# Modelling 2:
# Multiscale calculations

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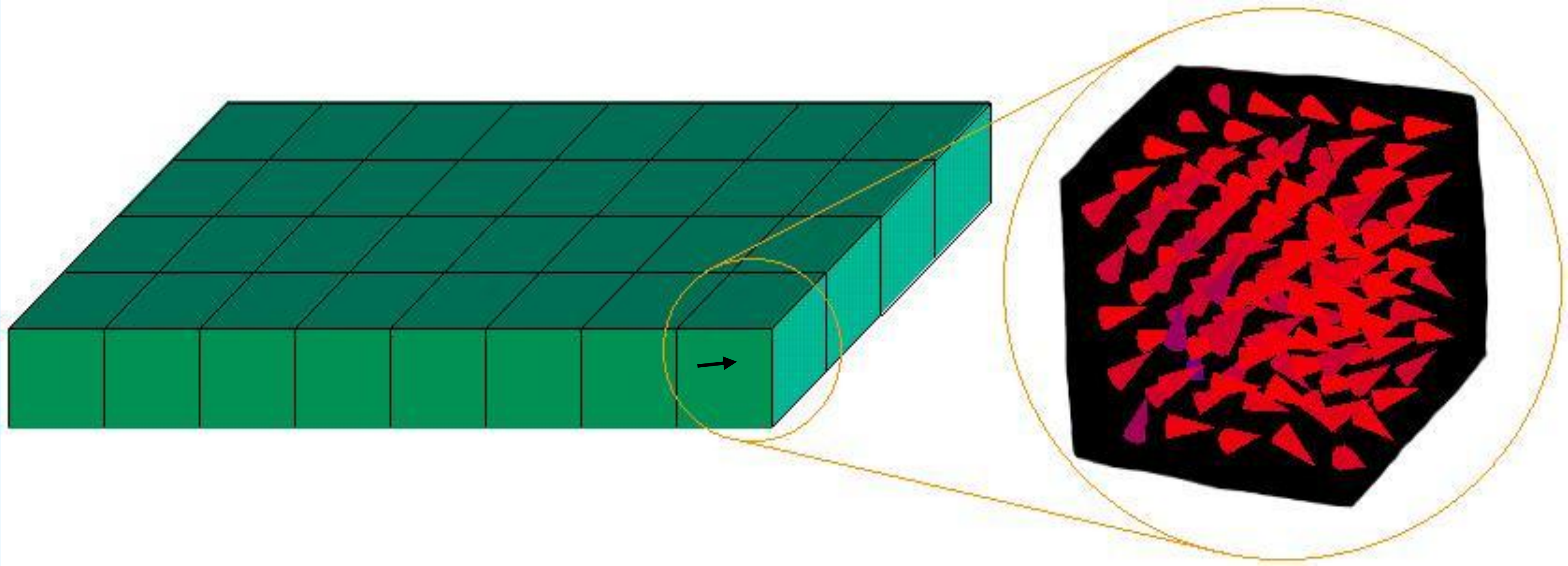
# Summary

- The lengthscale problem
- A simple multiscale approach to the properties of nanostructured materials
- Studies of soft/hard magnetic bilayers
- Dynamics and the Landau-Lifshitz- Bloch (LLB) equation of motion
- LLB-micromagnetics and dynamic properties for large-scale simulations at elevated temperatures
- The two timescales of Heat Assisted reversal; experiments and LLB-micromagnetic model

# Multiscale magnetism

- Need is for links between ab-initio and atomistic models
- BUT comparison with experiments involves simulations of large systems.
- Typically magnetic materials are ‘nanostructured’, ie designed with grain sizes around 5-10nm.
- Permalloy for example consists of very strongly exchange coupled grains.
- Such a ‘continuous’ thin film cannot be simulated atomistically



- For pump-probe simulations it would be ideal to have a ‘macrospin’ approximation to the atomistic model

# Length scales

Electronic ⟶ atomistic ⟶ micromagnetic

- Here the atomistic ⟶ micromagnetic process is illustrated using
  - Simple approach using macrocells and LLG-based micromagnetics
  - Introduction of the Landau-Lifshitz-Bloch (LLB) equation and LLB-micromagnetics
  - pump-probe experiments simulated by LLB-micromagnetics

# Magnetic Recording goes 'nano'

- Media grain sizes currently around 7-8 nm. Must be reduced to 5nm or below for 1TBit/sqin and beyond
- 'Ultimate' recording densities (around 50TBit/sqin would need around 3nm FePt grains
- Some advanced media designs require complex composite structures, eg soft/hard layers
- To what extent can micromagnetics cope with these advanced structures?

# The need for atomistic/multiscale approaches (recap)

- 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.

# Micromagnetic exchange

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

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

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.

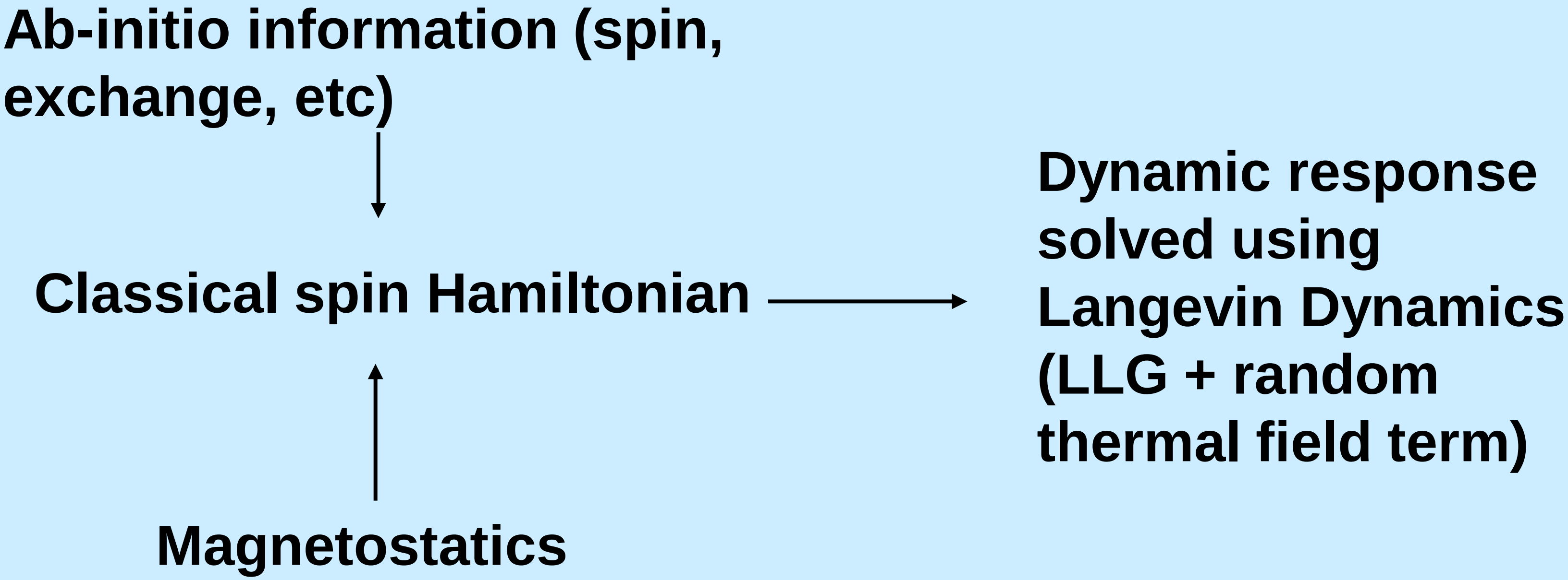
# Atomistic model

- 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**

**Magnetostatics**

**Dynamic response solved using Langevin Dynamics (LLG + random thermal field term)**

# Dynamic behaviour

- Dynamic behaviour of the magnetisation is based on the Landau-Lifshitz-Gilbert equation

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

- Where $\gamma_0$ is the gyromagnetic ratio and $\alpha$ is a damping constant

# Langevin Dynamics

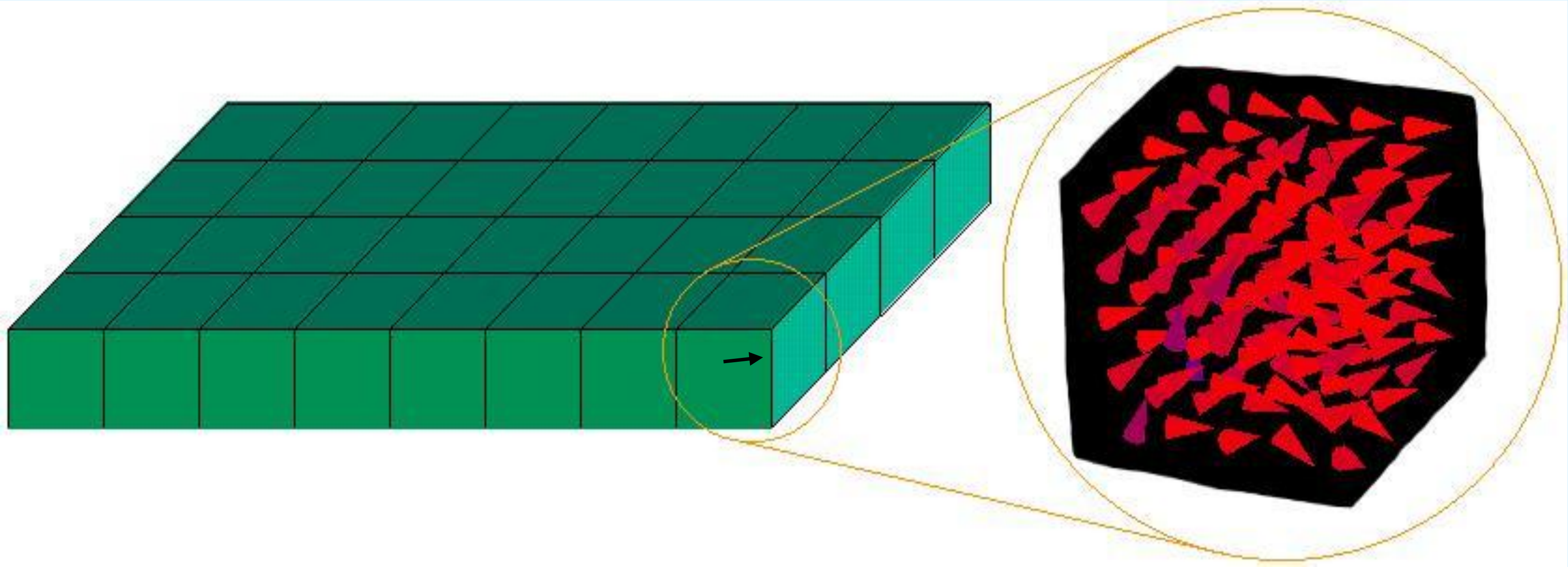
- Based on the Landau-Lifshitz-Gilbert equations with an additional stochastic field term h(t).
- From the Fluctuation-Dissipation theorem, the thermal field must must have the statistical properties

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

- From which the random term at each timestep can be determined.
- h(t) is added to the local field at each timestep.

# Magnetostatic term (2 approaches)

1. Use FFT at atomic level. This is exact but time consuming.
2. Average the magnetisation over ‘macrocells’ containing a few hundred atoms. The field from this magnetisation can be calculated using standard micromagnetic techniques. Most often this technique reduces the magnetostatic problem to a relatively small calculation.



- Average moments used to calculate fields
- Neglects short wavelength fluctuations of the magnetostatic field.
- However, this should not be a bad approximation since short wavelength fluctuations will be dominated by exchange.

# Scaling models.

- The problem – introduction of short-wavelength fluctuations into micromagnetics
- Solutions:

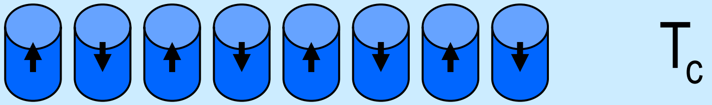
1. Coarse graining (V.V. Dobrovitski, M. I. Katsnelson and B. N. Harmon, J. Magn. Magn. Mater. 221, L235 (2000), PRL 90, 6, 067201 (2003)
2. Renormalisation group theory (G. Grinstein and R. H. Koch, Phys. Rev. Lett. 90, 207201 (2003) )
3. Numerical calibration of M(T), K(T) … (M Kirschner et al J Appl. Phys., 9710E301(2005))

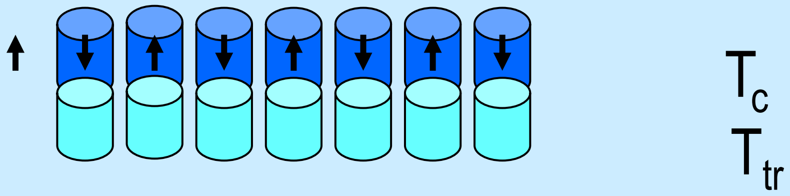
- These approaches scale the normal micromagnetic parameters and do not take explicit account of interfaces
- Here we describe a 'multiscale' model which explicitly links micromagnetic and atomistic regions.

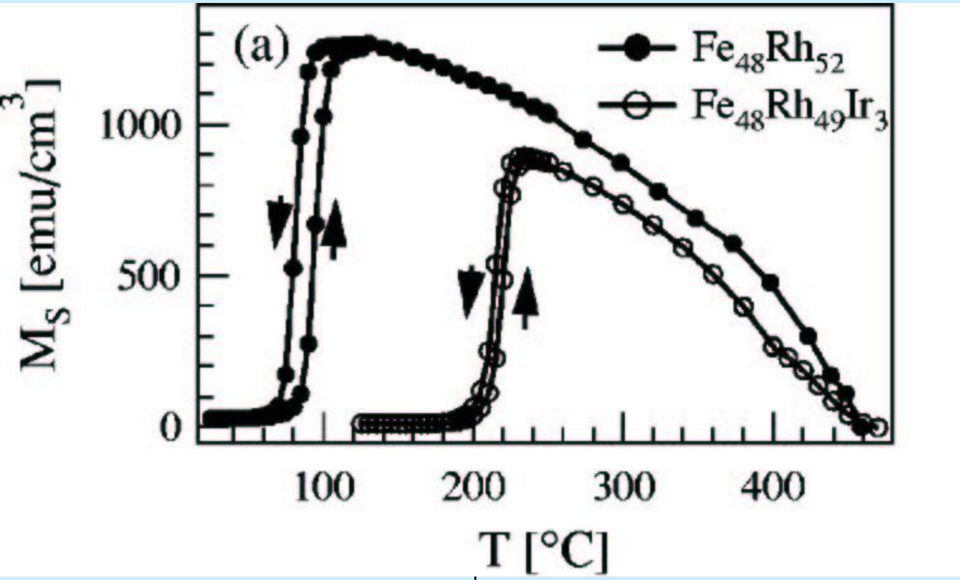
# Multiscale models

- H. Kronmuller, R. Fischer, R. Hertel and T. Leineweber, J. Magn. Magn. Mater. 175, 177 (1997); H. Kronmuller and M. Bachmann, Physica B 306, 96 (2001).
- F. Garcia-Sanchez and O. Chubykalo-Fesenko, O. Mryasov and R.W. Chantrell and K.Yu. Guslienko, APL 87, 122501 (2005)
- The technique involves partitioning the system into regions (such as interfaces) where an atomistic approach is required, and ‘bulk’ regions in which the normal micromagnetic approach (with suitably scaled parameters) can be applied.
- Here we illustrate the approach using as an example calculations of exchange spring behaviour in FePt/FeRh composite media – proposed by Thiele et al (APL, 82, 2003) for write temperature reduction in HAMR
- Also applied to the exchange spring bilayers proposed by Suess et al (J. Magn. Magn. Mater. **287**, 41 (2005), Appl. Phys. Lett. **87**, 012504 (2005)).

# Composite media using metamagnetic transition to soft underlayer

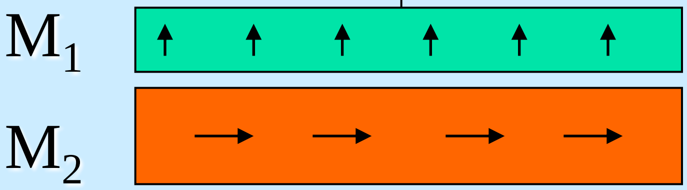






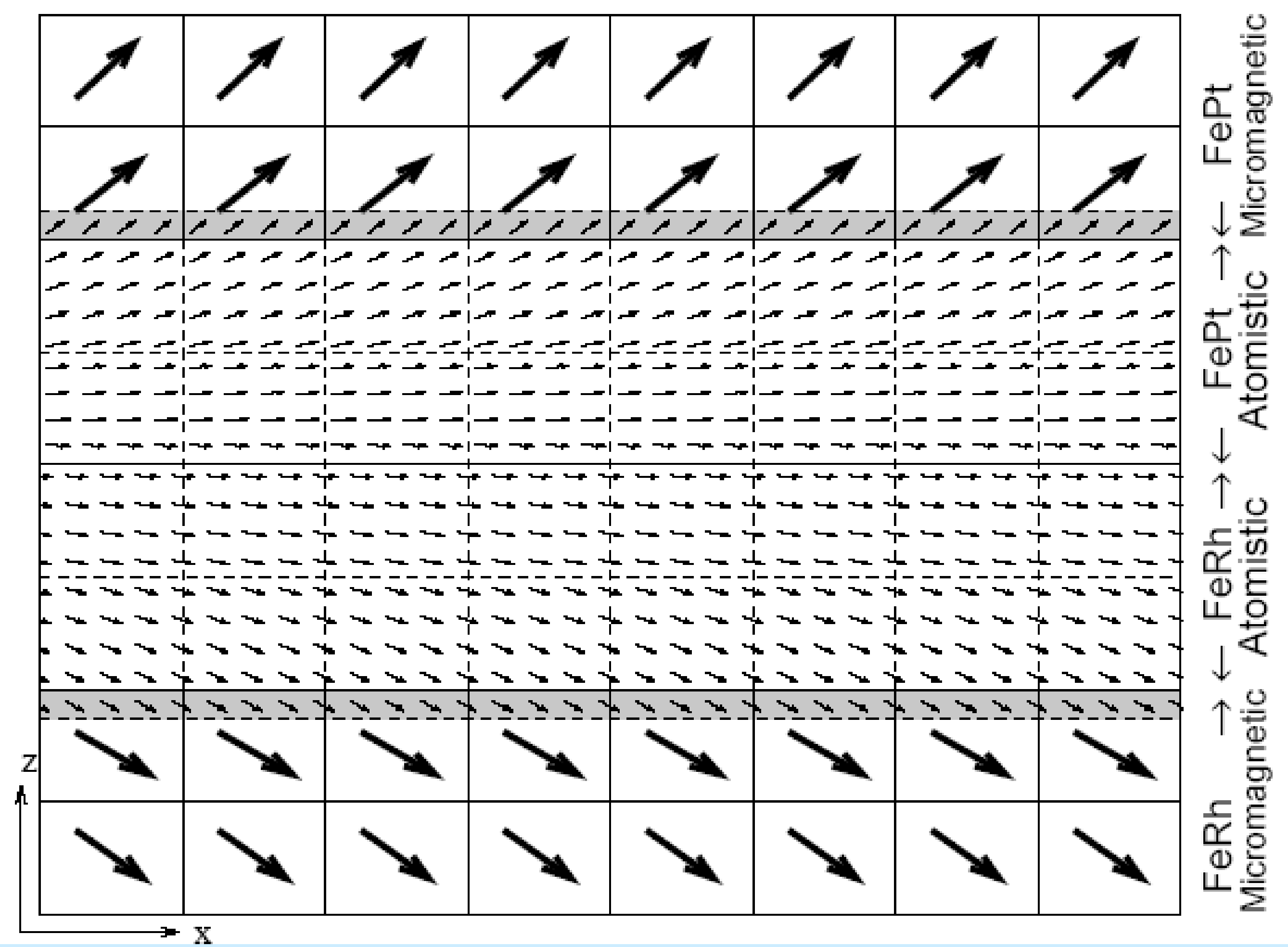
AFM -> FM

Thiele, Maat, Fullerton
APL, 82, 2003
Exchange spring films for HAMR



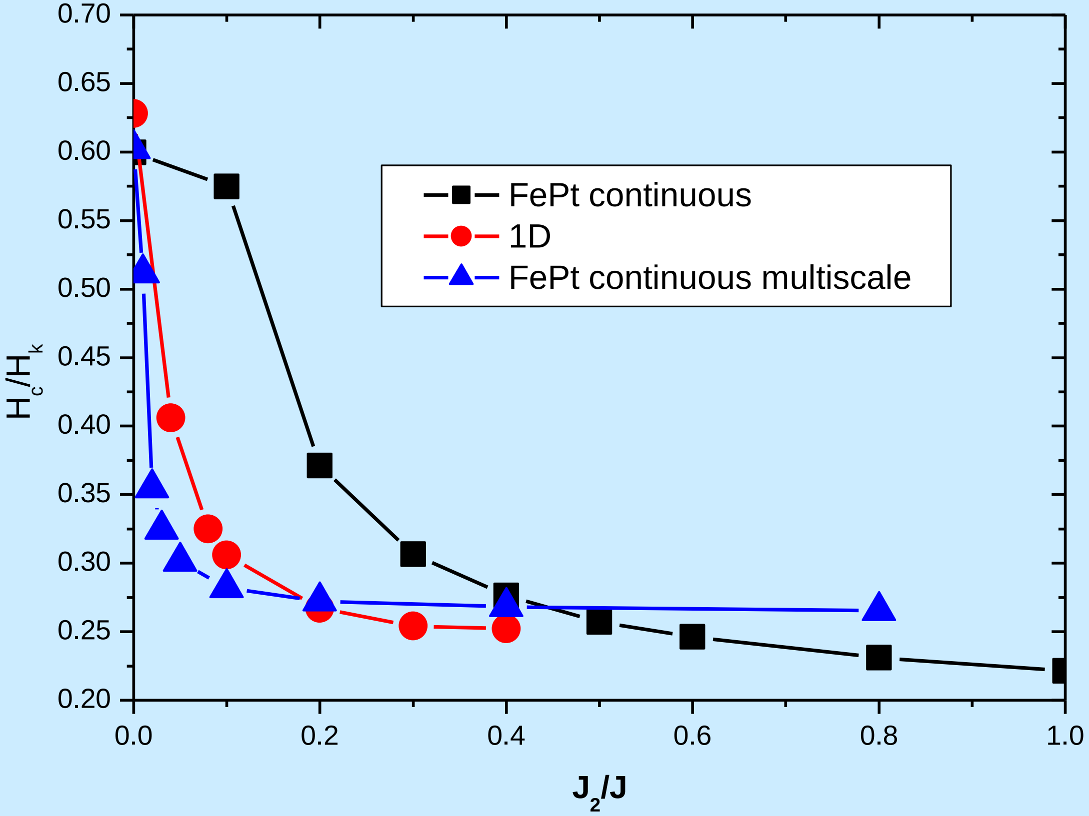
- hard layer with perp. anisotropy (FePt)
- soft layer with AF-F transition (FeRh)

**Physical mechanism:** crossing AF-F critical temperature induces Magnetization in soft layer and decreases $H_c$ of hard layer in 2-3 times within narrow T-interval due to interlayer exchange coupling



Schematic outline of the multiscale approach. Atomistic and micromagnetic layers are indicated. Coupling between the regions is achieved by a layer of ‘virtual’ atoms in the interfacial micromagnetic layer.

# Coercivity reduction due to soft layer



- $H_c$ depends on the interfacial coupling $J_s$
- Numerical results (multiscale) agree reasonably well with (1D) semi-analytical results (FePt continuous)
- Poor agreement with micromagnetic model

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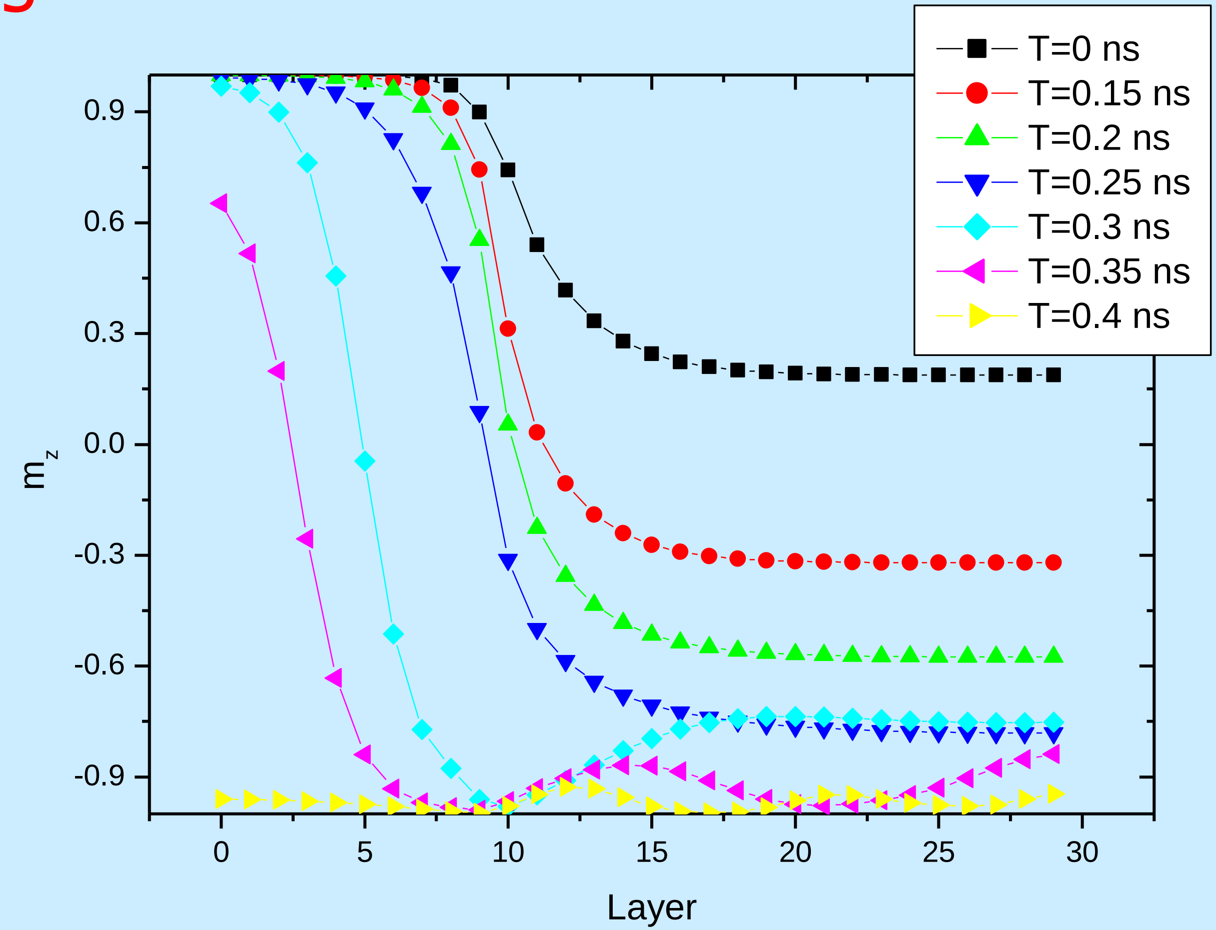
# Exchange spring behaviour (multiscale model) – propagation of DW



15 nm FePt 30 nm FeRh, H= 0.55 $H_k$ = 2 T

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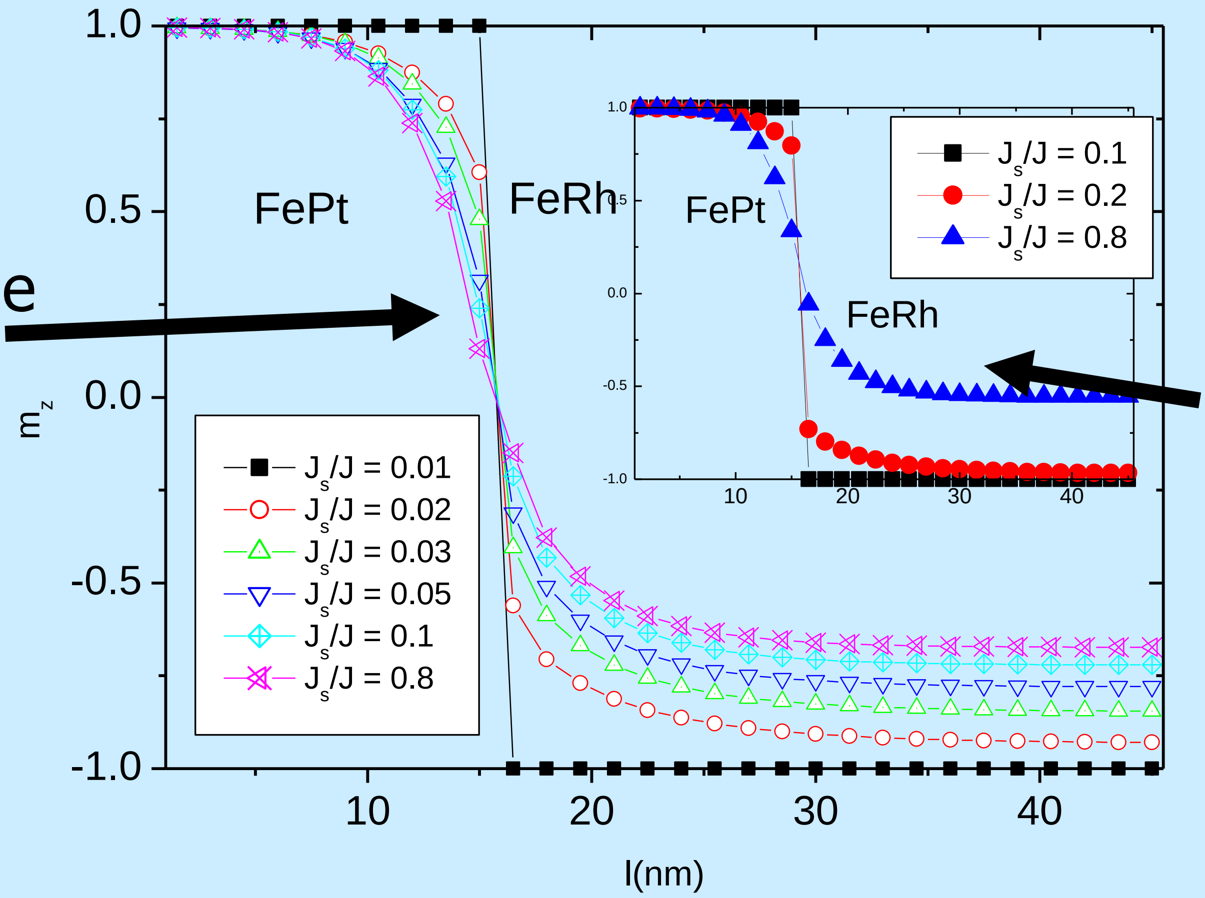
# Comparison with micromagnetic model

Domain Walls for applied field near coercive field in multiscale calculations



Multiscale model

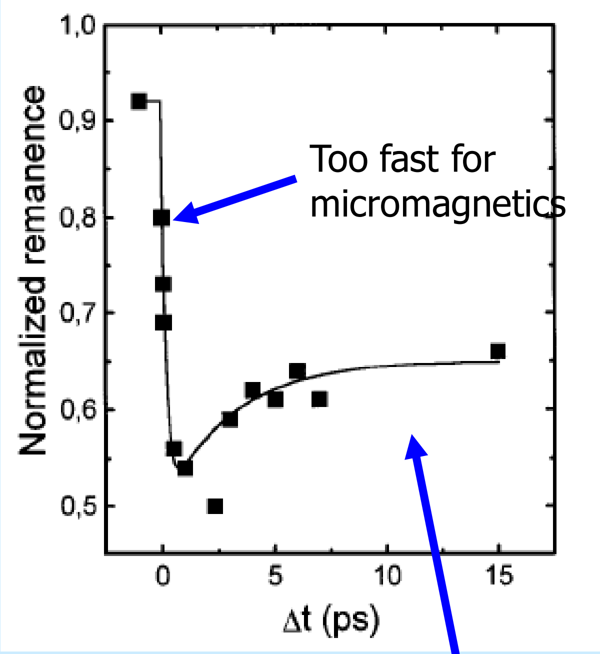
Micromagnetic model

- Tendency of micromagnetic formalism to under estimate the the exchange energy allows non-physically large spatial variation of M.
- Explains the need for large interface coupling (according to micromagnetics)to give coercivity reduction

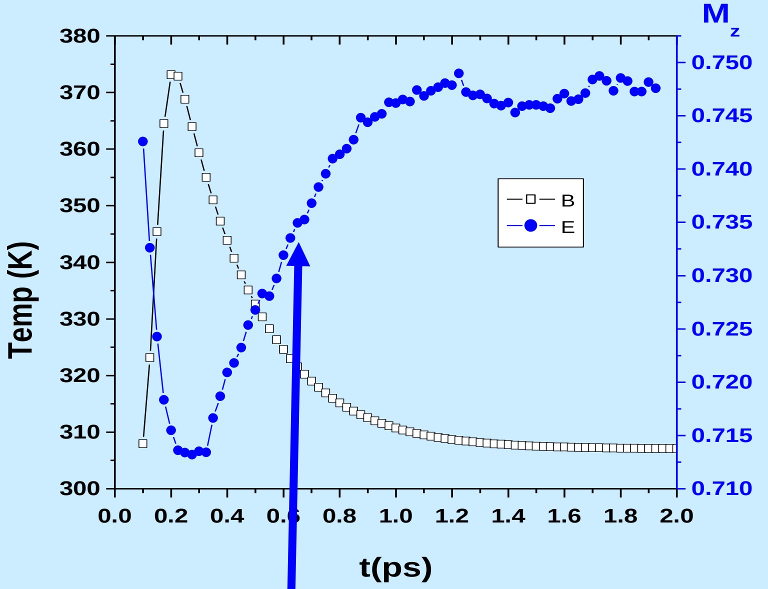
# Multiscale calculations and the LLB equation

- Large scale (micromagnetic) simulations essentially work with one spin/computational cell
- Single spin LLG equation cannot reproduce this reversal mechanism (conserves |M|)
- Pump- probe simulations require an alternative approach
- Landau-Lifshitz-Bloch (LLB) equation?

# Ultrafast demagnetisation

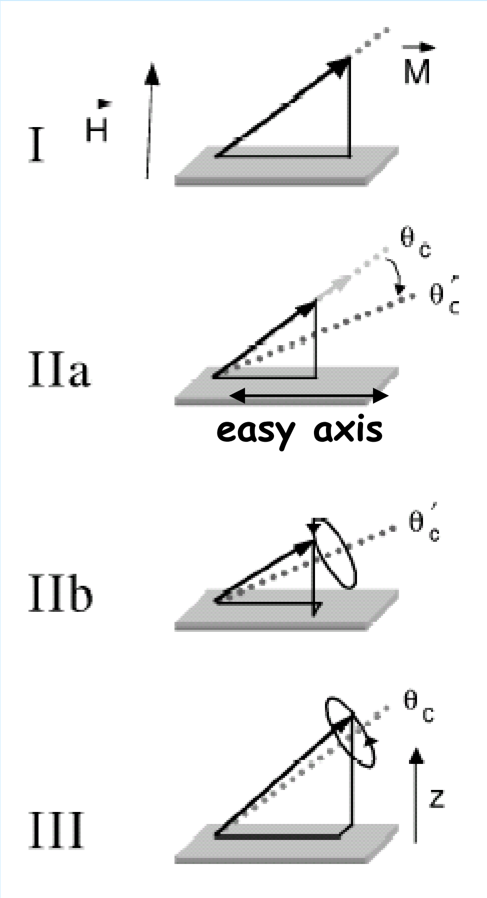


Too fast for micromagnetics

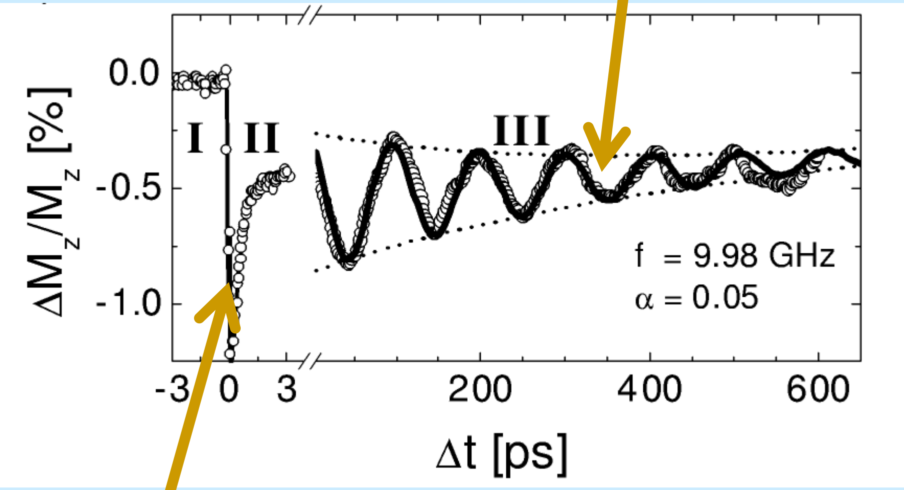


- Experiments on Ni (Beaurepaire et al PRL <u>76</u> 4250 (1996)
- Atomistic calculations for peak temperature of 375K
- These work because the atomistic treatment gets right the (sub-picosecond) longitudinal relaxation time. Only possible for atomic-level theory.

# Magnetisation precession during all-optical FMR

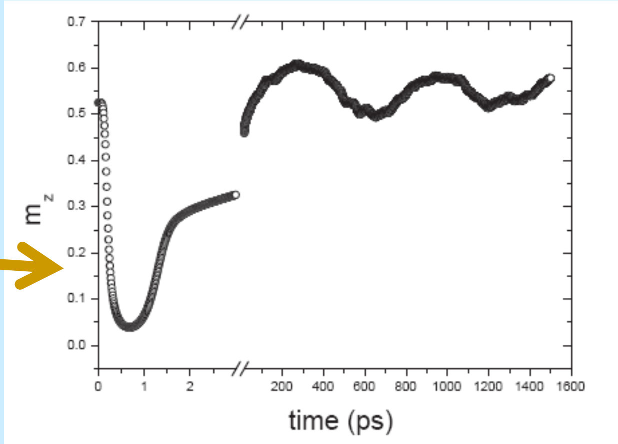


Micromagnetics can do this, BUT NB α is temperature dependent (as predicted by atomistic simulations)



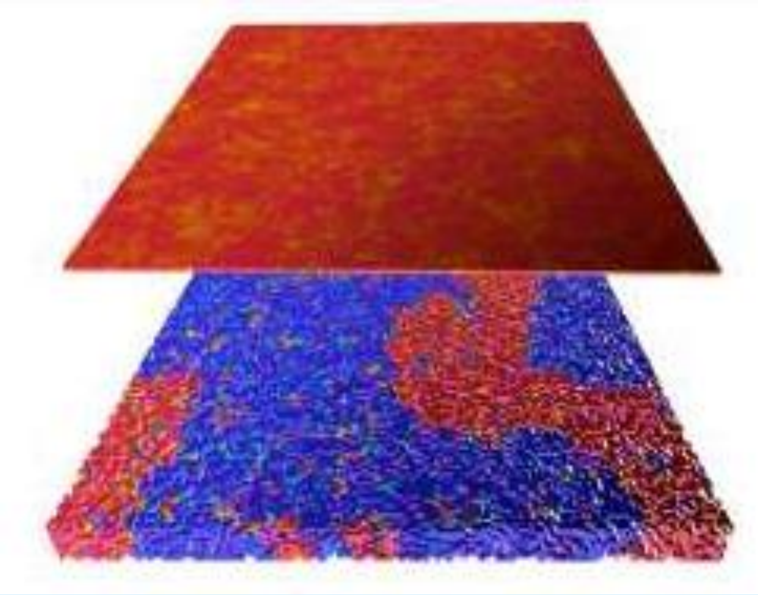
But it cannot do this!

Atomistic + LLB-μ-mag calculations **can** (Atxitia et al APL 91, 232507 2007)

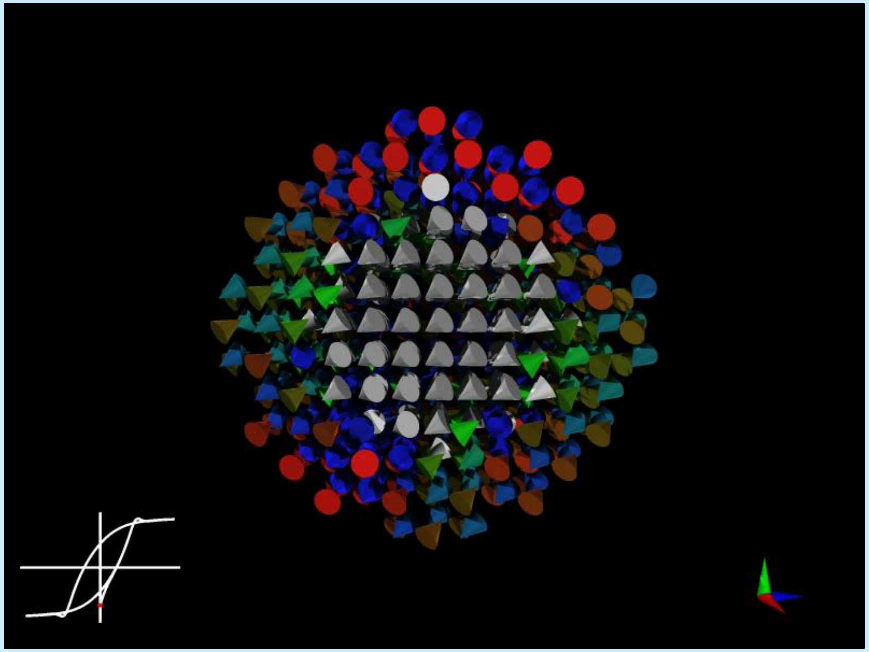


**M.van Kampen et al PRL 88 (2002) 227201**

# Complex nanostructures



Domain state model of FM/AF bilayer (Jerome Jackson)



Core/shell FM/AF structure (Dan Bate, Richard Evans, Rocio Yanes and Oksana Chubykalo-Fesenko)

# Extended micromagnetics; LLB equation

Longitudinal term introduces fluctuations of M

Transverse (LLG) term

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

- 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:

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

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

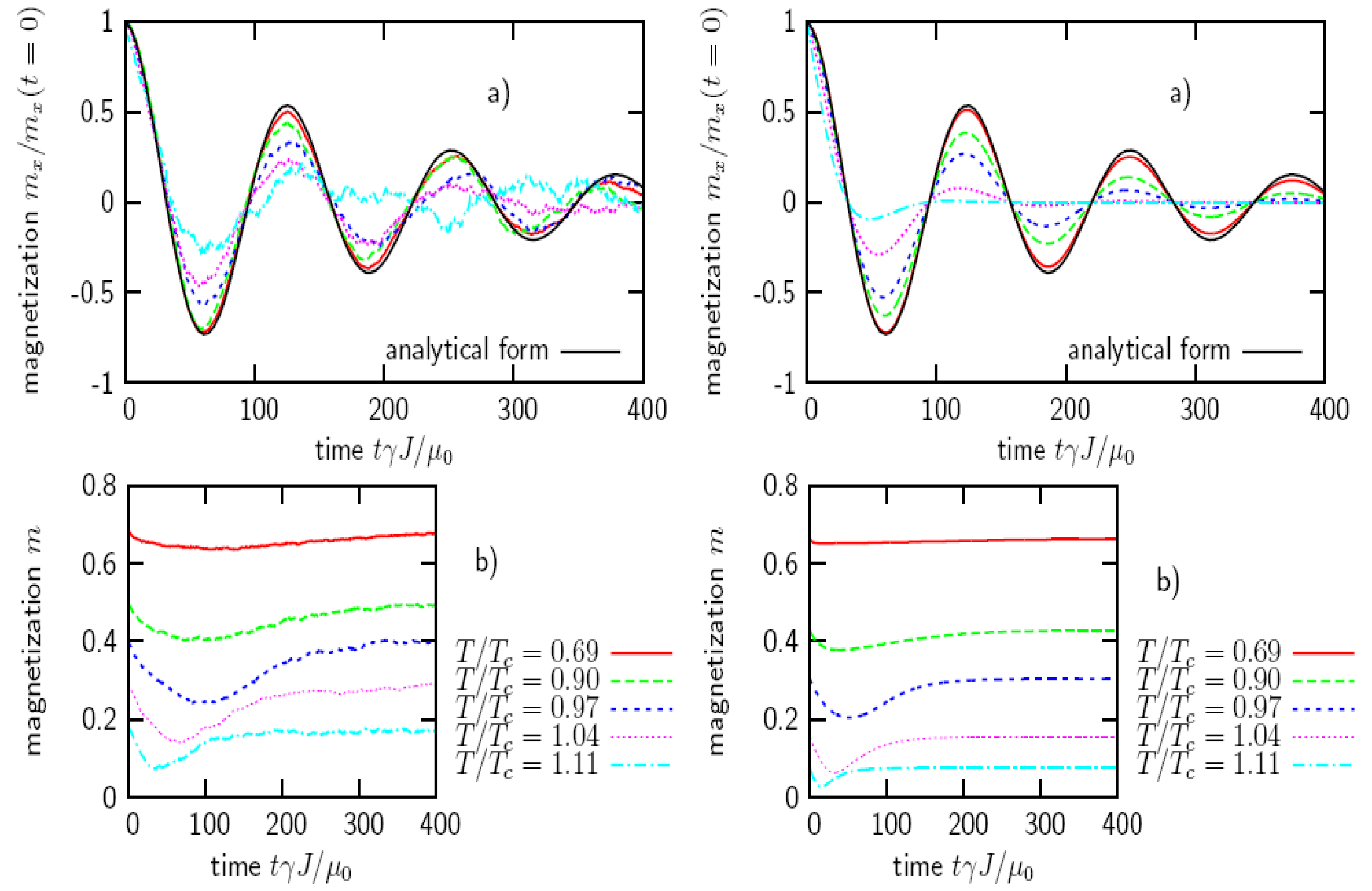
# Multiscale calculations

Electronic → atomistic → micromagnetic

Case by case basis, eg FePt (Mryasov et al, Europhys Lett., 69 805-811 (2005)

Landau-Lifshitz-Bloch equation

Treatment of the whole problem for FePt given by Kazantseva et al Phys. Rev. B 77, 184428 (2008)



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

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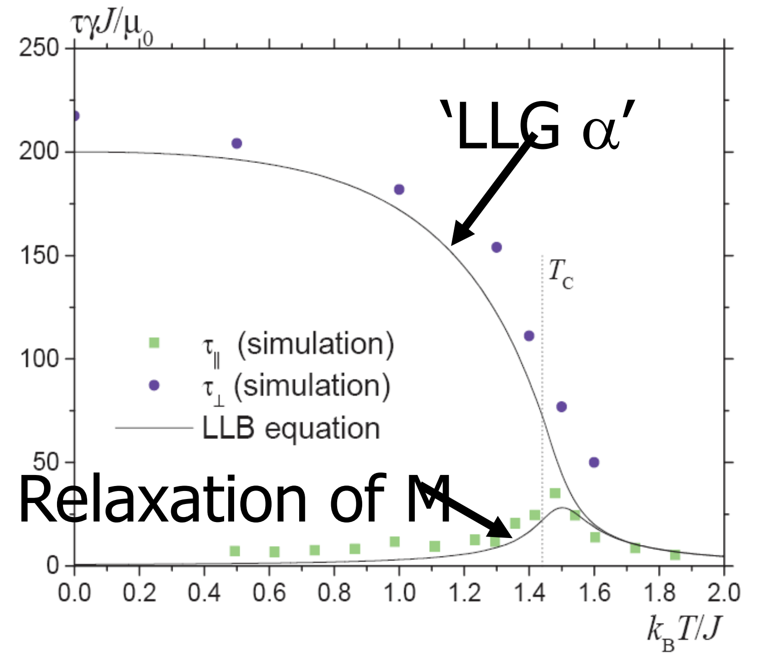
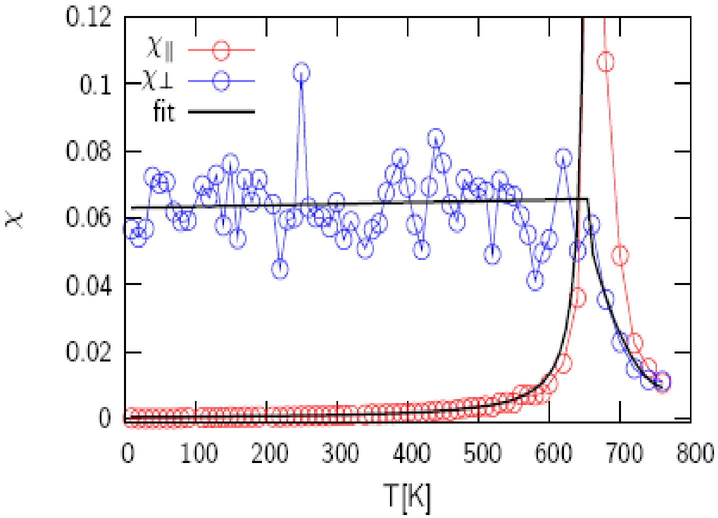
# Relaxation times



- Effective α 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.

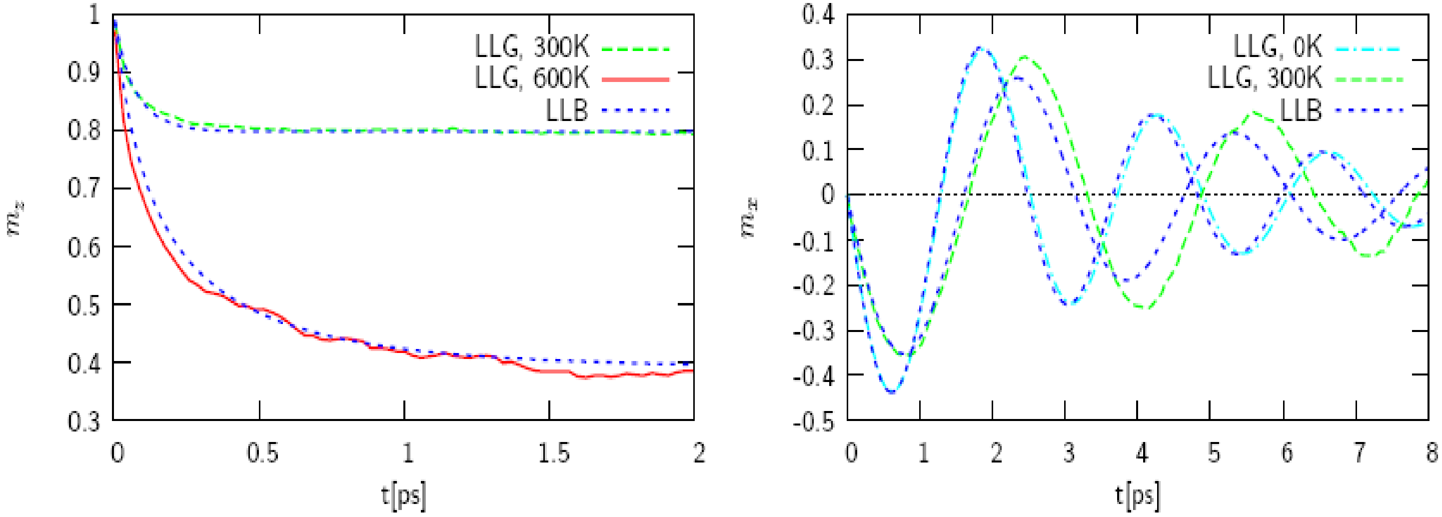
# LLB parameters

- 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.



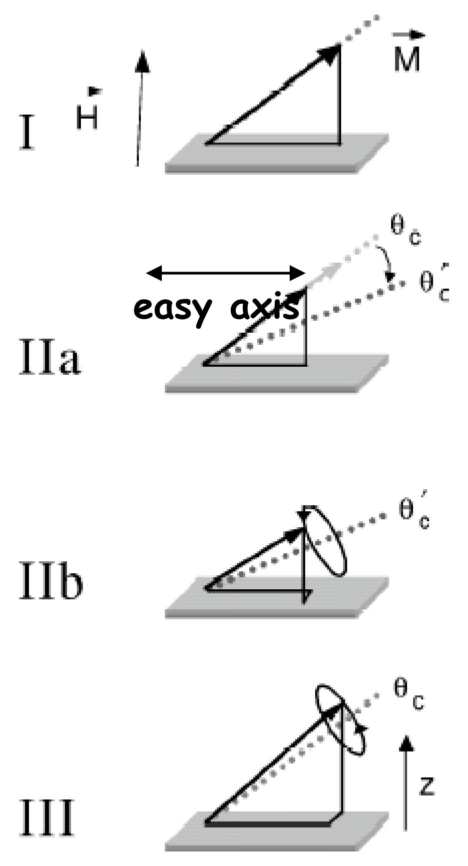
- spin model for FePt [4]
- LLG calculations for a grain of size $32 \times 32 \times 48$ spins
- $\tilde{\chi}_\perp$ (left fig.), $\tilde{\chi}_\parallel$ and $m_e$ were evaluated and used for LLB calculations

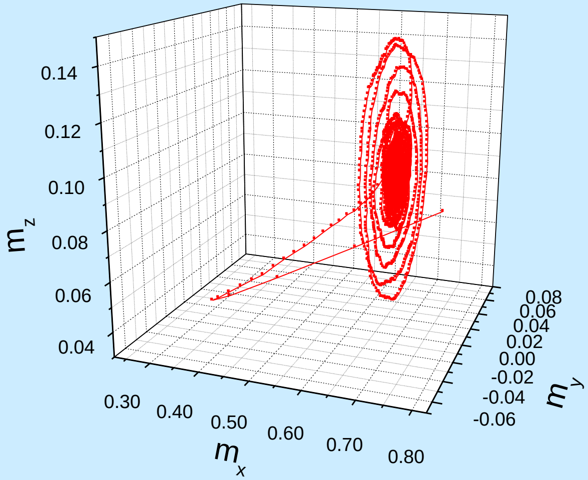
longitudinal relaxation (left) and transverse relaxation after 30° exitation (right) for atomistic LLG and macro-spin LLB modeling

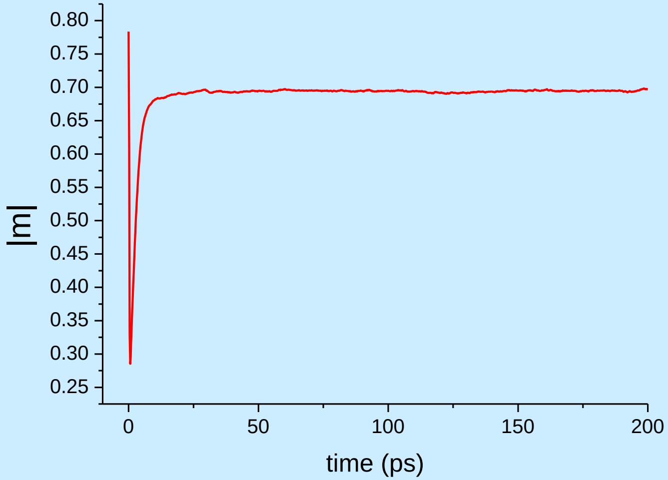


# Magnetisation precession during all-optical FMR

**Our simulation results**







$K(T=0)=5.3\ 10^6$ erg/cm$^3$
$M_s(T=0)=480$ emu/cm$^3$
$T_c=630$ K
$H_{ext}=0.2$ T

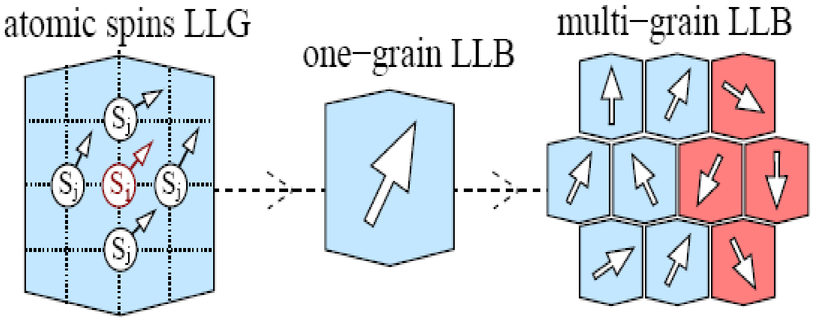
**M.van Kampen et al PRL 88 (2002) 227201**

# Reprise; Multi-scale modelling

- concept: multi-scale modelling

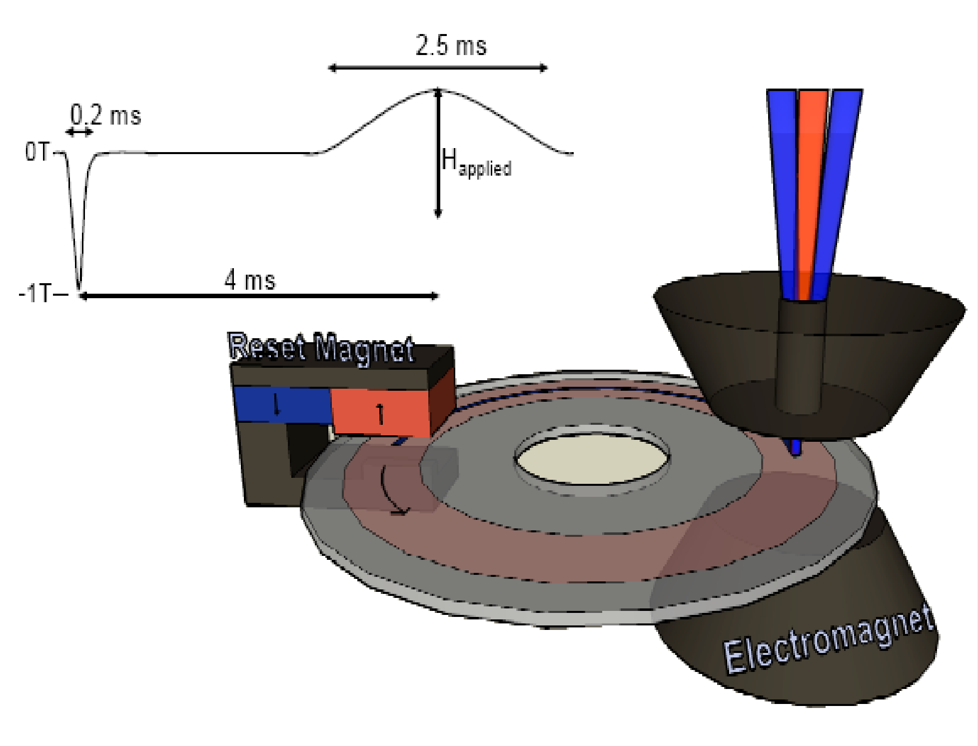
**electronic structure:** DFT theory, single (periodic) atom or clusters ⇔ **atomic spin model:** molecular dynamics: up to $10^6$ spins ⇔ **continuum theory:** micromagnetism: up to some $\mu m^3$

- simulation model using Landau-Lifshitz-Bloch equation



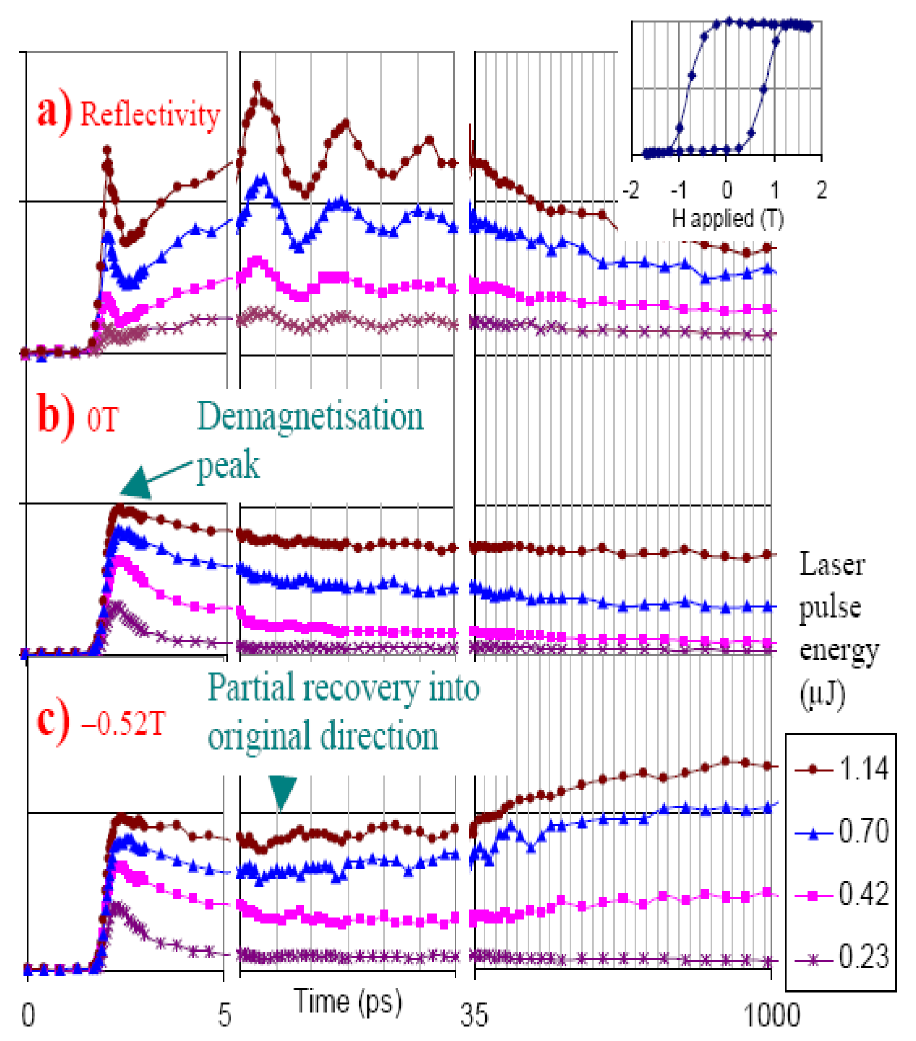
- This process is now possible for FePt
- Can be applied to other materials
- Final factor – does micromagnetic exchange really scale with $M^2$?

# 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

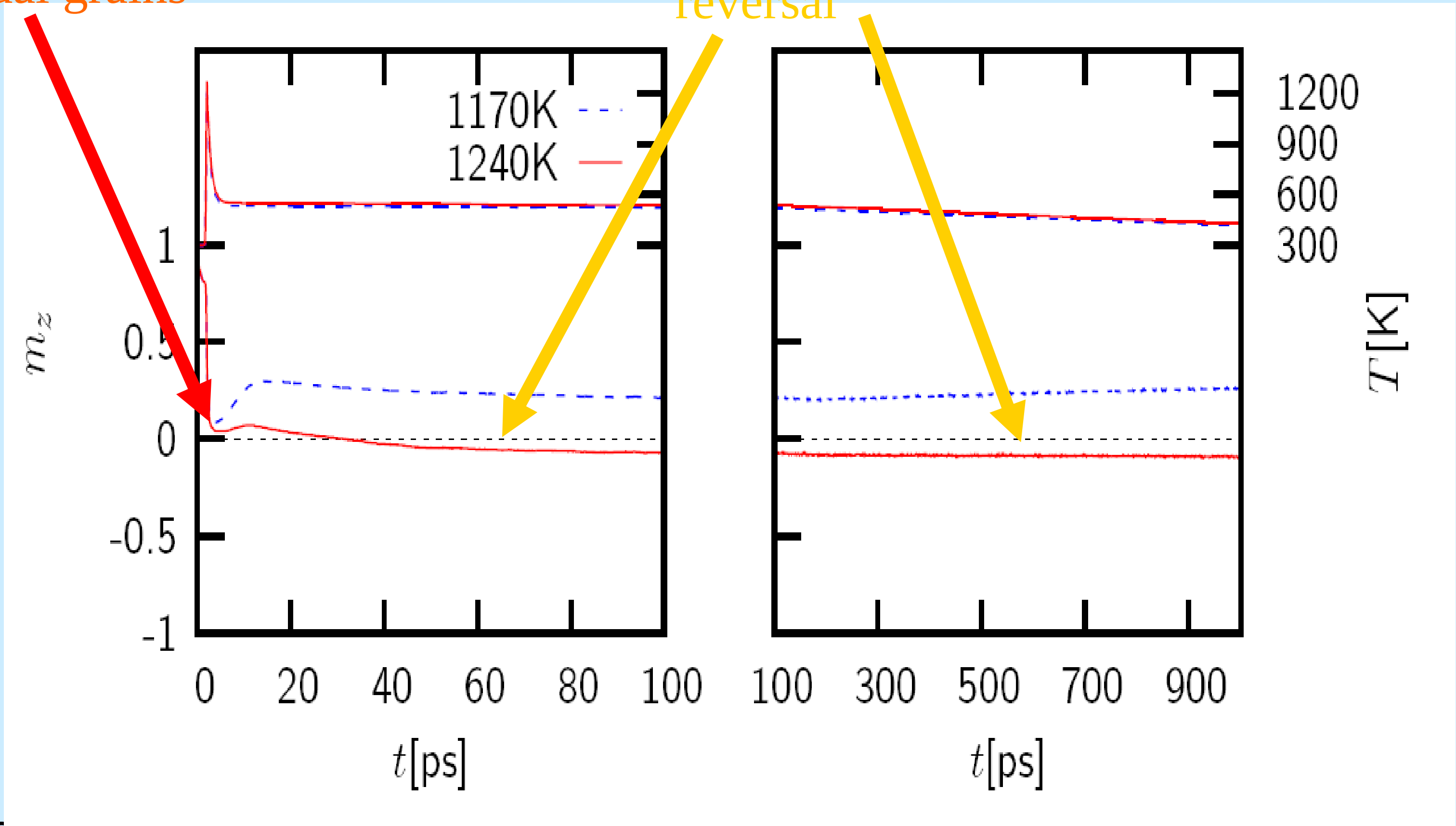


- Reversal occurs in a field of 0.52T (<< intrinsic coercivity of 1.4T
- Note 2 timescales. Associated with Longitudinal (initial fast reduction of M) and transverse (long timescale reversal over particle energy barriers) relaxation

# 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

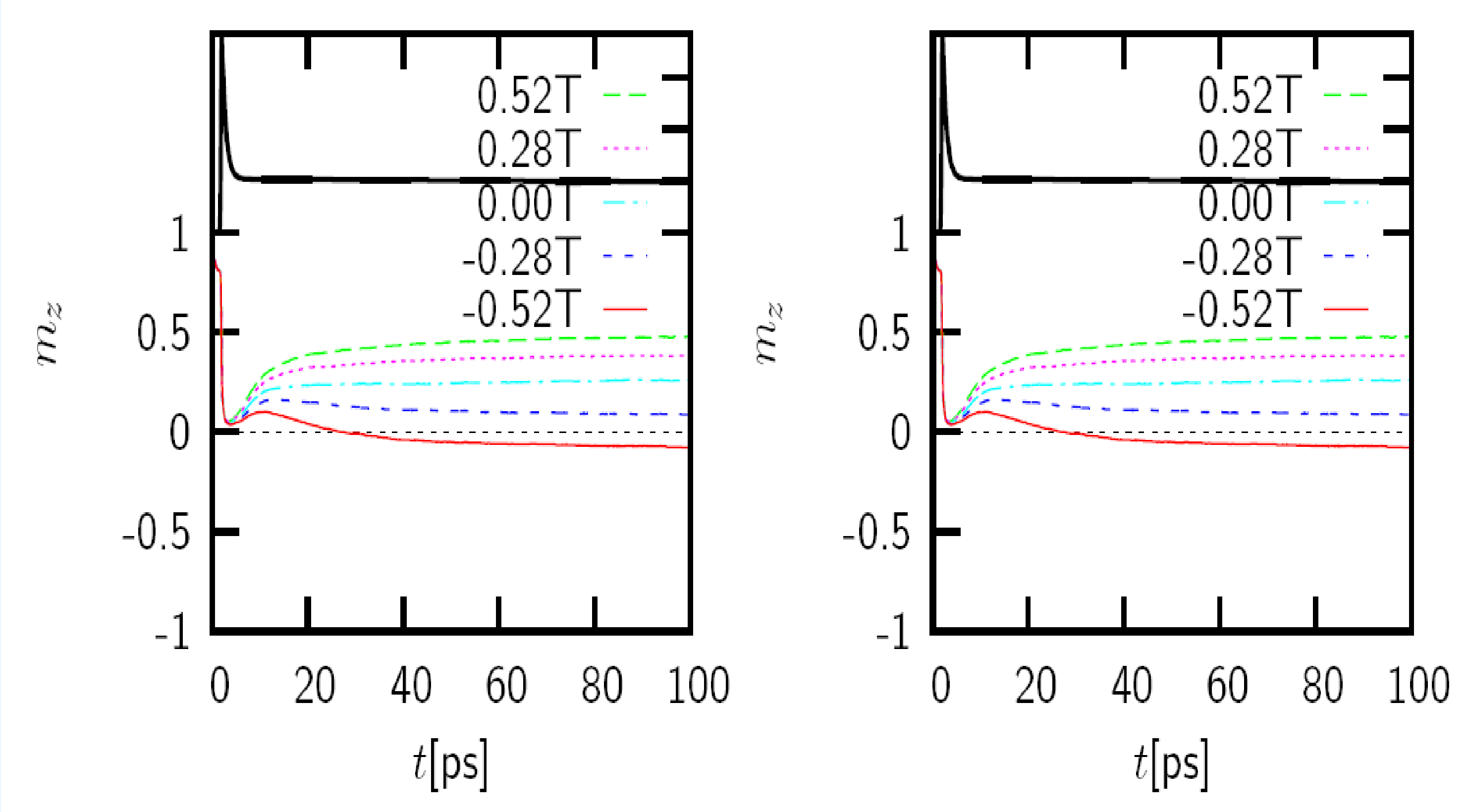


Demagnetisation/recovery of the magnetisation of individual grains

Superparamagnetic reversal

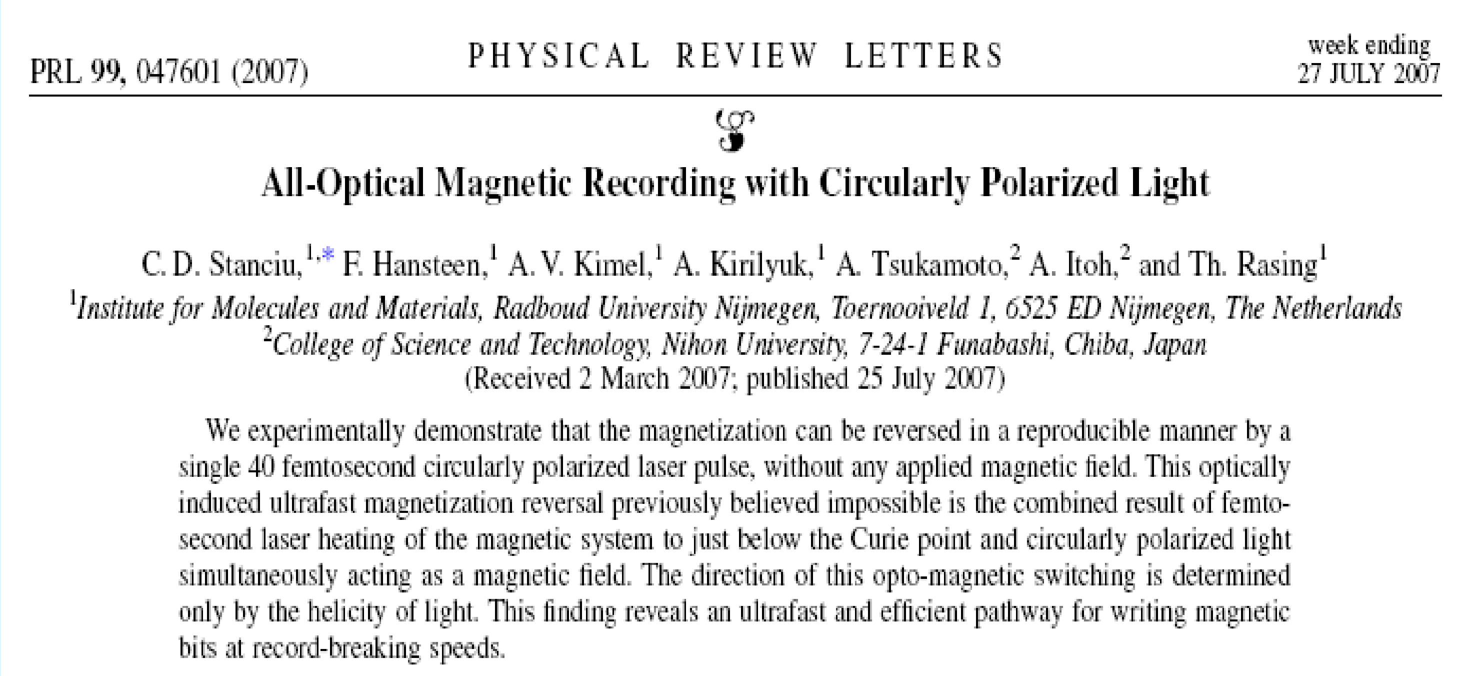
- Simulations show rapid demagnetisation followed by recovery on the short timescale. Over longer times the magnetisastion rotates into the field direction due to thermally activated transitions over energy barriers.
- This is consistent with experimental results

# Effect of the magnetic field



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

# Opto-magnetic reversal revisited



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

### All-Optical Magnetic Recording with Circularly Polarized Light

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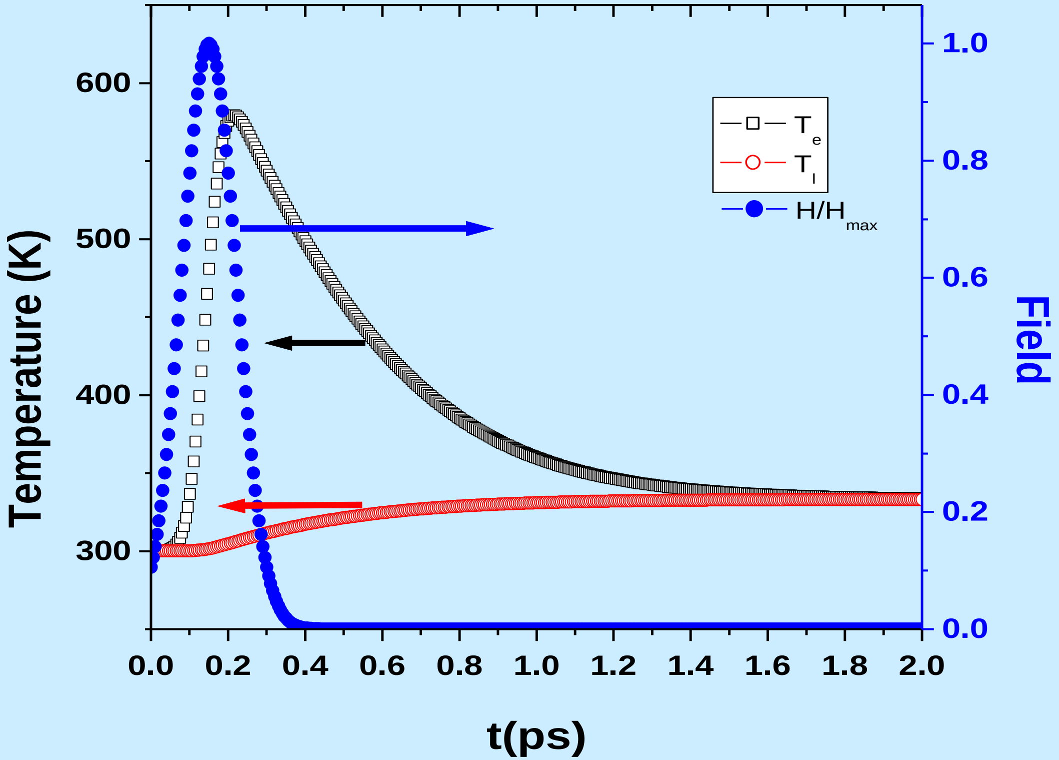
(Received 2 March 2007; published 25 July 2007)

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 $T_c$

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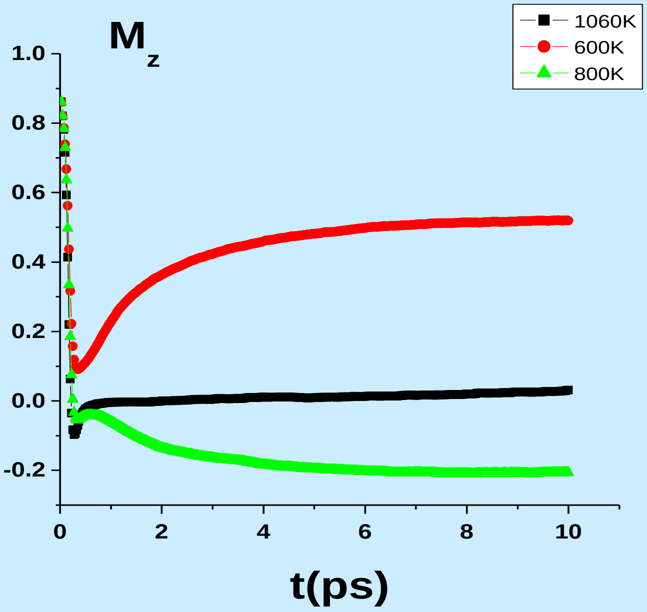
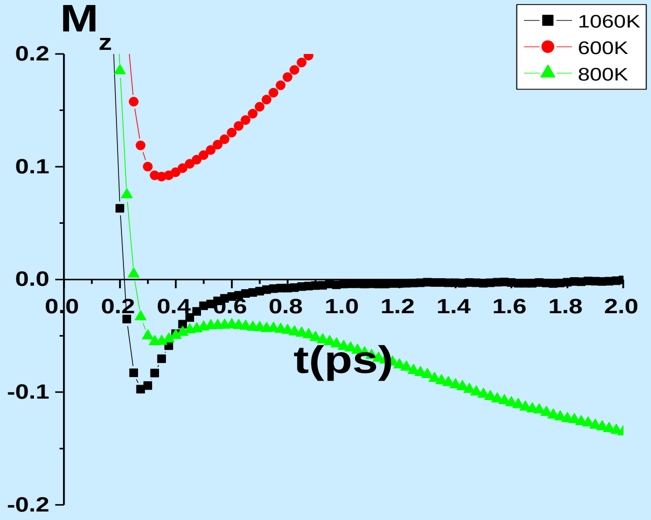
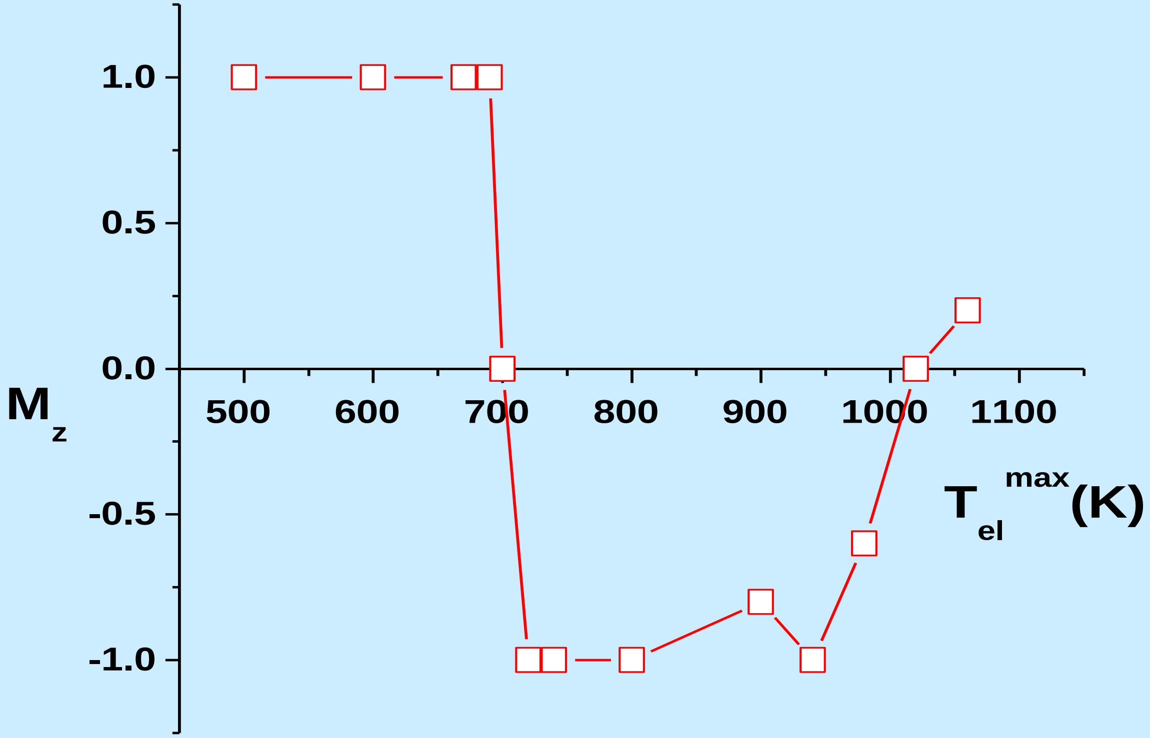
# Magnetisation dynamics (atomistic model)





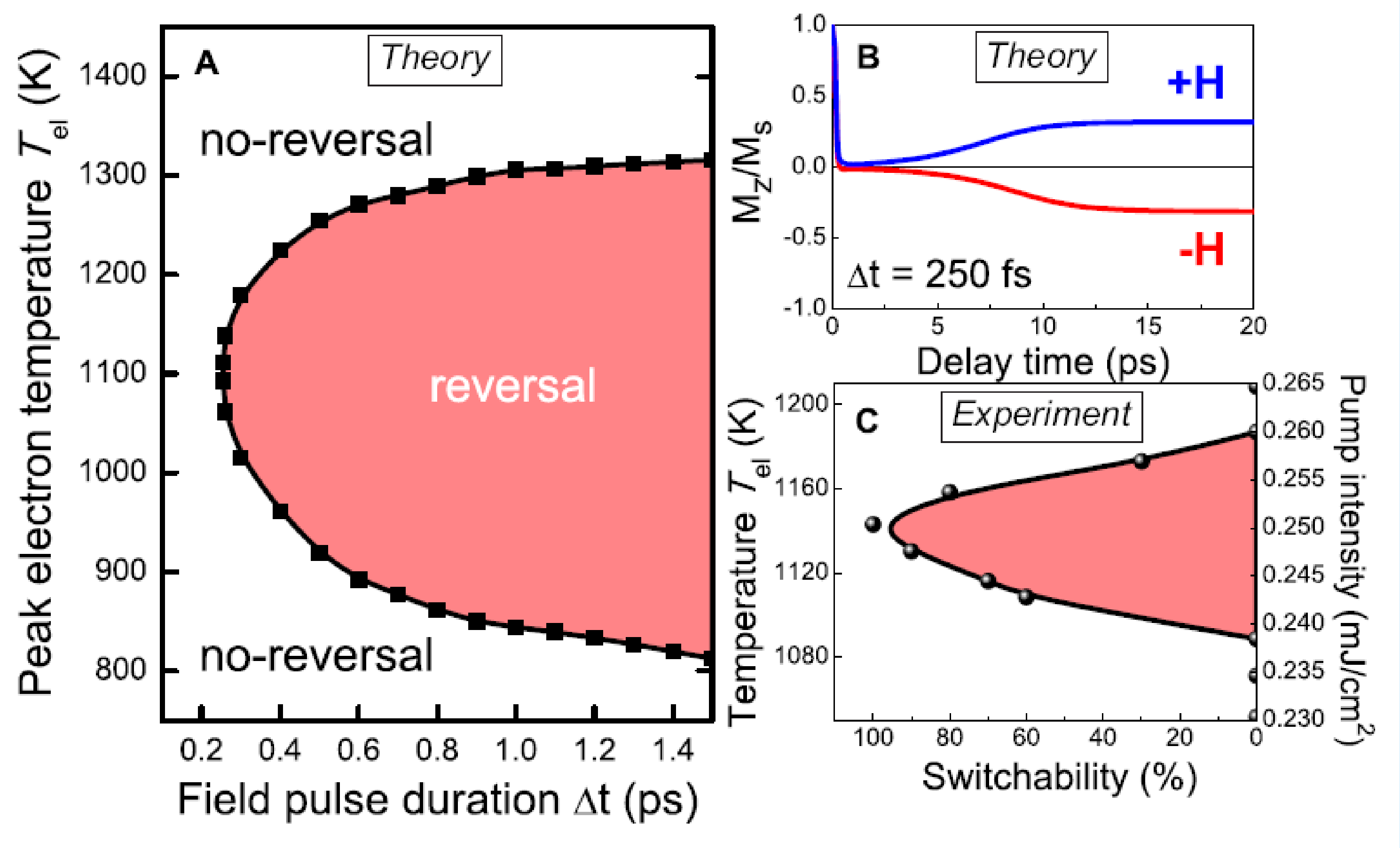
- ***Reversal is non-precessional*** – $m_x$ and $m_y$ remain zero. ***Linear reversal mechanism***
- Associated with increased magnetic susceptibility at high temperatures
- Too much laser power and the magnetisation is destroyed after reversal
- Narrow window for reversal

# ‘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., <u>103</u>, 117201 (2009)



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

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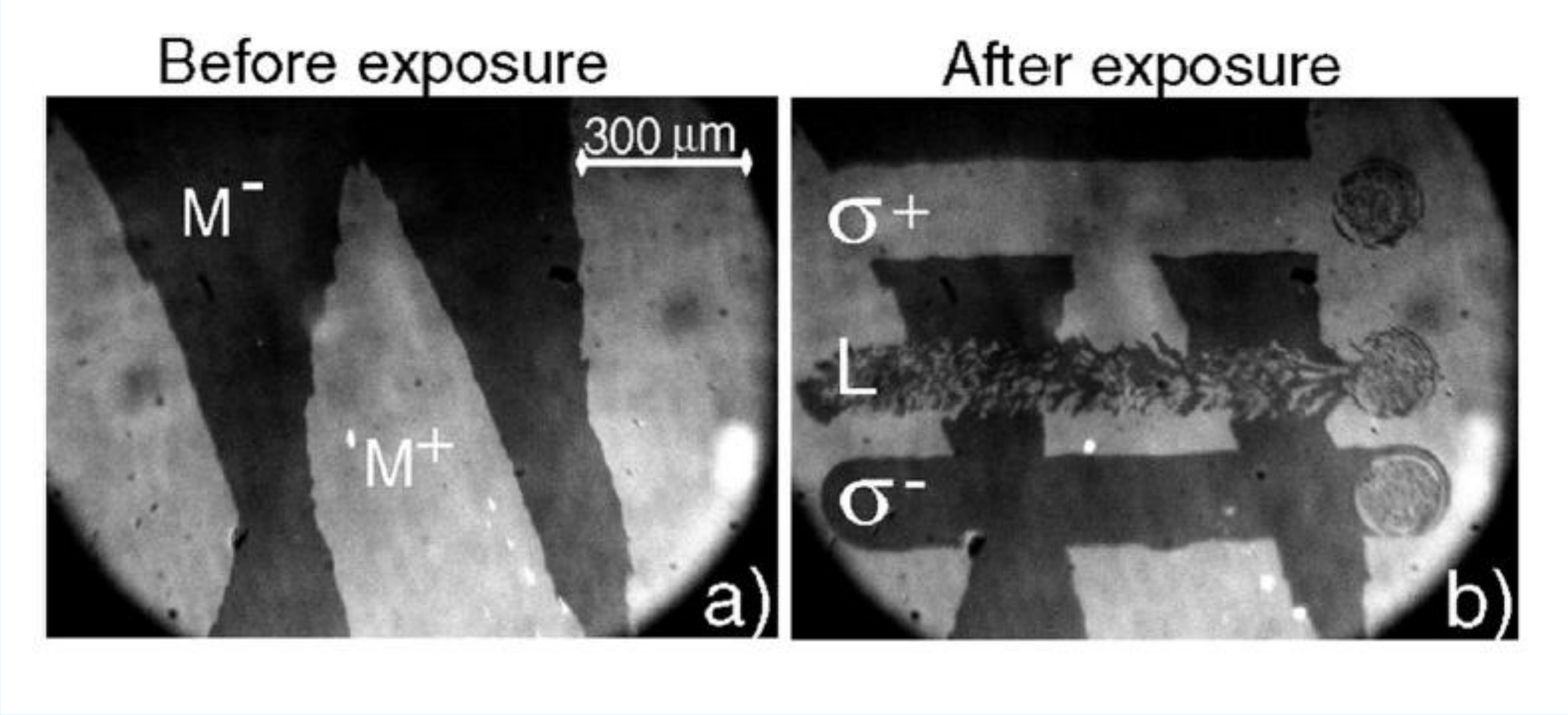
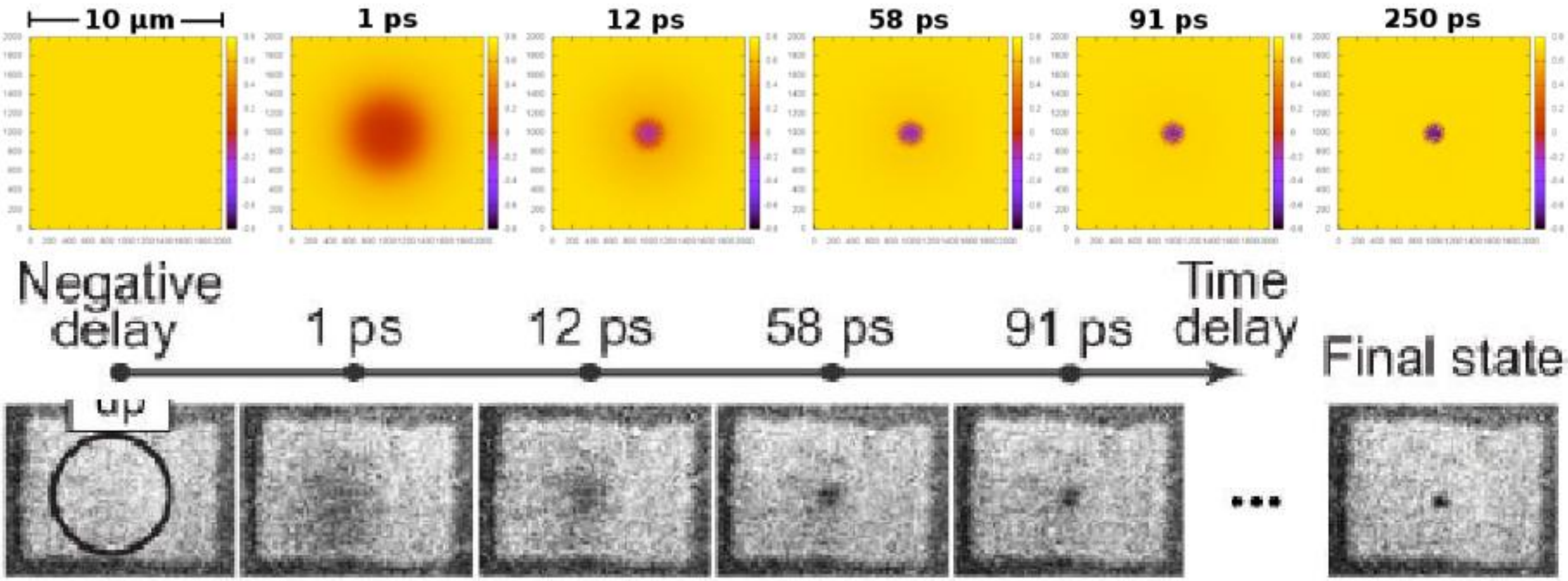
# Further evidence



# Large scale simulations (LLB micromagnetics)



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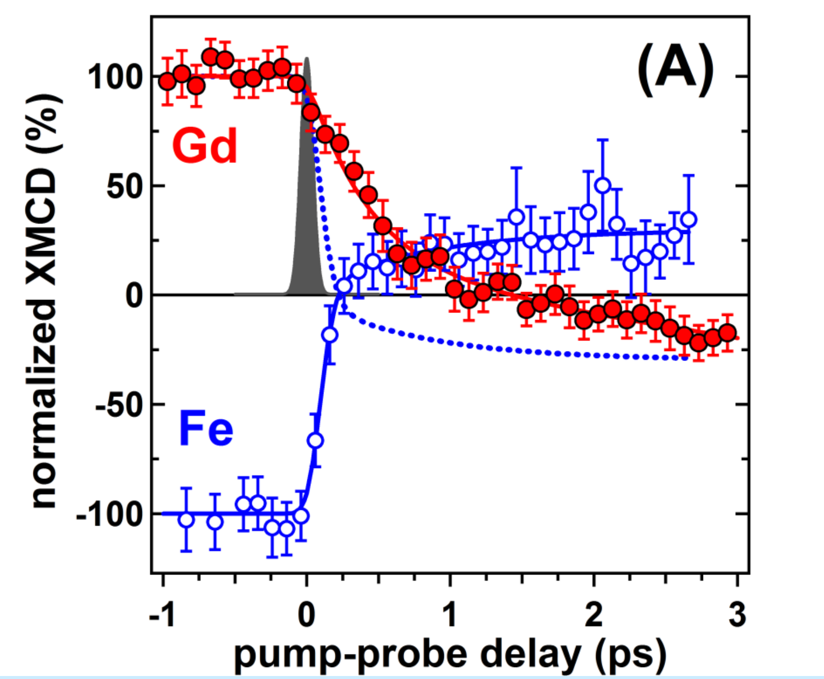
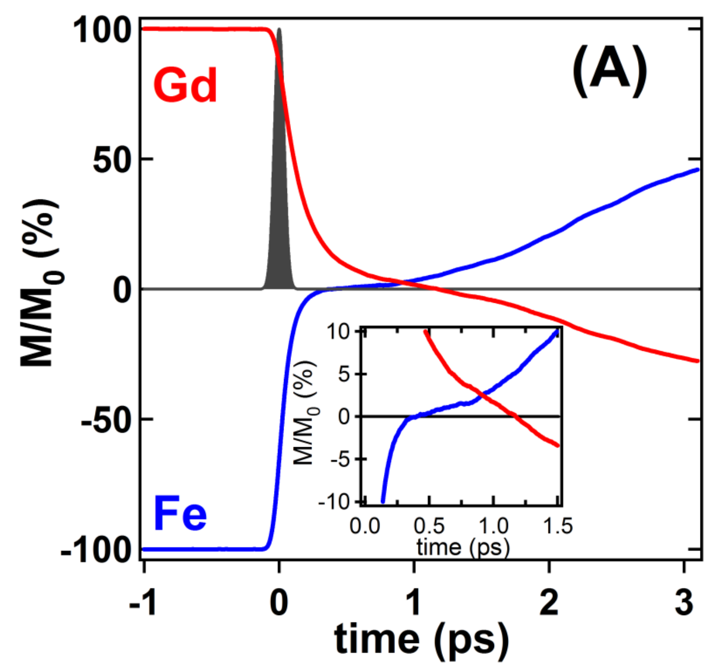
# Linear reversal in GdFeCo
# (I. Radu et. al., Nature, 472, 205 (2011))





- Experiments (left) in good agreement with atomistic model calculations (right)

# Differential sublattice demagnetisation

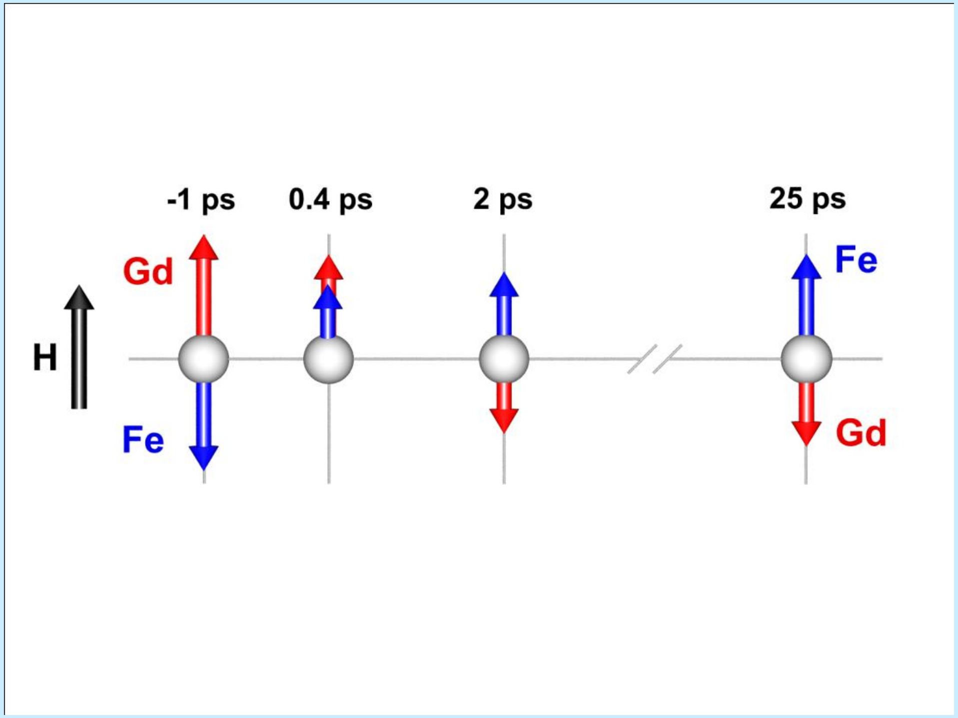
- TM and RE sublattices demagnetise at different rates *irrespective of the exchange interaction*.
- According to N. Kazantseva et. al., Europhys. Lett., 81, 27004 (2008), the demagnetisation time

$$\tau_D = \mu_s / \alpha$$

where $\mu_s$ is the atomic spin and $\alpha$ is the damping constant

- Consistent with experiments

# BUT



- Sublattices do not act independently
- Remarkable transient FM state produced for about 400fs!
- Seems to drive magnetisation 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!
- Finally, a problem which has received limited attention ……………………….

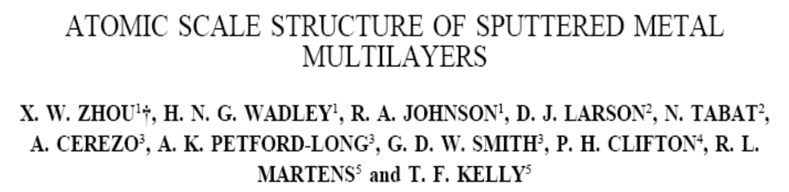
# Conclusions

- For many nanostructured magnetic systems micromagnetics has serious limitations.
  - Temperature dependence of the magnetic properties is not correctly predicted – cannot correctly deal with HAMR
  - Problems can occur at interfaces
- Solution is multiscale atomistic modelling, coupling electronic, atomistic and micromagnetic lengthscales. We distinguish 2 approaches
  - Scaling approaches – correctly scale M(T), K(T), A(T) within micromagnetics.
  - Multiscale approach – partitioning of material into atomistic and micromagnetic regions.
- Atomistic model has been developed using Heisenberg exchange.
- Soft/hard composite materials show a failure of micromagnetics to correctly predict the coercivity reduction at low interface coupling.
- 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.

# Future developments

- Micromagnetics will continue as the formalism of choice for large scale simulations
- However, multiscale calculations will become increasingly necessary as magnetic materials become more nanostructured
- 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?)

# Interfaces



ATOMIC SCALE STRUCTURE OF SPUTTERED METAL MULTILAYERS

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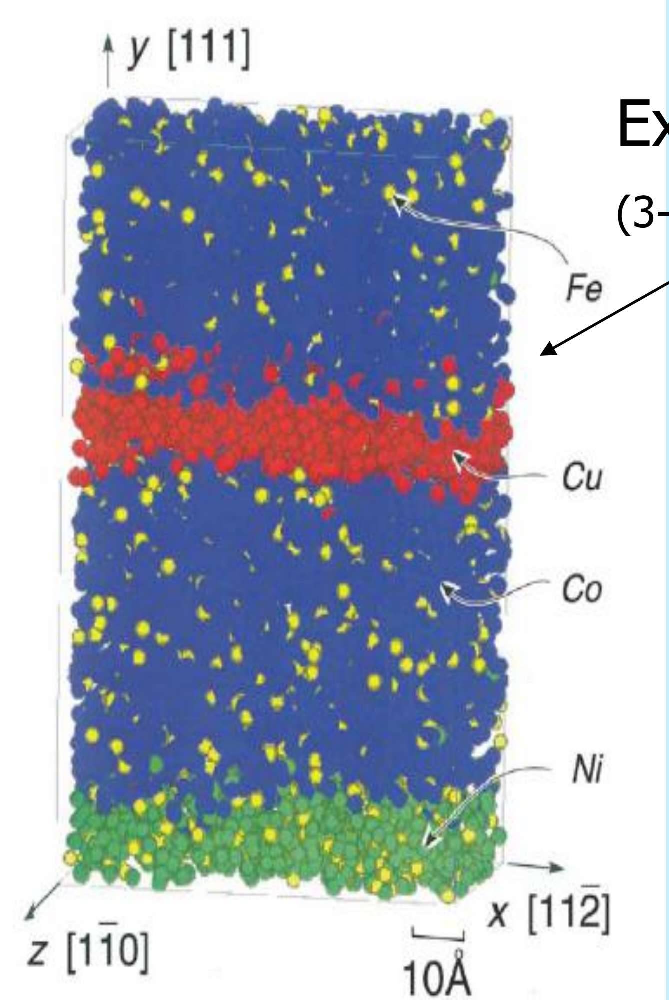
Acta mater. 49 (2001) 4005–4015

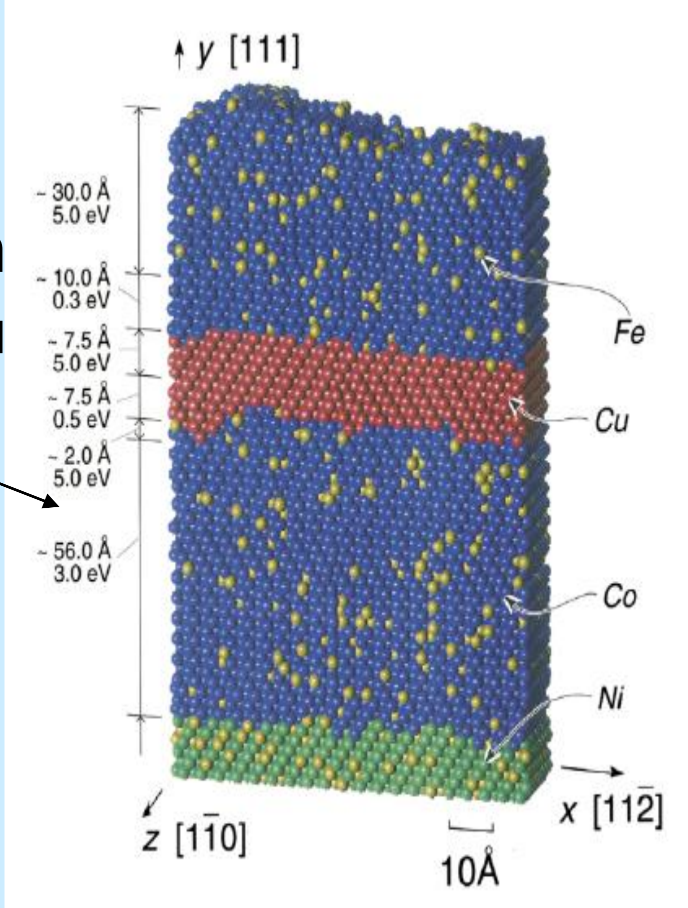


Experiment

(3-D atom probe)

Simulation

MD+Embedded atom potential



# What's next?

- 1948 Stoner-Wohlfarth (Brunsviga mechanical calculator)
- 1980s
  - Early - local mainframe computers
  - Late – Cray YMP
- 1990s teraflop machines
- 2000s
  - local teraflop machines
  - Petaflop supercomputers
- 2010 onwards – ENJOY!