\tau_{\chi}\mu)$

NB (very crude) direct simulation of the heat bath properties.

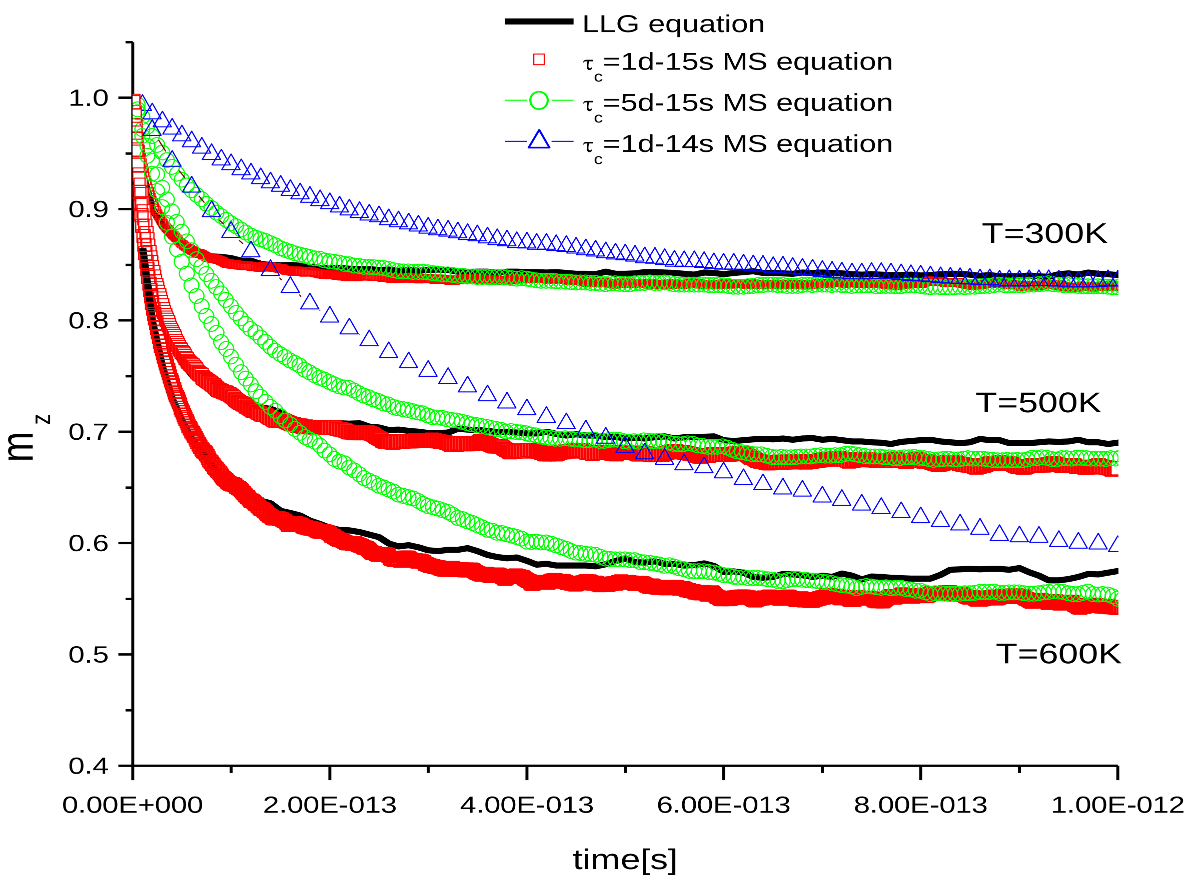
# Single-spin calculations – probability distributions



# Multispin calculations – equilibrium properties



# Correlations increase the longitudinal relaxation time



# Multiscale calculations and the LLB equation

- Large scale (micromagnetic) simulations essentially work with one spin/computational cell
- Single spin LLG equation cannot reproduce ultrafast reversal mechanisms at elevated temperature (conserves |M|)
- Pump- probe and HAMR simulations require an alternative approach
- Landau-Lifshitz-Bloch (LLB) equation?

# LLB equation

Transverse (LLG) term

Longitudinal term introduces fluctuations of M

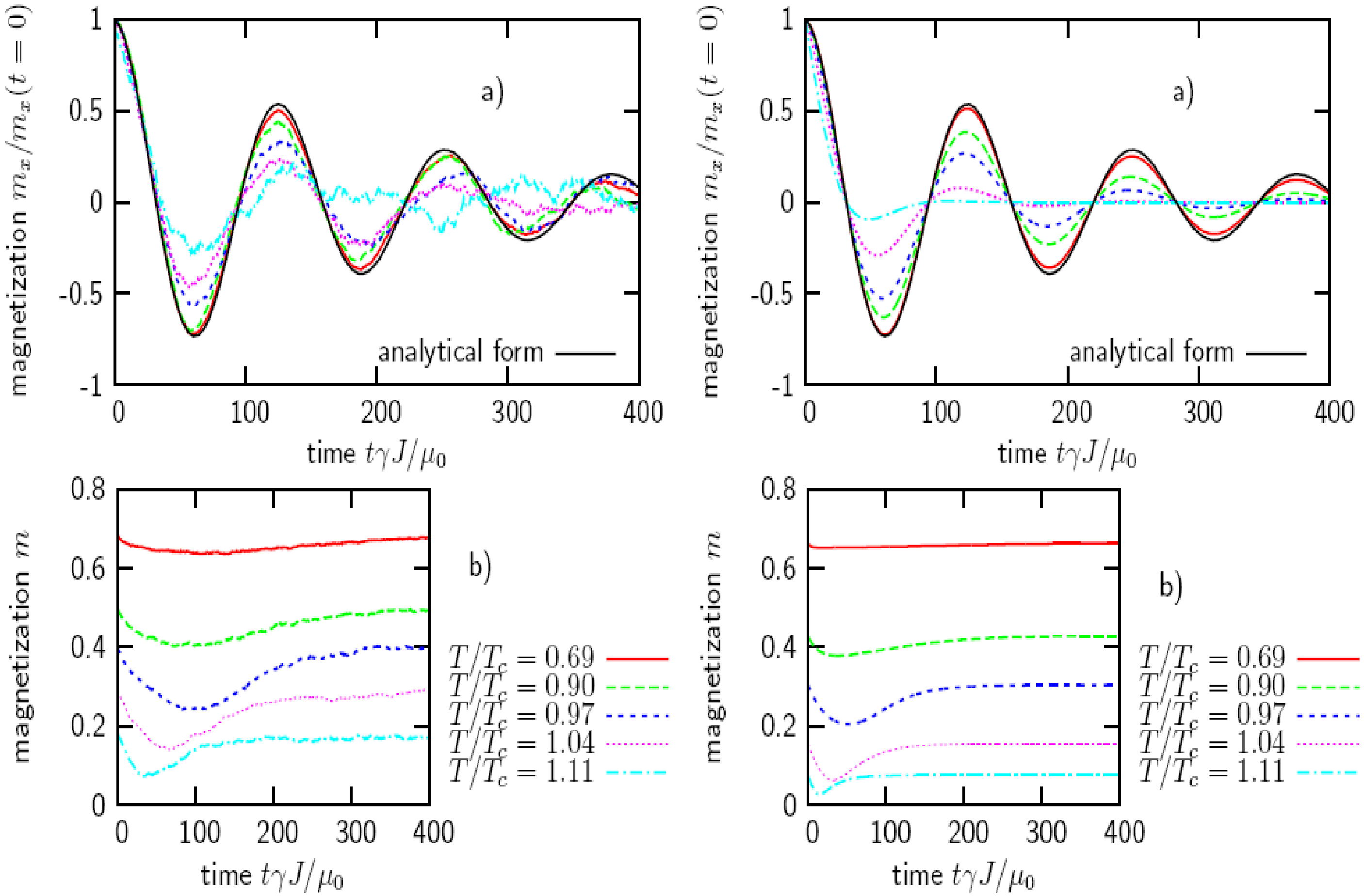
$$\dot{\mathbf{m}} = -\gamma[\mathbf{m}\times\mathbf{H}_{\text{eff}}] + \gamma\alpha_{\parallel}\frac{(\mathbf{m}\cdot\mathbf{H}_{\text{eff}})\mathbf{m}}{m^2} - \gamma\alpha_{\perp}\frac{[\mathbf{m}\times[\mathbf{m}\times\mathbf{H}_{\text{eff}}]]}{m^2}$$

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

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

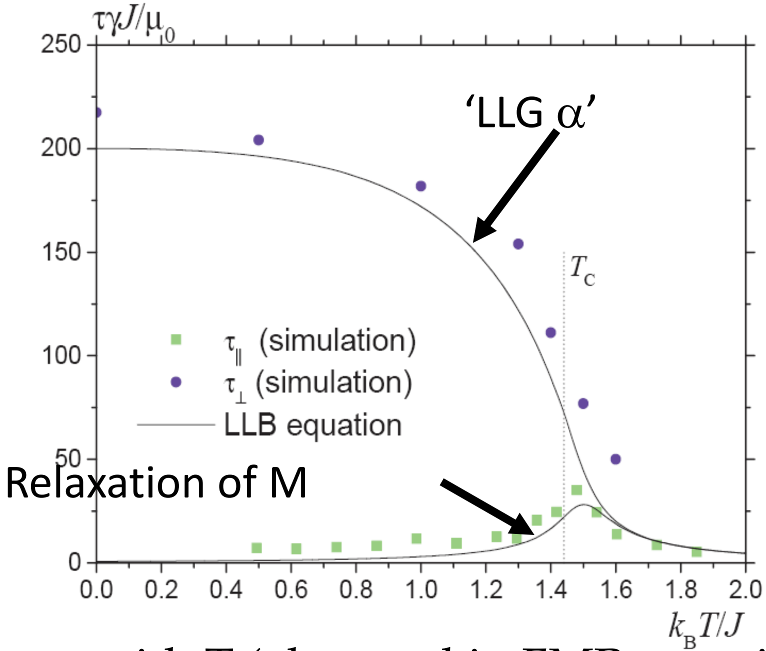
here $\mathbf{H}$ is applied field and $m_e$ is zero-field equilibrium spin polarization

the second term is an expression for the anisotropy field




- Precessional dynamics for atomistic model (left) and (single spin) LLB equation (right)

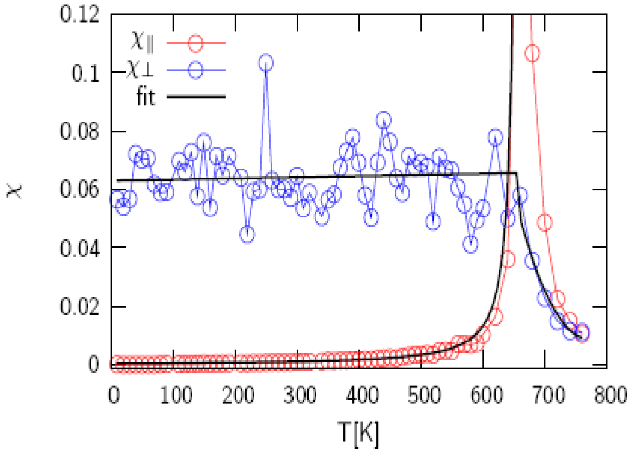
# Relaxation times




- Effective a increases with T (observed in FMR experiments)
- Critical slowing down at $T_c$
- Longitudinal relaxation is in the ps regime except very close to $T_c$
- Atomistic calculations remarkably well reproduced by the LLB equation
- Makes LLB equation a good candidate to replace LLG equation in micromagnetics.

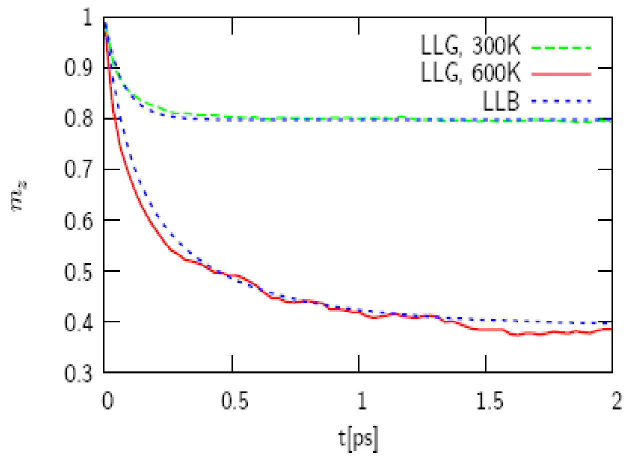
# LLB parameters

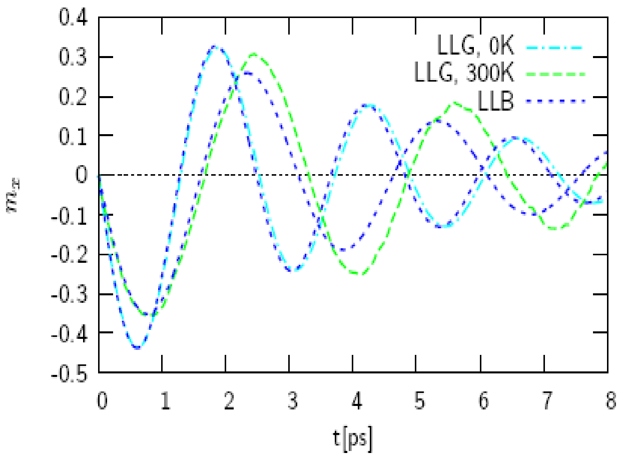
- Important parameters are;
  - Longitudinal and transverse susceptibility
  - K(T), M(T)
- These can be determined from Mean Field theory.
- Also possible to determine the parameters numerically by comparison with the Atomistic model.
- In the following we use numerically determined parameters in the LLB equation and compare the dynamics behaviour with calculations from the atomistic model.



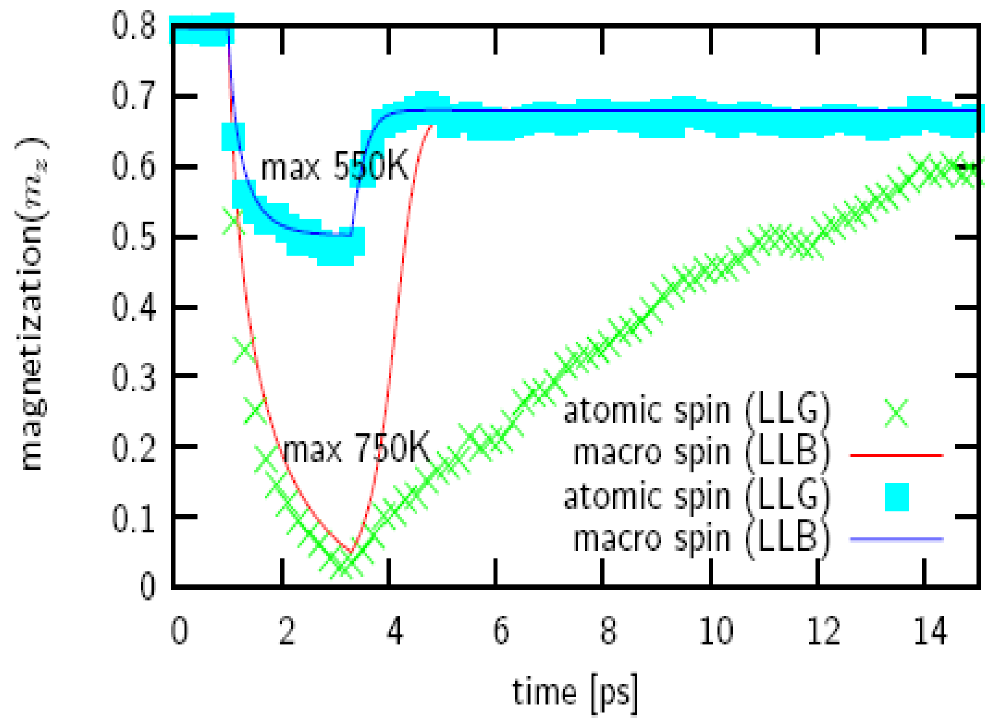

- spin model for FePt [4]
- LLG calculations for a grain of size $32 \times 32 \times 48$ spins
- $\tilde{\chi}_\perp$ (left fig.), $\tilde{\chi}_\parallel$ and $m_e$ were evaluated and used for LLB calculations

longitudinal relaxation (left) and transverse relaxation after $30^\circ$ exitation (right) for atomistic LLG and macro-spin LLB modeling



# Comparison with (macrospin) LLB equation




- Single LLB spin cannot reproduce  the slow recovery with a single longitudinal relaxation time.
- State dependent relaxation time?
- Big advantage in terms of computational efficiency.
- LLB equation is an excellent candidate approach to complete the multiscale formalism

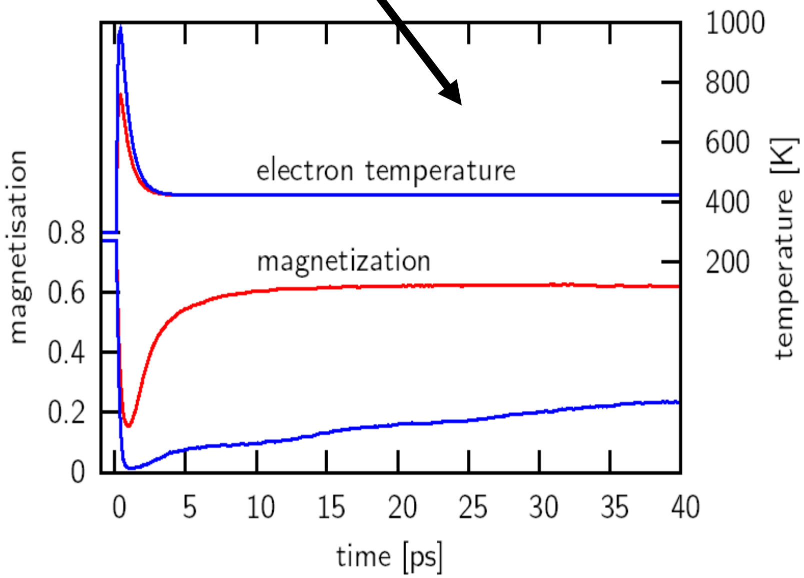
# Slow recovery – multispin LLB

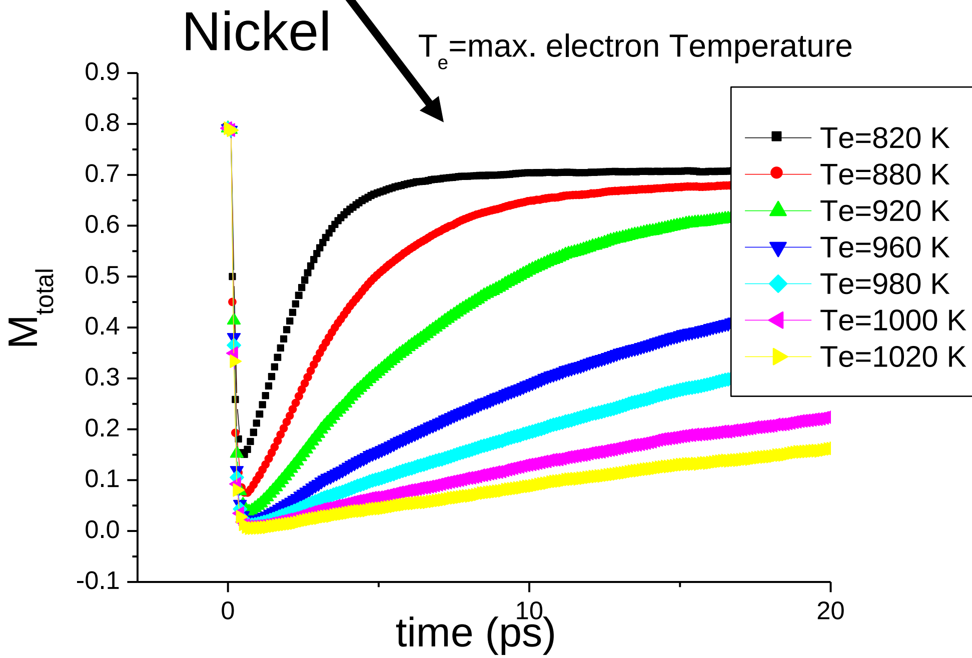
- Essentially micromagnetics with LLG replaced by LLB to simulate the dynamics.
- Exchange between cells taken as $\propto M^2$ (mean-field result)
- Capable of simulating the uncorrelated state after demagnetisation.

# Comparison of atomistic and LLB-μmag model

Atomistic model

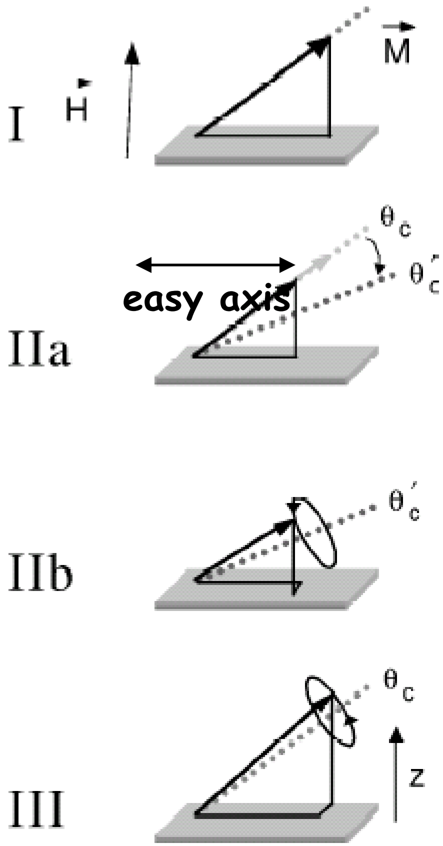
LLB-μmag



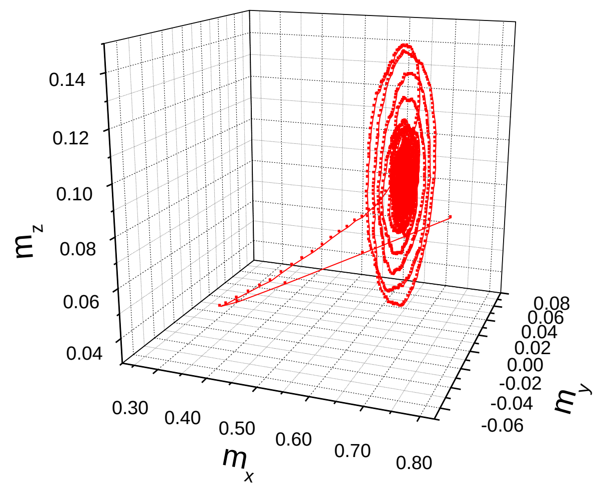

- Calculations with the LLB-μmag model agree well with atomistic calculations, *including the slow recovery*

# Magnetisation precession during all-optical FMR

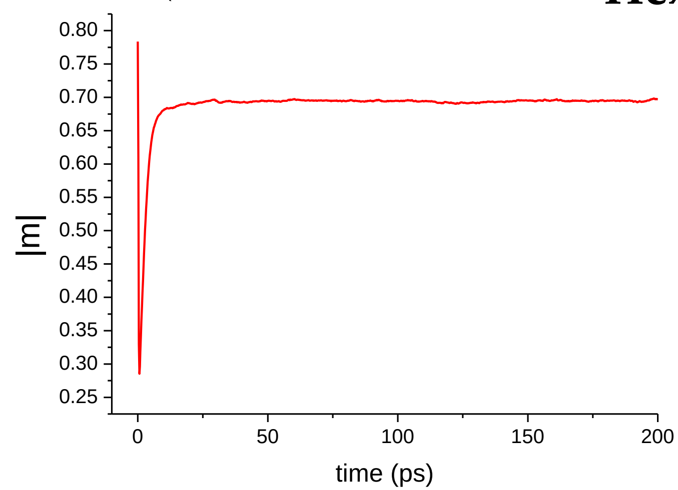



**M.van Kampen et al PRL 88 (2002) 227201**

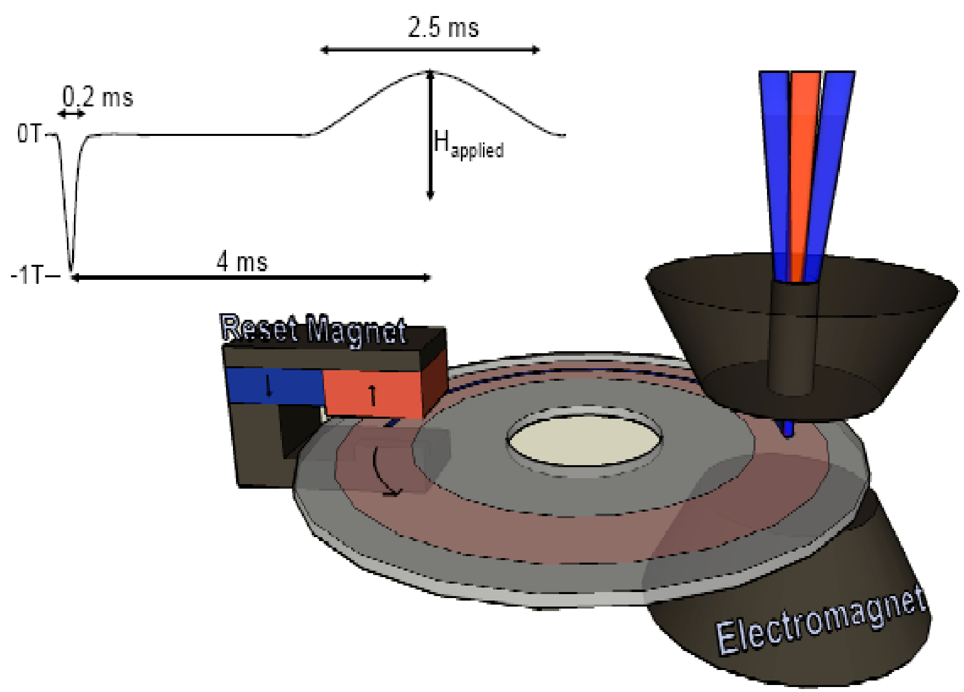
**Our simulation results**




**K(T=0)=5.3 106 erg/cm3**
**Ms(T=0)= 480 emu/cm3**
**Tc=630 K**
**Hext=0.2 T**

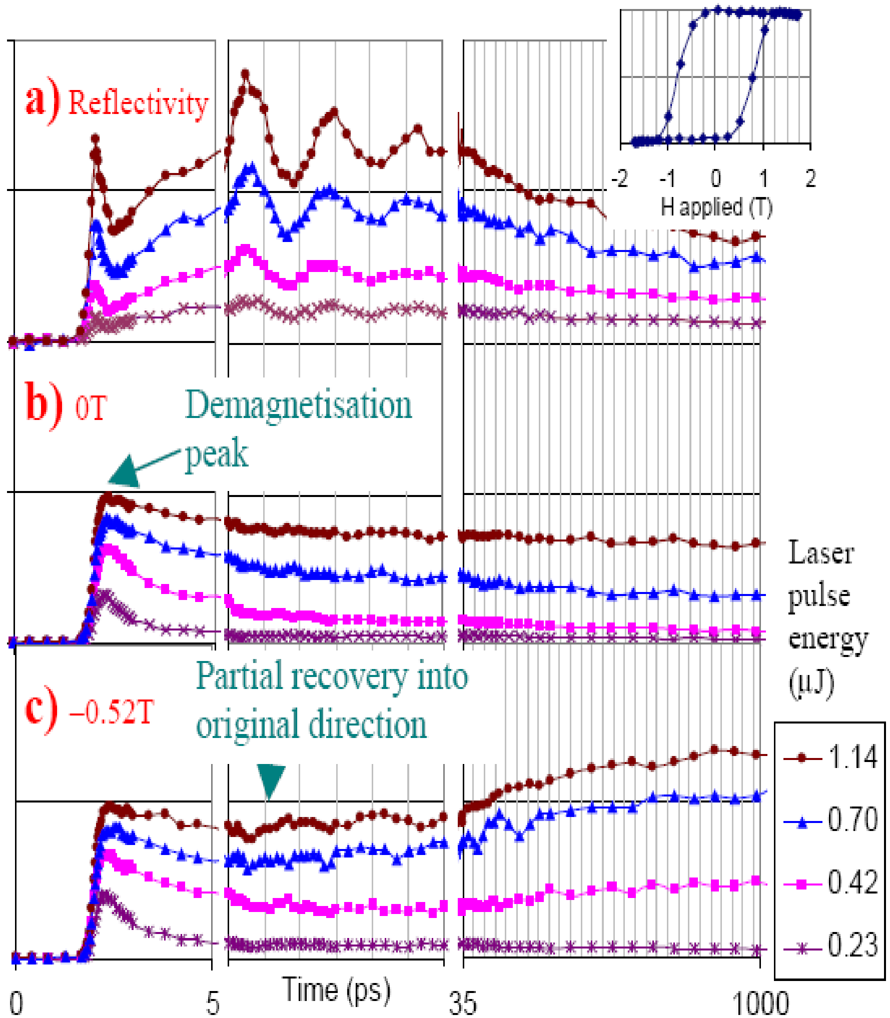


# Experimental studies of Heat Assisted Reversal and comparison with LLB-micromagnetic model




- Experimental set-up (Chris Bunce, York)
- Uses hard drive as a spin-stand to alternate between reset field and reversal field
- Sample used – specially prepared CoPt multilayer (G Ju, Seagate)

# Results

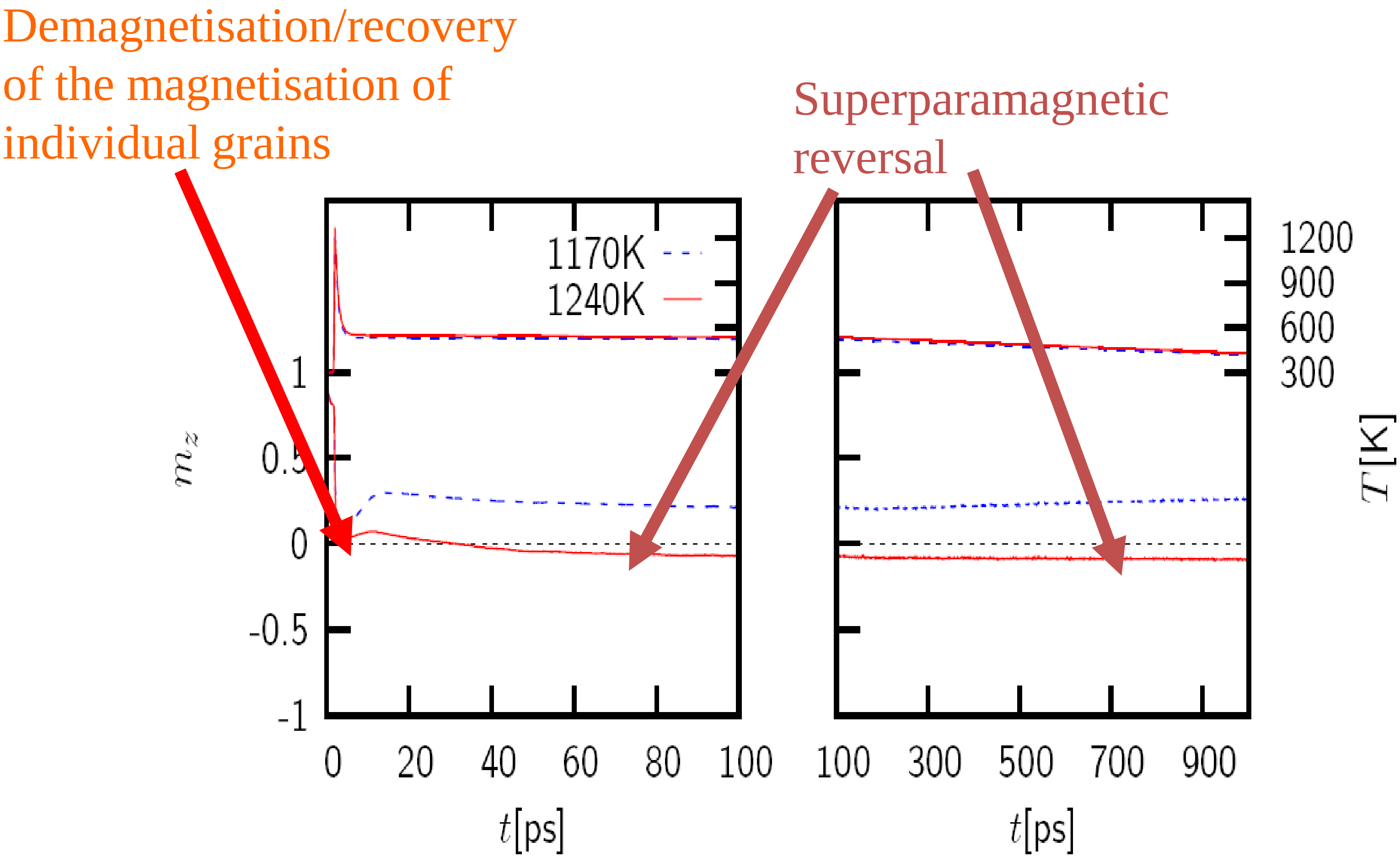



- Reversal occurs in a field of 0.52T (<< intrinsic coercivity of 1.4T
- Note 2 timescales. Associated with Longitudinal (initial fast reduction of M) and transverse (long timescale reversal over particle energy barriers) relaxation

# The computational model

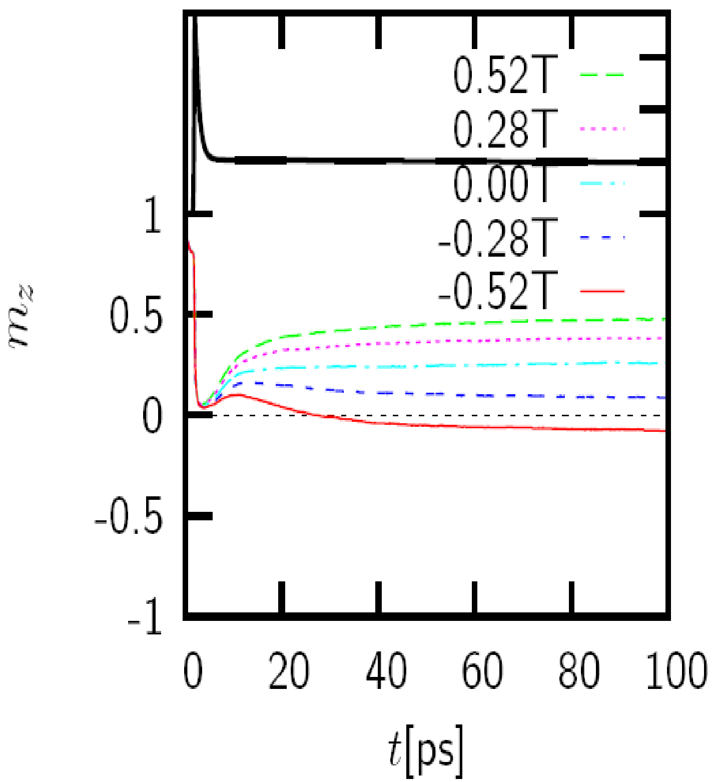
- Film is modelled as a set of grains coupled by exchange and magnetostatic interactions.
- The dynamic behaviour of the grains is modelled using the Landau-Lifshitz-Bloch (LLB) equation.
- The LLB equation allows fluctuations in the magnitude of M. This is necessary in calculations close to or beyond $T_c$.
- The LLB equation can respond on timescales of picoseconds via the longitudinal relaxation time (rapid changes in the magnitude of M) and hundreds of ps - transverse relaxation over energy barriers.
- LLG equation cannot reproduce the longitudinal relaxation
- The film is subjected to a time varying temperature from the laser pulse calculated using a two-temperature model.

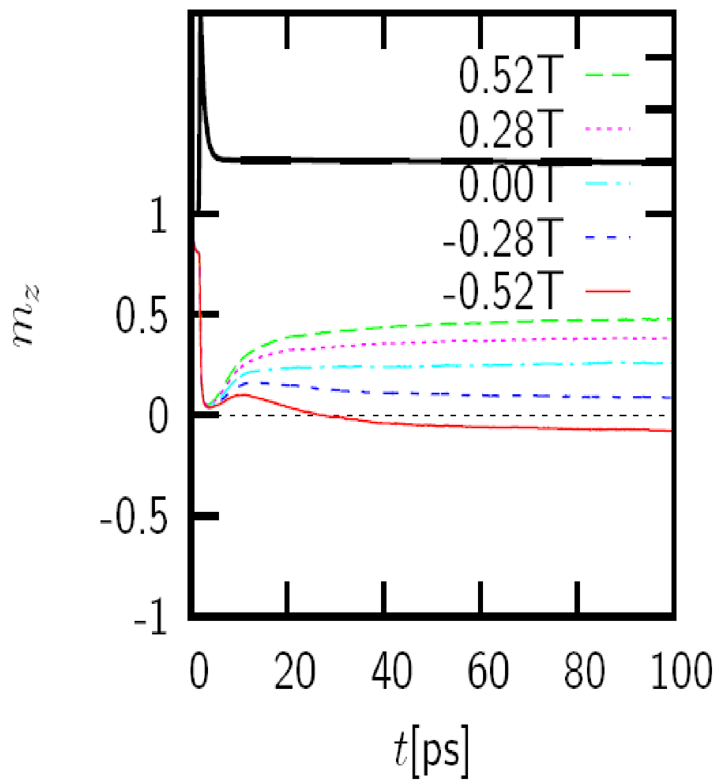
# Calculated results




- Simulations show rapid demagnetisation followed by recovery on the short timescale. Over longer times the magnetisastion rotates into the field direction due to thermally activated transitions over energy barriers.
- This is consistent with experimental results

# Effect of the magnetic field




- Also qualitatively in agreement with experiments
- LLB equation is very successful in describing high temperature dynamics

# Opto-magnetic reversal

PRL 99, 047601 (2007) PHYSICAL REVIEW LETTERS week ending 27 JULY 2007

## All-Optical Magnetic Recording with Circularly Polarized Light

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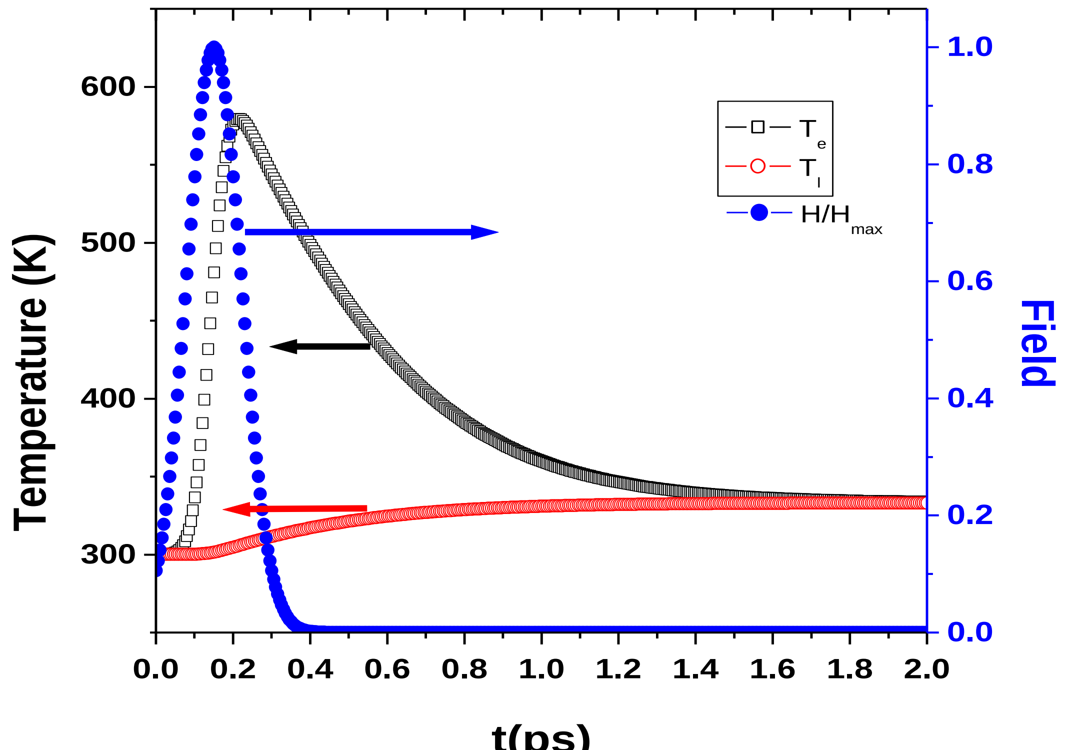
(Received 2 March 2007; published 25 July 2007)

We experimentally demonstrate that the magnetization can be reversed in a reproducible manner by a single 40 femtosecond circularly polarized laser pulse, without any applied magnetic field. This optically induced ultrafast magnetization reversal previously believed impossible is the combined result of femtosecond laser heating of the magnetic system to just below the Curie point and circularly polarized light simultaneously acting as a magnetic field. The direction of this opto-magnetic switching is determined only by the helicity of light. This finding reveals an ultrafast and efficient pathway for writing magnetic bits at record-breaking speeds.

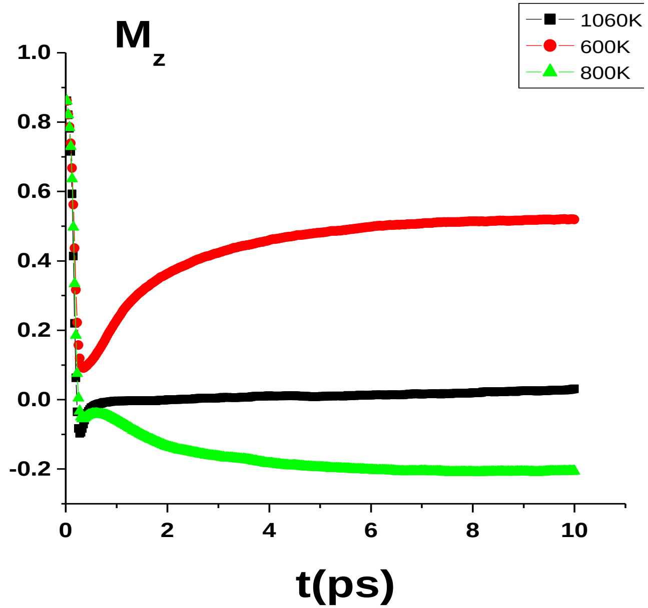
- What is the reversal mechanism?
- Is it possible to represent it with a spin model?

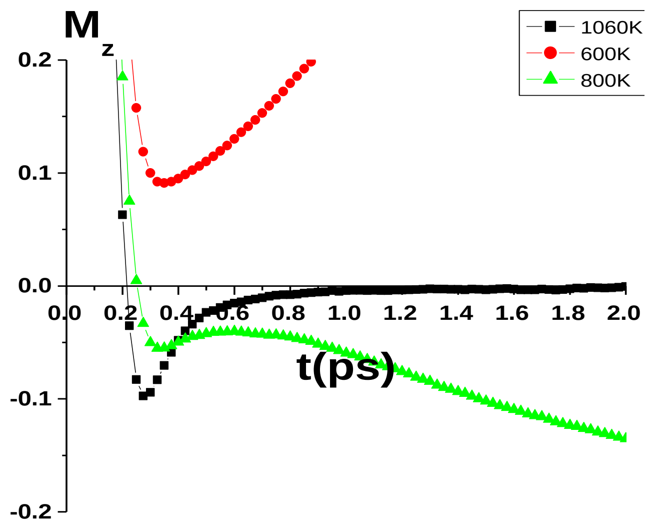
# Fields and temperatures




- Simple '2-temperature' model
- Problem – energy associated with the laser pulse (here expressed as an effective temperature) persists much longer than the magnetic field.
- Equlibrium temperature much lower than $T_c$

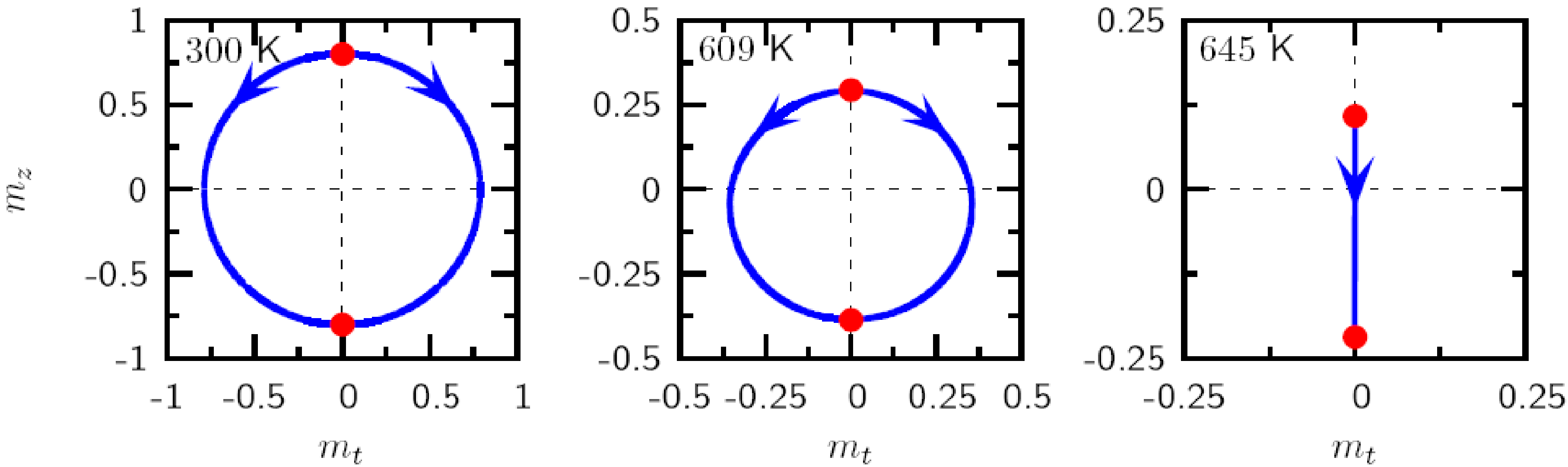
# Magnetisation dynamics (atomistic model)



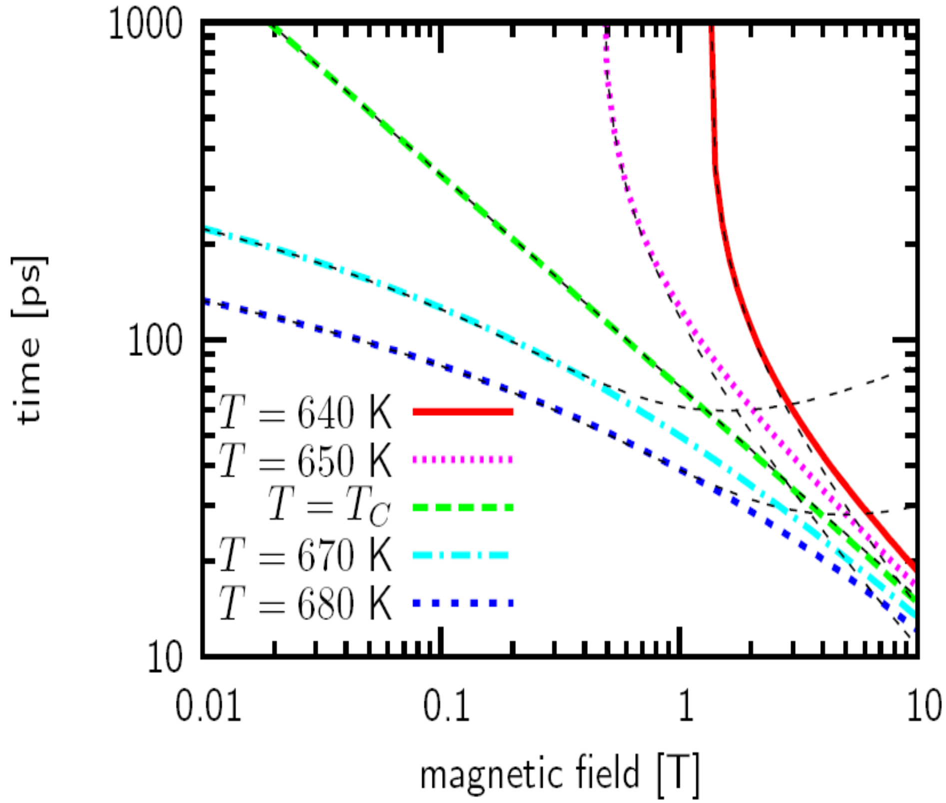

- ***Reversal is non-precessional*** – $m_x$ and $m_y$ remain zero. ***Linear reversal mechanism***
- Associated with increased magnetic susceptibility at high temperatures
- Too much laser power and the magnetisation is destroyed after reversal
- Narrow window for reversal

# Linear reversal



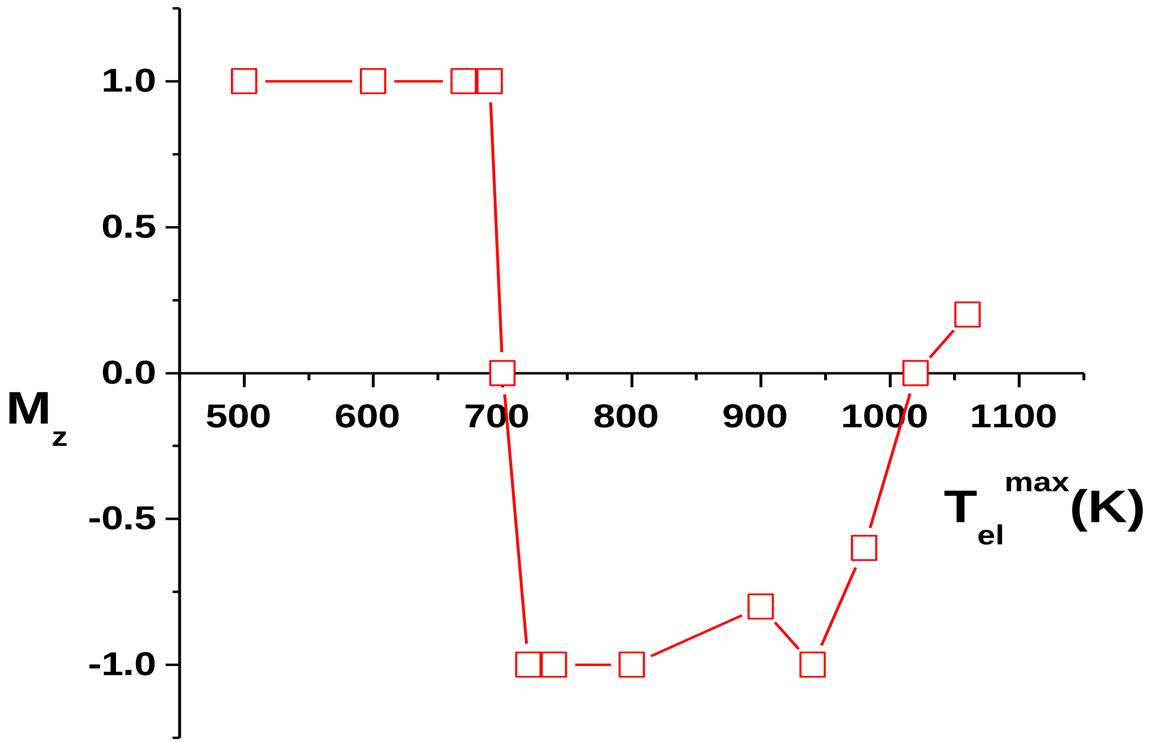

New reversal mechanism via a strongly non-uniform (demagnetised) state.
VERY fast (timescale of longitudinal relaxation)
Micromagnetics with LLG equation cannot reproduce behaviour

# Analytical calculations of relaxation times using LLB equation



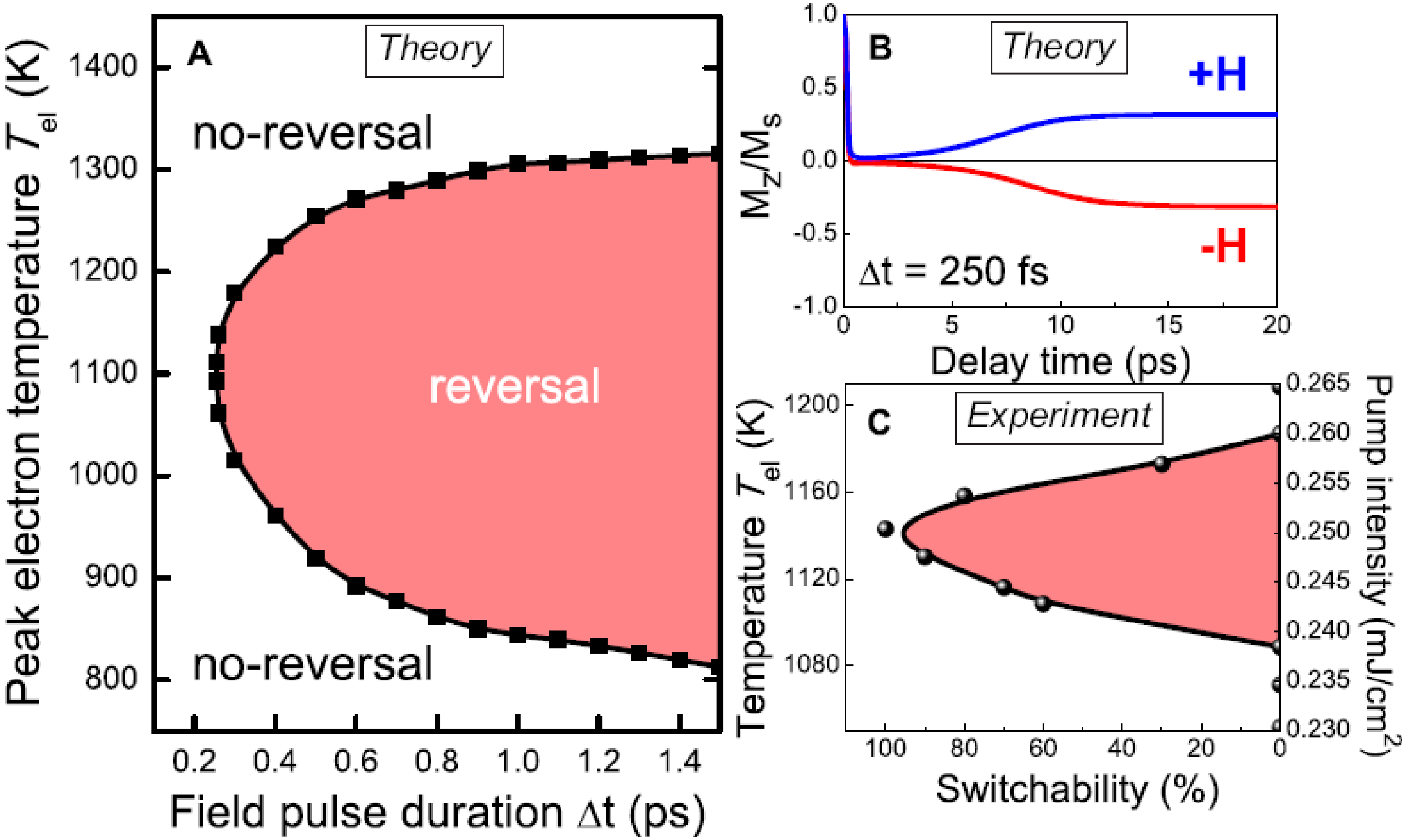

Large fields required for ps reversal (Kazantseva et al, EPL, in press)

# 'Reversal window'




- Well defined temperature range for reversal
- Critical temperature for the onset of linear reversal
- BUT atomistic calculations are very CPU intensive
- LLB micromagnetic model used for large scale calculations

# Reversal 'phase diagram'
Vahaplar et al Phys. Rev. Lett., 103, 117201 (2009)




- Note the criticality of the experimental results
- Characteristic of linear reversal

# End of the story? Not quite!

- Calculations suggest a thermodynamic contribution (linear reversal).
- But
  - Energy transfer channels are not well represented
  - What is the origin of the field – Inverse Faraday Effect?
  - Electron/phonon coupling plays a role
  - Role of the R-E – is this important?
- These require detailed studies at the ab-initio level – the multiscale problem still remains!

# Conclusions

- Atomistic model has been developed using Heisenberg exchange.
- The Landau-Lifshitz-Bloch (LLB) equation incorporates much of the physics of the atomistic calculations
- LLB-micromagnetics is proposed, essentially using the LLB equation in a micromagnetic formalism.
- LLB-micromagnetics is shown to be successful in simulating ultrafast dynamics at elevated temperatures. Important for pump-probe simulations and models of HAMR. Also thermally assisted MRAM?
- New (linear) reversal demonstrated with sub-picosecond reversal times
- Demonstrates the probable thermodynamic origin of Opto-Magnetic reversal.

# Future developments

- Micromagnetics will continue as the formalism of choice for large scale simulations under normal conditions of temperature and timescale
- However, atomistic and multiscale calculations are vital for ultrafast dynamics
- Challenges
  - Picosecond dynamics
  - Damping mechanisms
  - Introduction of spin torque
  - Link between magnetic and transport models
  - Models of atomic level microstructure are necessary. (The ultimate problem of magnetism vs microstructure?)

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