Advanced micromagnetics and atomistic simulations of magnets

Richard F L Evans

ESM 2024

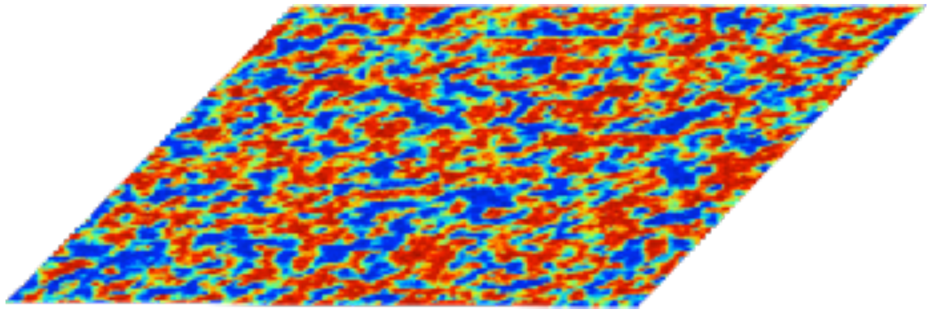
Overview

- Landau-Lifshitz-Bloch micromagnetics

- Applications of atomistic spin dynamics: Non-collinear antiferromagnets and their temperature dependent anisotropy

- 2D magnetism and the Mermin-Wagner theorem

- Simulations of ultrafast magnetisation processes



Landau Lifshitz Bloch micromagnetics

Next generation micromagnetics: Landau Lifshitz Bloch equation

- Conventional micromagnetics ubiquitous but does a poor job of thermodynamics of magnetic materials

- Atomistic models in principle resolve this but horrendously computationally expensive

- **Landau Lifshitz-Bloch micromagnetics** is an advanced micromagnetic approach which attempts to correctly simulate the intrinsic thermodynamic properties of magnets

- Still only a partial solution - crystal structure, interfaces, surfaces, local defects, finite size effects all still not really accessible to a micromagnetic model

Landau Lifshitz Bloch (LLB) equation

- An additional dynamic term compared to the LLG equation

$$
\begin{aligned}
\dot{\mathbf{m}} = \gamma[\mathbf{m} \times \mathbf{H}_{\text{eff}}] + \frac{|\gamma|\alpha_{||}}{m^2}(\mathbf{m} \cdot \mathbf{H}_{\text{eff}})\mathbf{m} \\
- \frac{|\gamma|\alpha_{\perp}}{m^2}[\mathbf{m} \times [\mathbf{m} \times (\mathbf{H}_{\text{eff}} + \boldsymbol{\eta}_{\perp})]] + \boldsymbol{\eta}_{||}
\end{aligned}
$$

- Derived from the thermodynamic behaviour of a collection of classical spins by D. Garanin [1]

- Longitudinal fluctuations (and damping) of the magnetization are now included in the dynamics, enabling simulations up to and above the Curie temperature

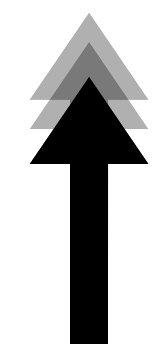
- Also quantum flavours of the LLB

[1] D. A. Garanin, Phys. Rev. B 55, 3050 (1997)

Longitudinal term in the Landau Lifshitz Bloch (LLB) equation

- Longitudinal fluctuations of the magnetization have their own dynamics

$$\frac{|\gamma|\alpha_{||}}{m^2}(\mathbf{m}\cdot\mathbf{H}_{\mathrm{eff}})\mathbf{m}$$



- Different effects below and above the Curie temperature, T_c

- The effective magnetic field that constrains the magnetization length is given by

$$\mathbf{H}_{\mathrm{eff}} = \mathbf{H} + \mathbf{H}_A + \begin{cases} \frac{1}{2\tilde{\chi}_{\|}}\left(1 - \frac{m^2}{m_{\mathrm{e}}^2}\right)\mathbf{m}, & T \lesssim T_{\mathrm{c}} \\ -\frac{1}{\tilde{\chi}_{\|}}\left(1 + \frac{3}{5}\frac{T_{\mathrm{c}}}{T-T_{\mathrm{c}}}m^2\right)\mathbf{m}, & T \gtrsim T_{\mathrm{c}} \end{cases}$$

Energy terms in the Landau Lifshitz Bloch (LLB) equation

- Conventional energy terms used in micromagnetics cause numerical issues for the LLB, as any “applied” magnetic field will cause the moment length to grow

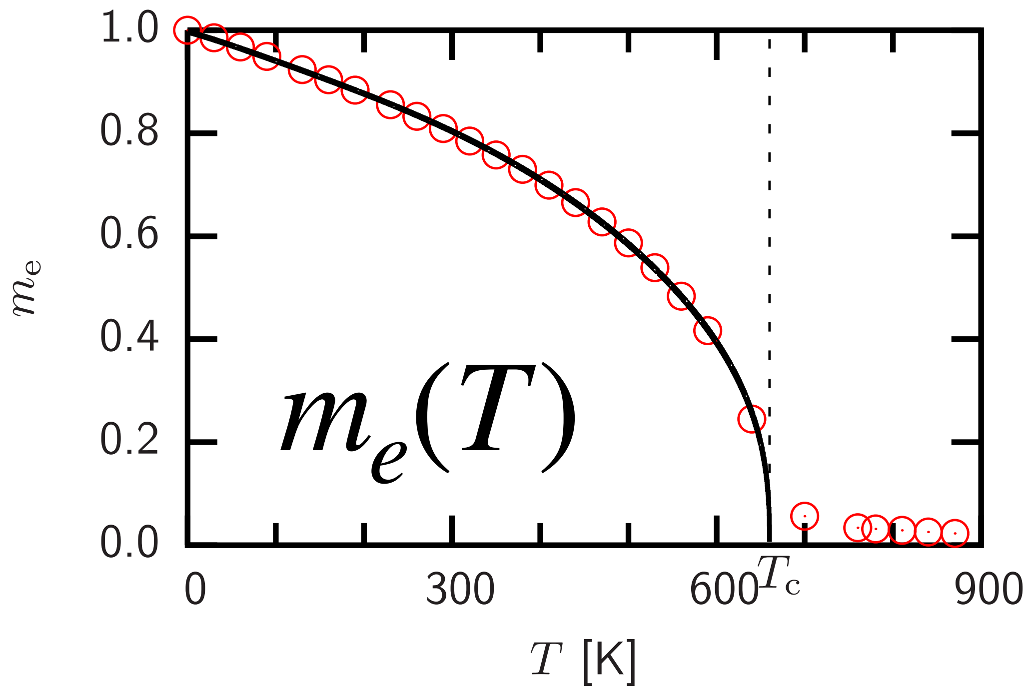
$$\frac{|\gamma|\alpha_{||}}{m^2}(\mathbf{m}\cdot\mathbf{H}_{\mathrm{eff}})\mathbf{m}$$

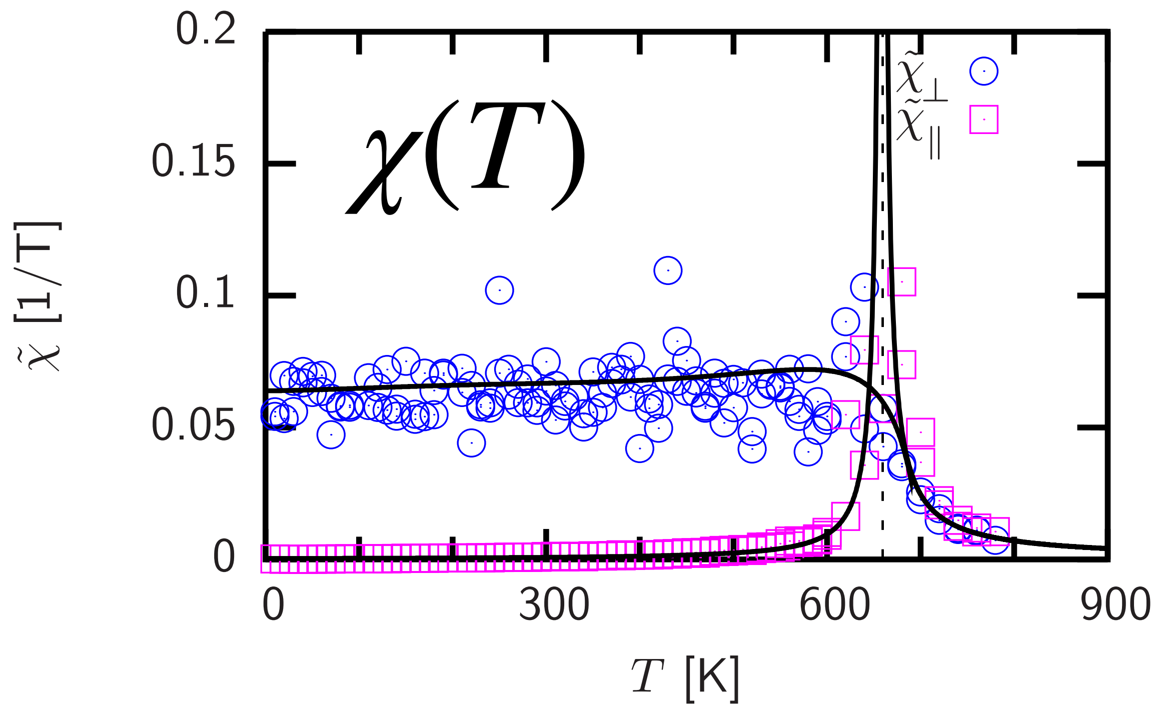
- Therefore need to treat internal fields in a special way so that in **thermal equilibrium**, the net magnetic field is **zero**

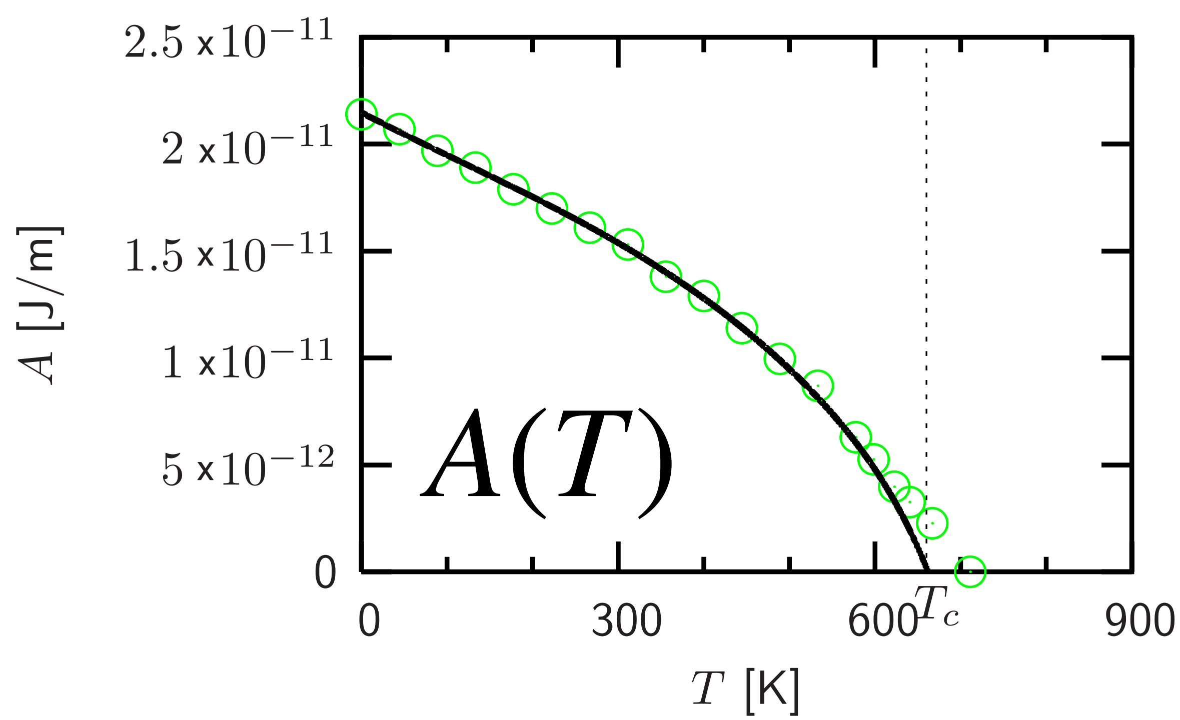
$$\frac{F}{M_{\mathrm{s}}^0 V} = \begin{cases} \frac{m_x^2+m_y^2}{2\tilde{\chi}_\perp} + \frac{\left(m^2-m_{\mathrm{e}}^2\right)^2}{8\tilde{\chi}_\parallel m_{\mathrm{e}}^2}, & T \leqslant T_{\mathrm{c}} \\ \frac{m_x^2+m_y^2}{2\tilde{\chi}_\perp} + \frac{3}{20\tilde{\chi}_\parallel}\frac{T_{\mathrm{c}}}{T-T_{\mathrm{c}}}\left(m^2+\frac{5}{3}\frac{T-T_{\mathrm{c}}}{T_{\mathrm{c}}}\right)^2, & T > T_{\mathrm{c}} \end{cases}$$

Evans *et al*, Phys. Rev. B **85**, 014433 (2012)

Parameters for the LLB equation can be derived from mean field or atomistic/multiscale simulations

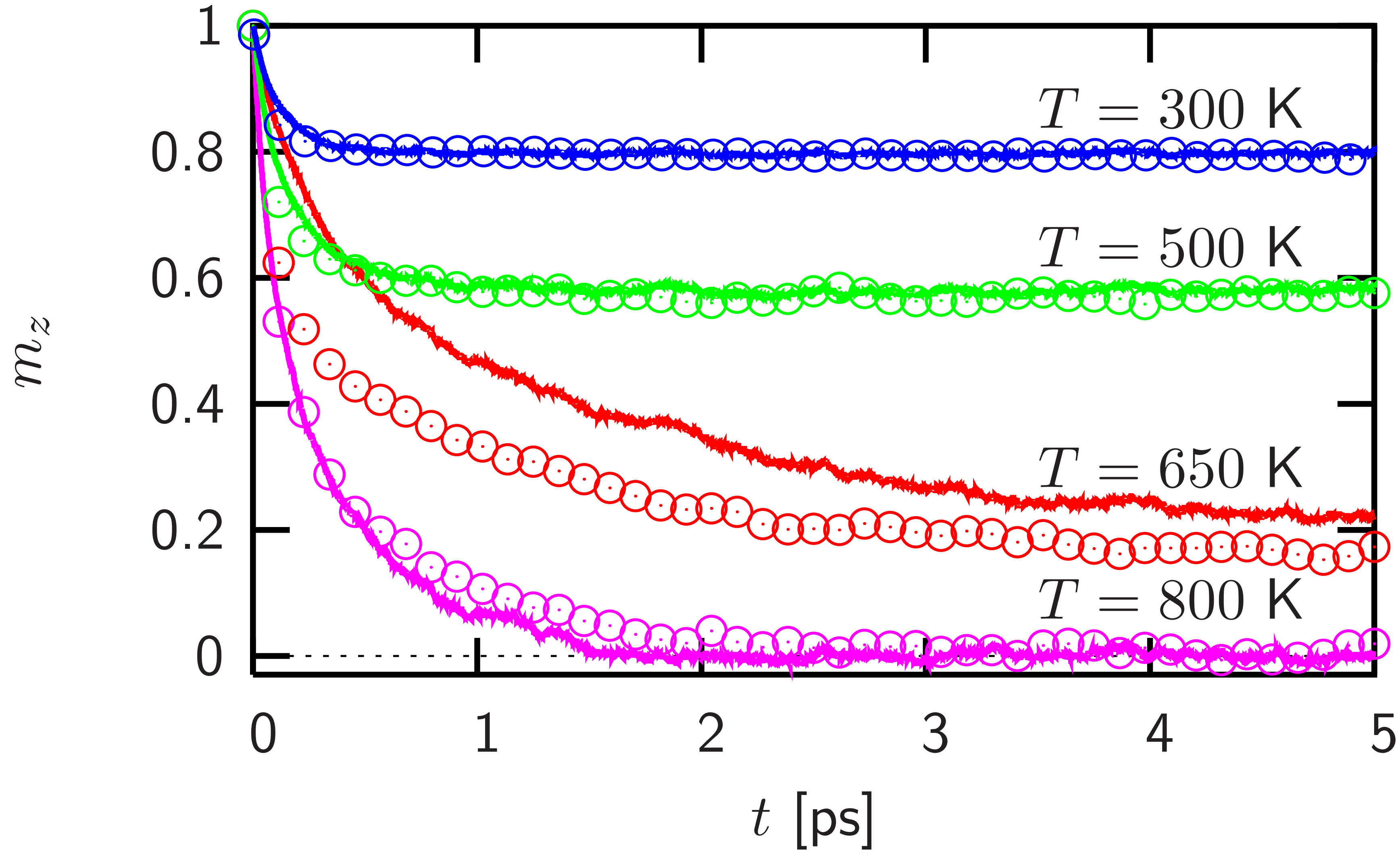






Kazantseva *et al*, Phys. Rev. B **77**, 184428 (2008)

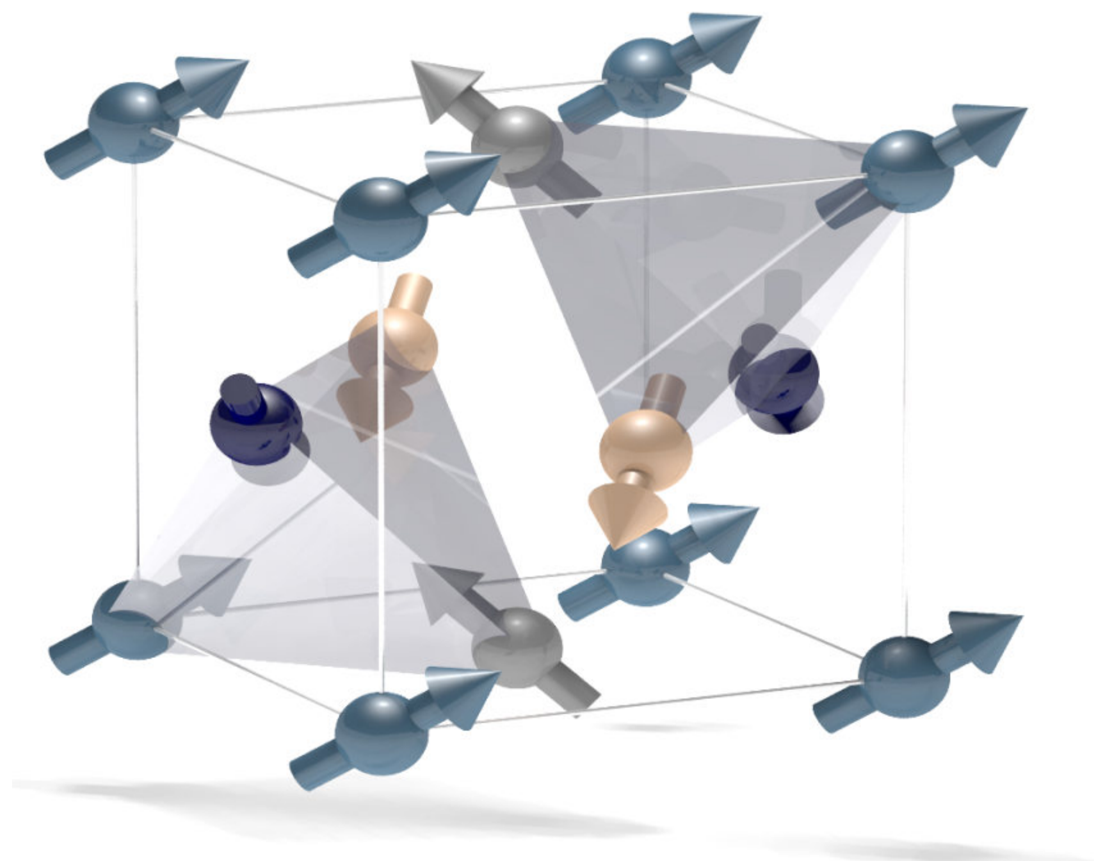
Comparative dynamics for LLB and atomistic simulations



Kazantseva *et al*, Phys. Rev. B **77**, 184428 (2008)

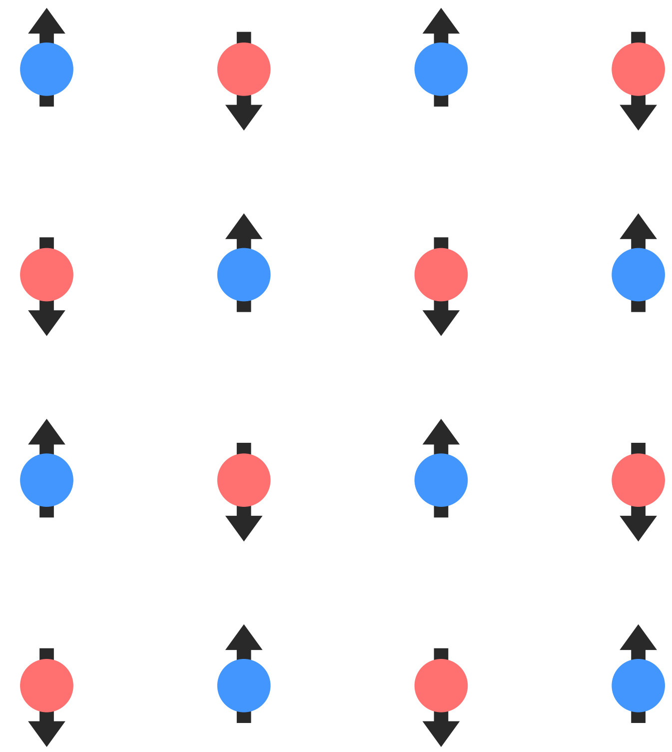
Applications of atomistic spin dynamics

Temperature dependent properties and dynamics of $IrMn_3$ antiferromagnets



Sarah Jenkins and Richard F L Evans

Simple antiferromagnets



- ‘Simple’ antiferromagnets consist of two magnetic sublattices

- Total magnetic moment is zero (macroscopically)

- Can consider two antiparallel contributions from each ‘colour’ of spin

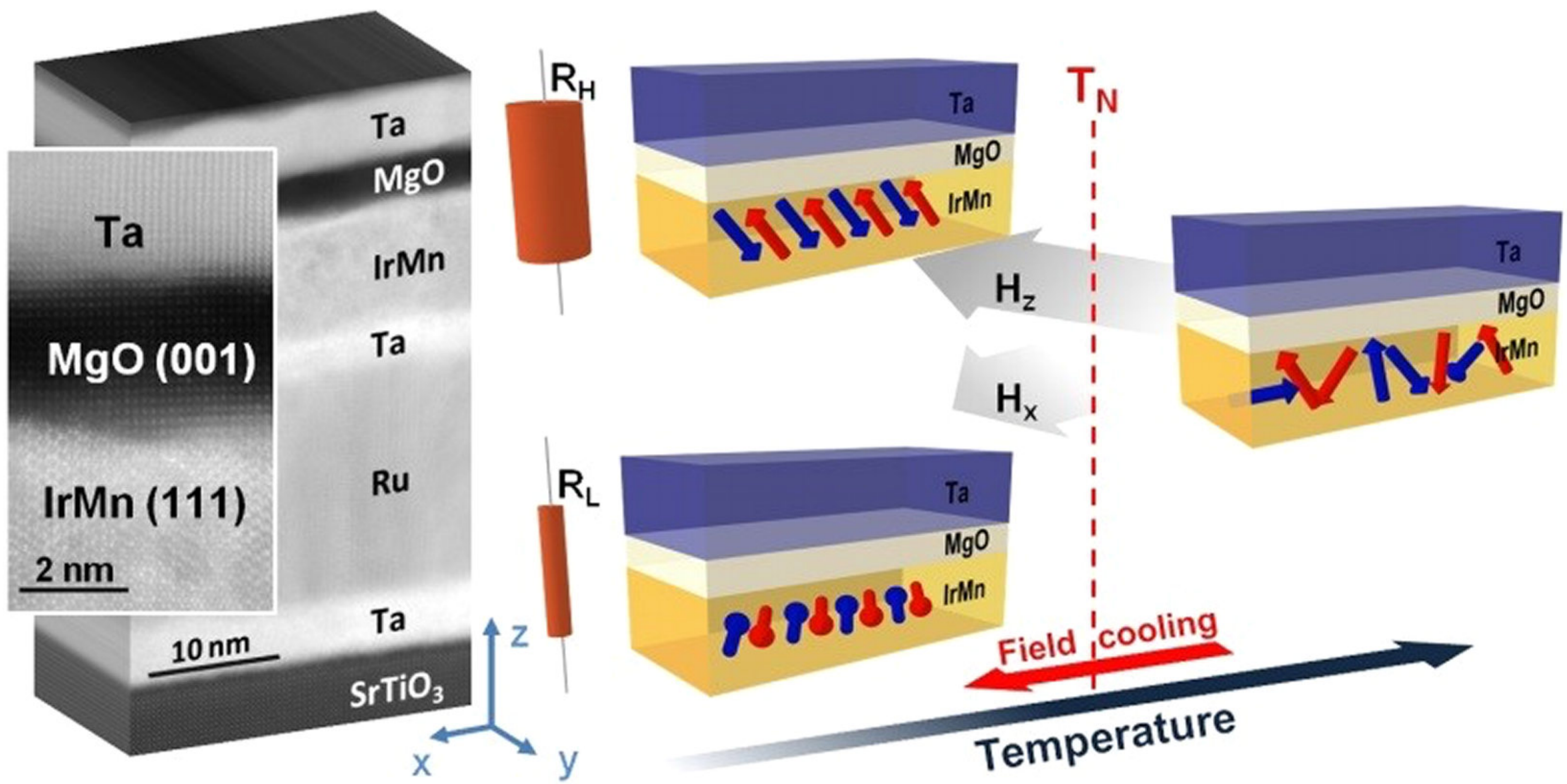
$$\mathbf{m}_a = \sum_a \mathbf{S}_a \qquad \mathbf{m}_b = \sum_b \mathbf{S}_b$$

- This is called the **sublattice magnetization**

- The Néel vector *n* is the equivalent order parameter for antiferromagnets

$$\mathbf{n} = \mathbf{m}_a - \mathbf{m}_b$$

Motivation: exchange bias and antiferromagnetic spintronics



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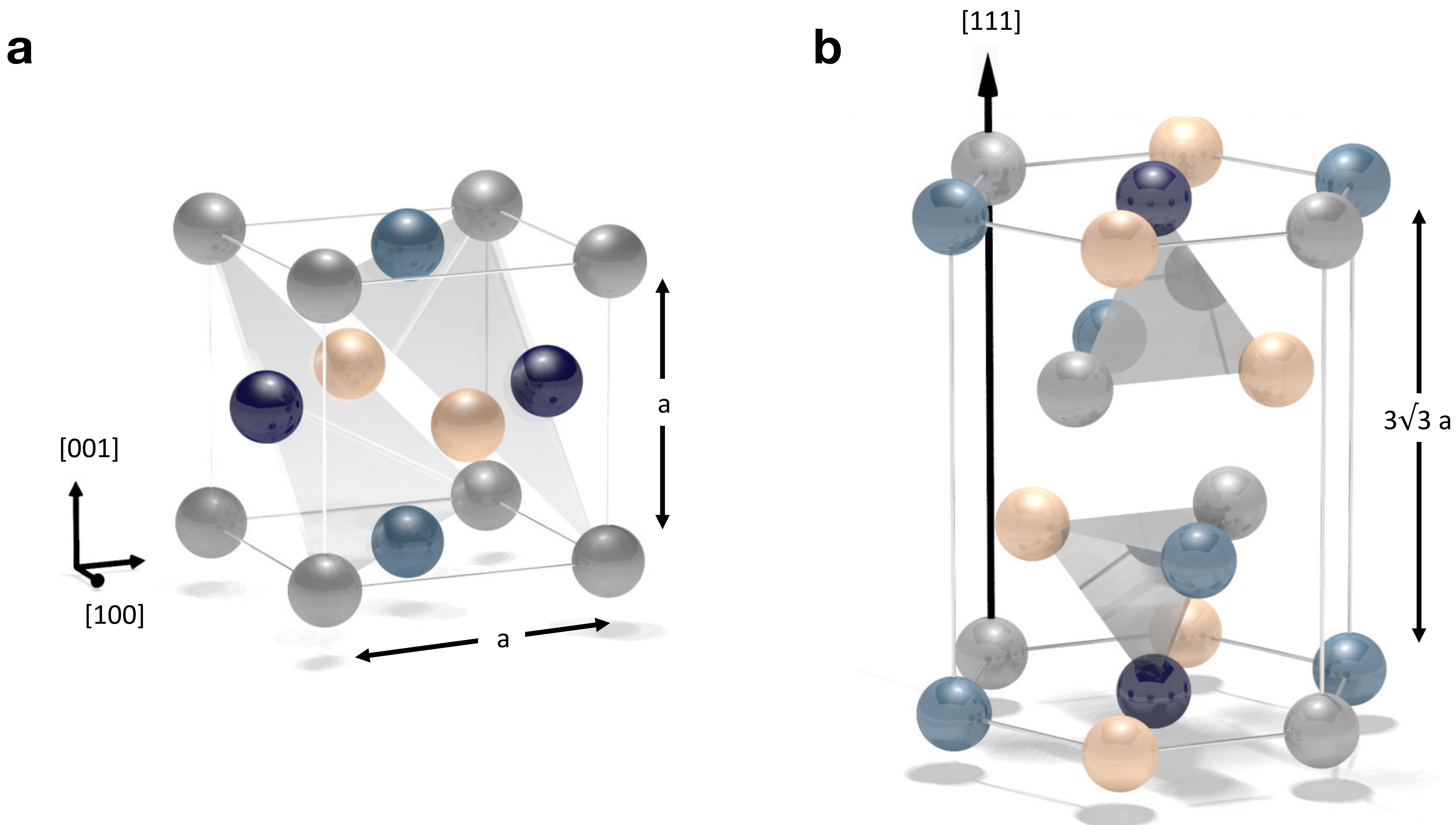
http://nabis.fisi.polimi.it/research-areas/antiferromagnet-spintronics/

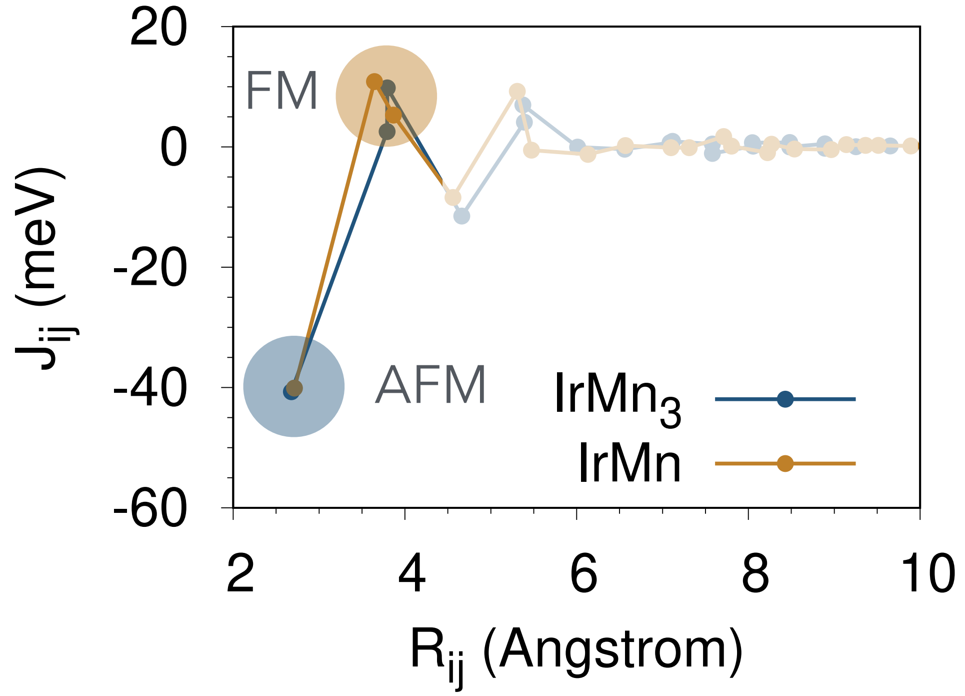
Crystal structures of IrMn



Atomistic spin model of IrMn

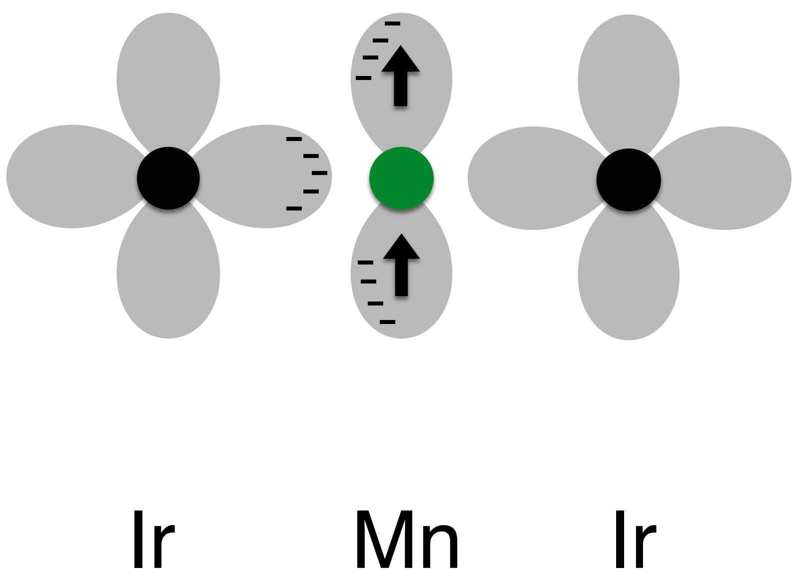
$$\mathscr{H} = -\sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_{i,j} \frac{k_{\mathrm{N}}}{2} \left(\mathbf{S}_i \cdot \mathbf{e}_{ij}\right)^2$$

J_{NN}-J_{NNN} model



Szunyogh et al, Phys. Rev. B ***79****, 020403 R (2009)*

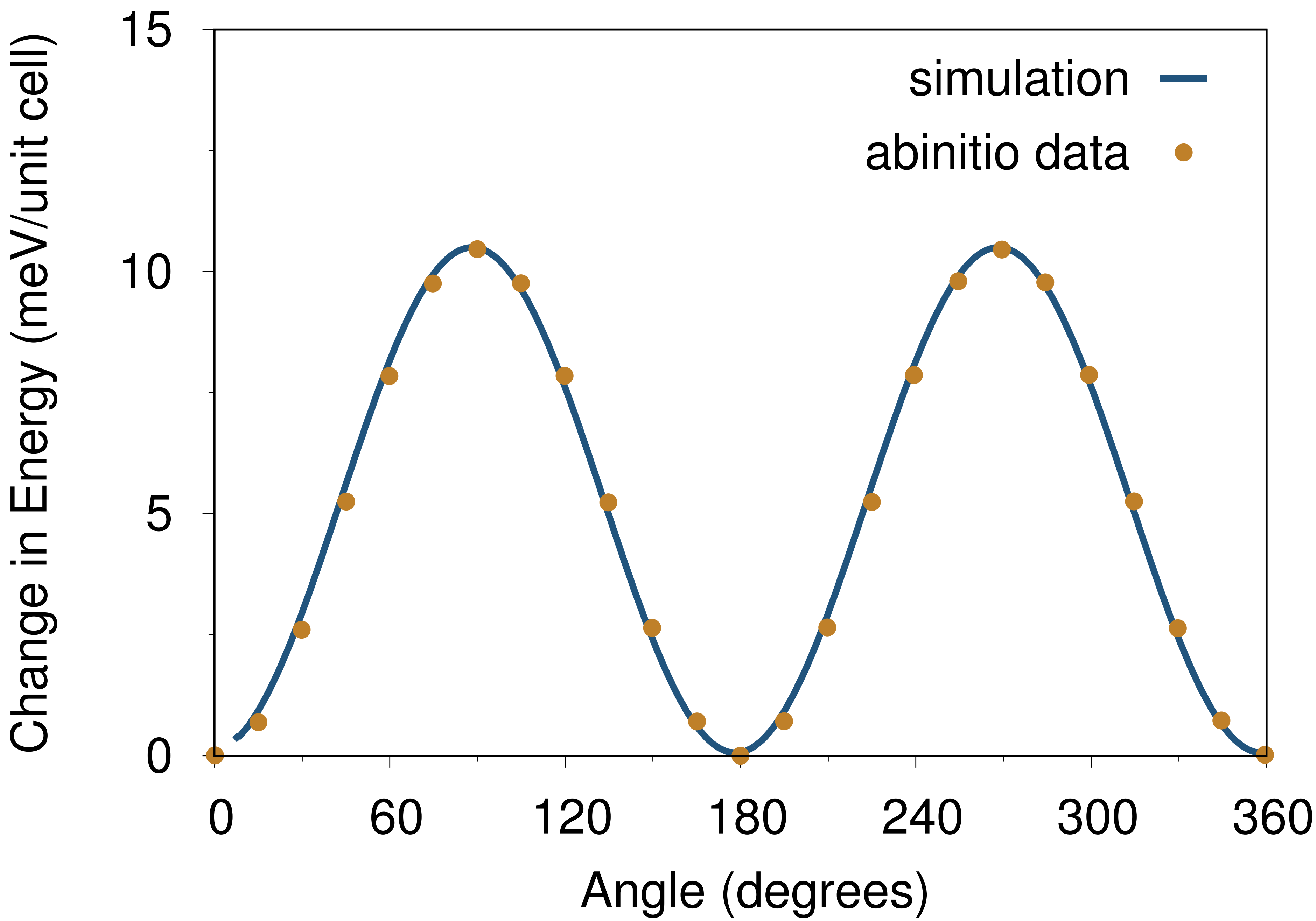
Néel pair anisotropy

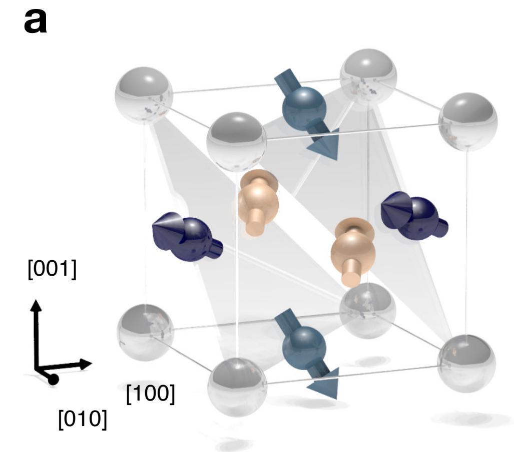


L. Néel, J. Phys. Radium ***15****, 376 (1954)*

S Jenkins et al, Journal of Applied Physics 124 152105 (2018)

Comparison of ab-initio data and the Neel pair anisotropy

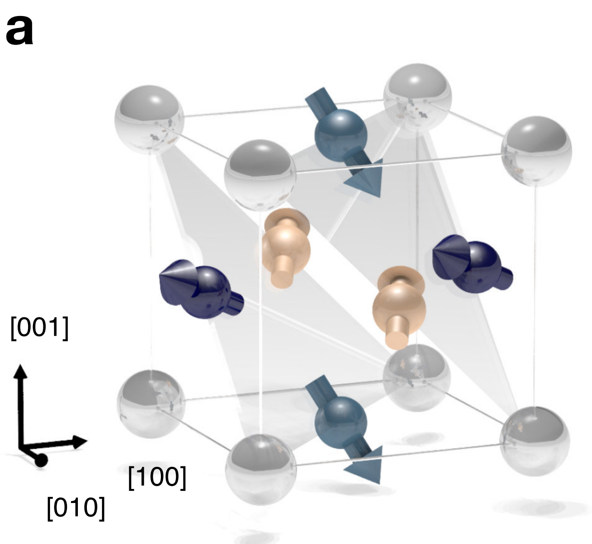




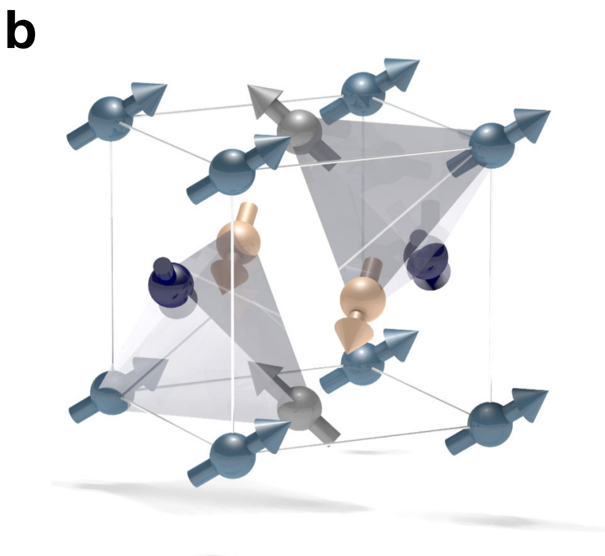
$L1_2$ (T1) phase

Szunyogh et al, Phys. Rev. B ***79****, 020403 R (2009)*

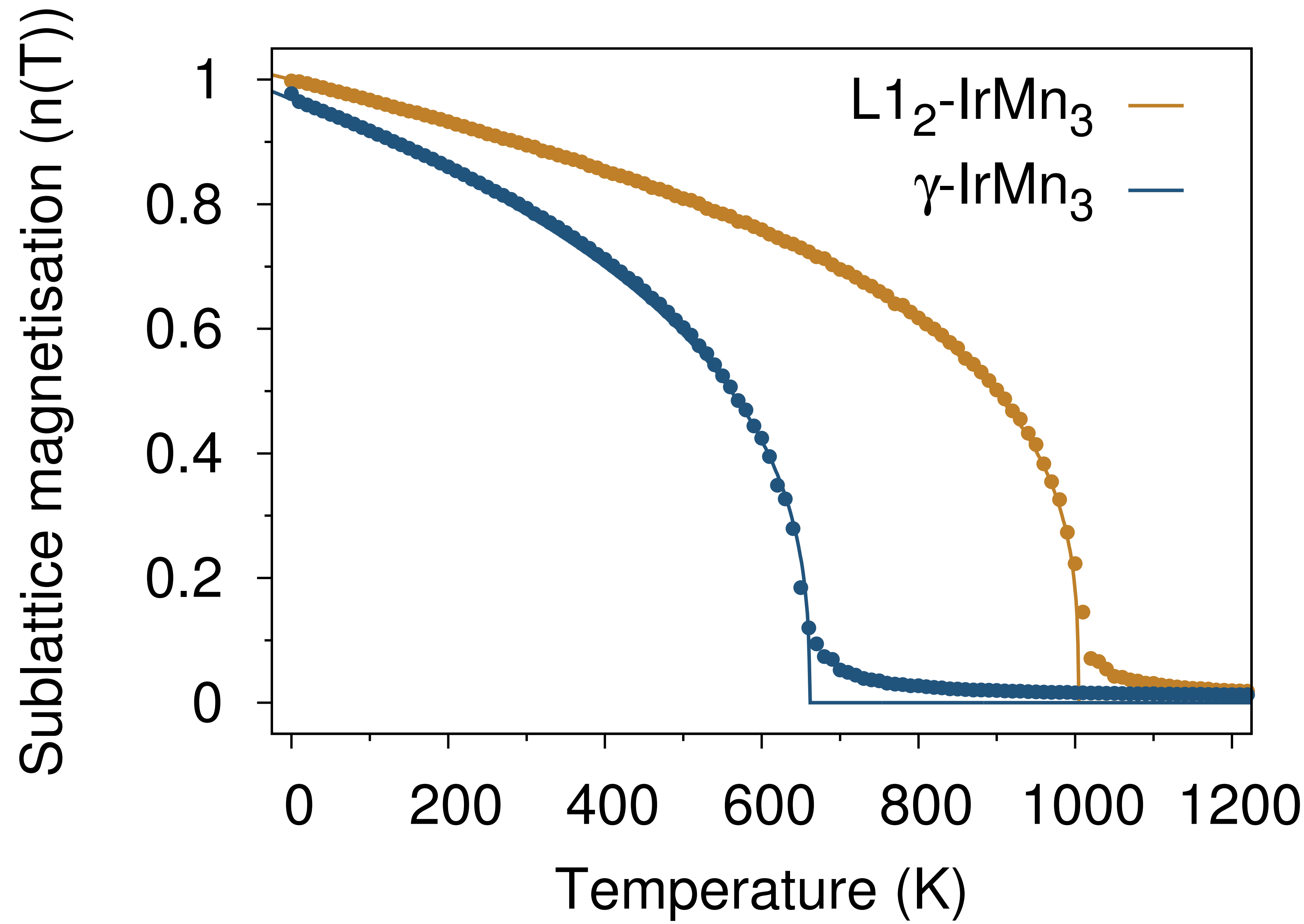
Magnetic structure and Néel temperatures of $IrMn_3$



Ordered $L1_2$–$IrMn_3$

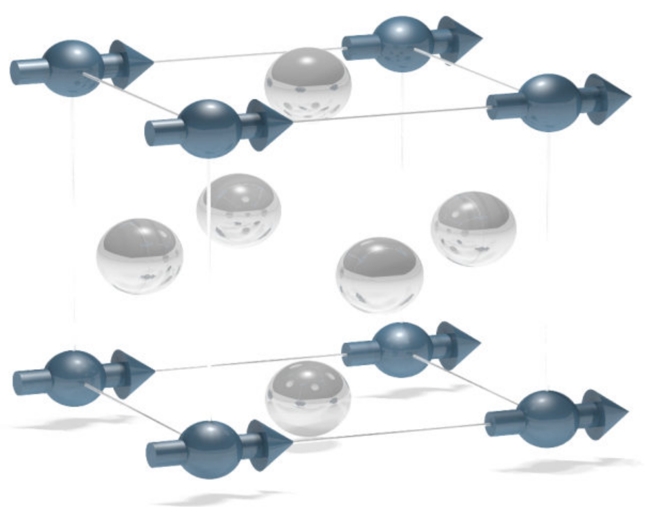


Disordered γ –$IrMn_3$

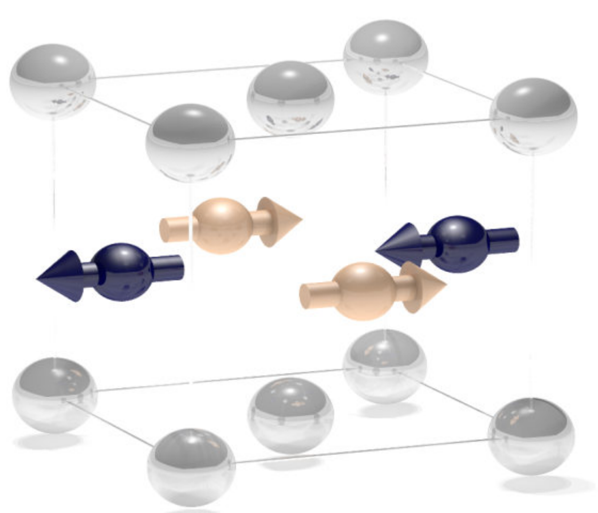


Sarah Jenkins *et al*, Physical Review Materials **5** 034406 (2021)

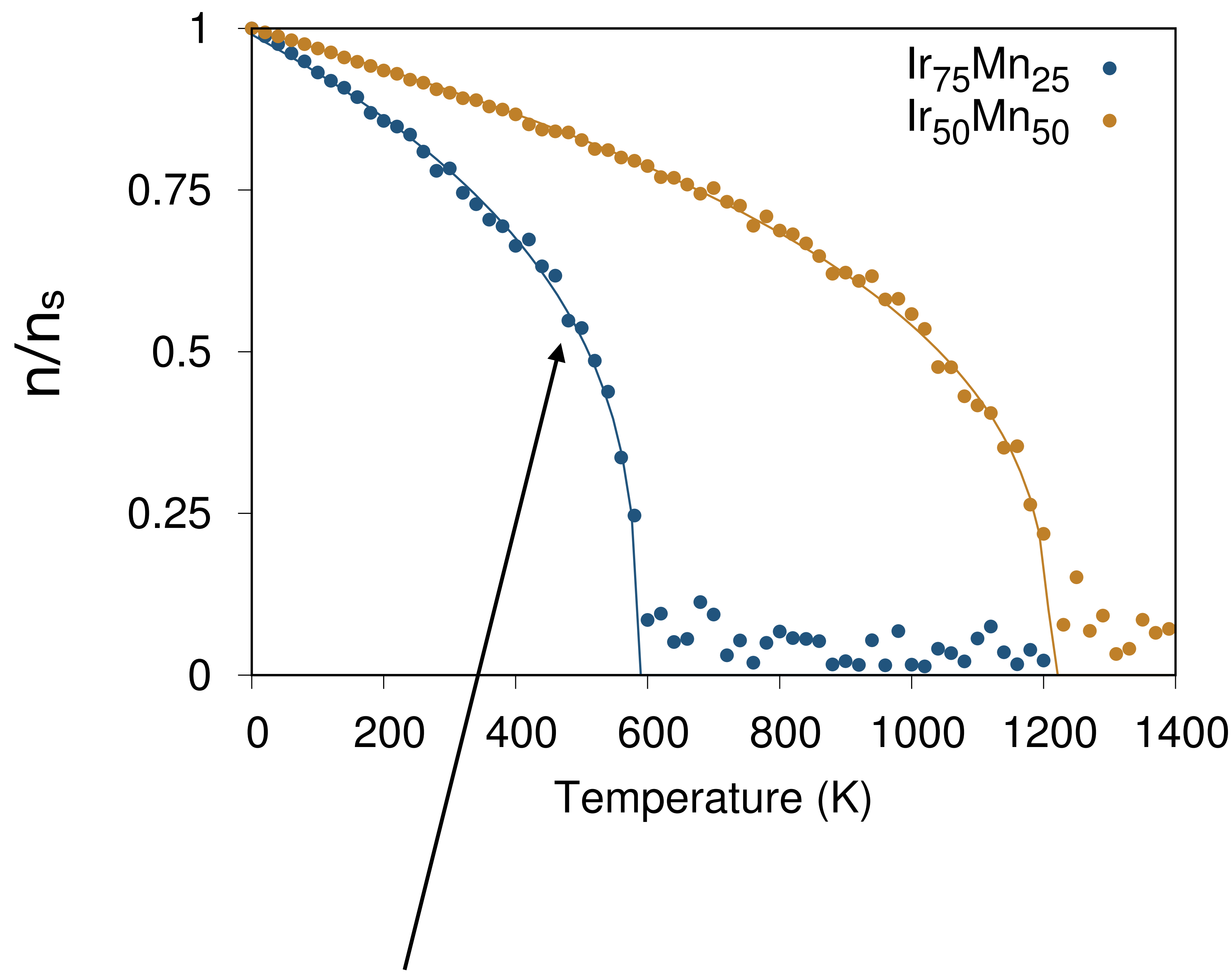
Magnetic structure and Néel temperatures of Ir_xMn_{1-x} alloys



Ordered $Ir_{75}Mn_{25}$

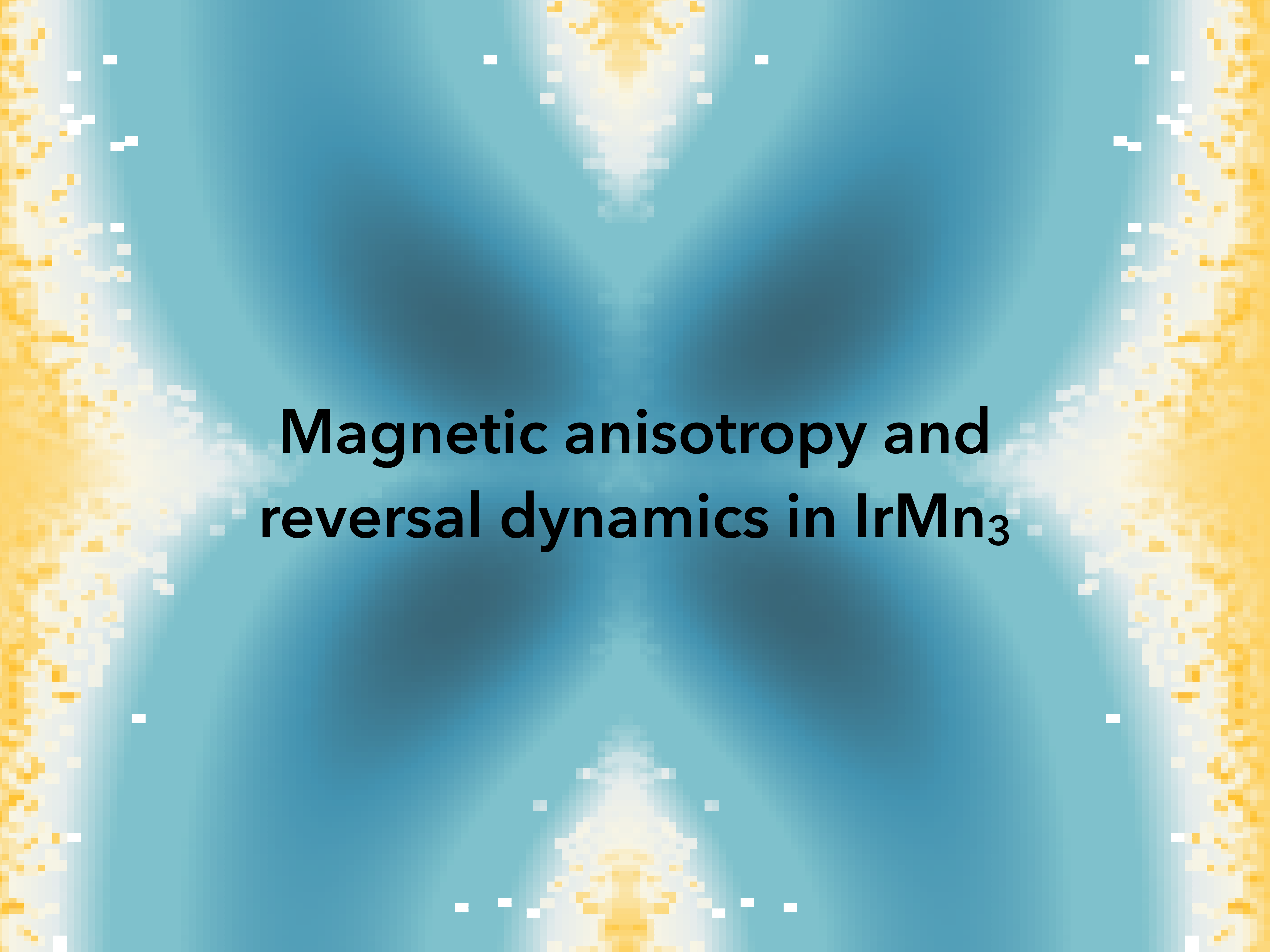


Ordered $Ir_{50}Mn_{50}$



World's most expensive ferromagnet…

Sarah Jenkins *et al*, Physical Review Materials **5** 034406 (2021)



Magnetic anisotropy and reversal dynamics in $IrMn_3$

Previous calculations of the strength of the anisotropy of IrMn

Experimental

Measuring the mean blocking temperature.

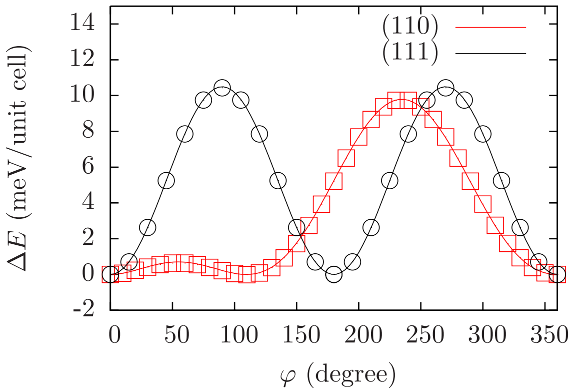
$$f = f_0 \exp\left(\frac{-\Delta E}{k_B T}\right)$$

10×10^5 J/m^3

Vallejo-Fernandez et al, APL 97, (2010)

Theoretical

Calculated the anisotropy using *ab-initio* methods.



300×10^5 J/m^3

Szunyogh et al, Phys Rev B 83, (2011)

Anisotropy in IrMn - cubic or uniaxial?

Callen Callen theory

$$\frac{K_{AF}(T)}{K_{AF}(0)} = \left(\frac{n_{AF}(T)}{n_{AF}(0)}\right)^{l}$$

$$\boxed{\begin{array}{ll}\text{Uniaxial:} & l = 3 \\ \text{Cubic:} & l = 10\end{array}}$$

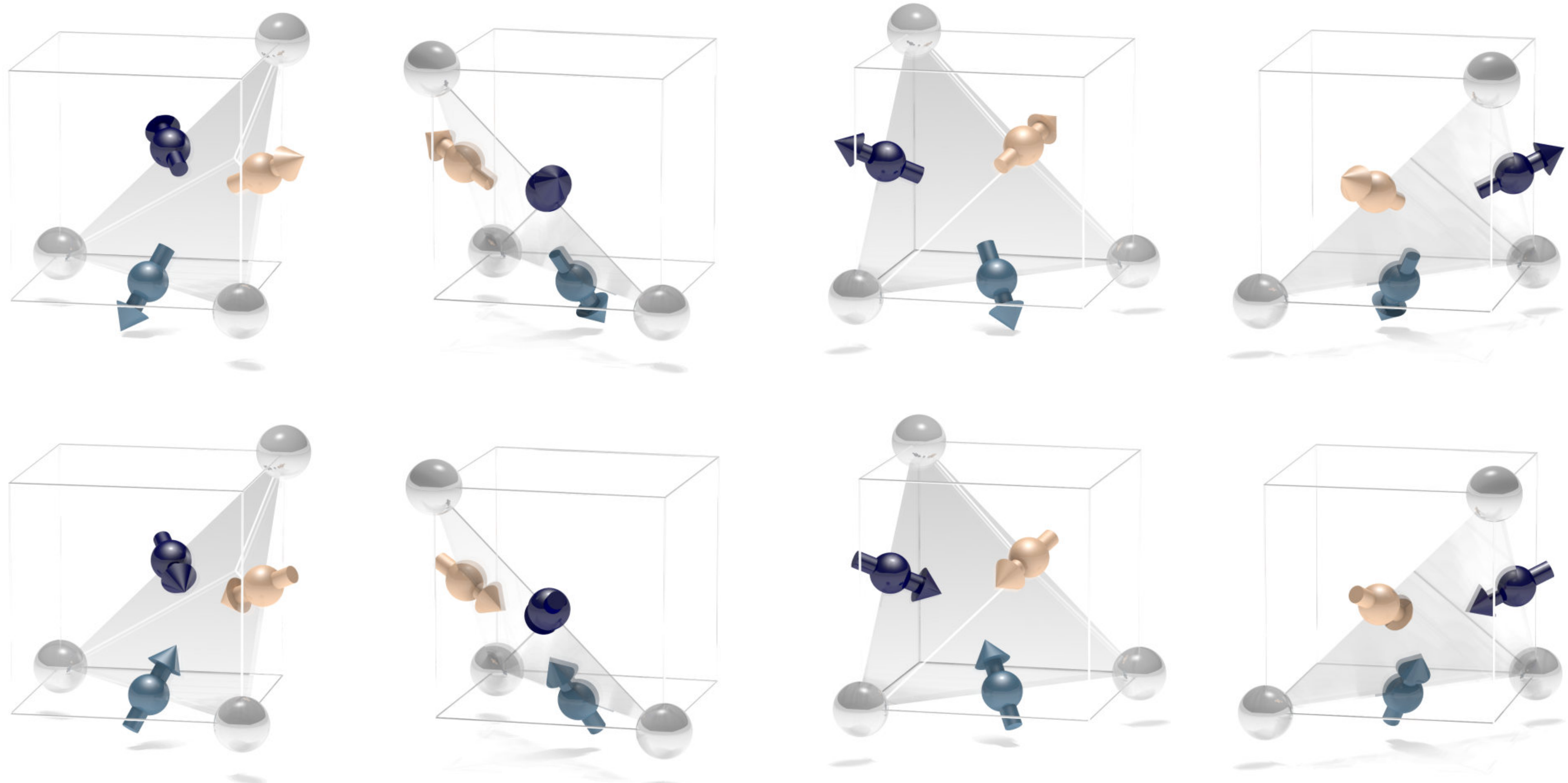
Experimental

Fits a uniaxial anisotropy from the temperature dependance of the sub lattice magnetisation (l ~ 3)

Theoretical

Calculates a uniaxial energy barrier for individual spins by rotating the ground state around the [111] direction.

T1 ground state magnetic structure



8 minimum energy ground states

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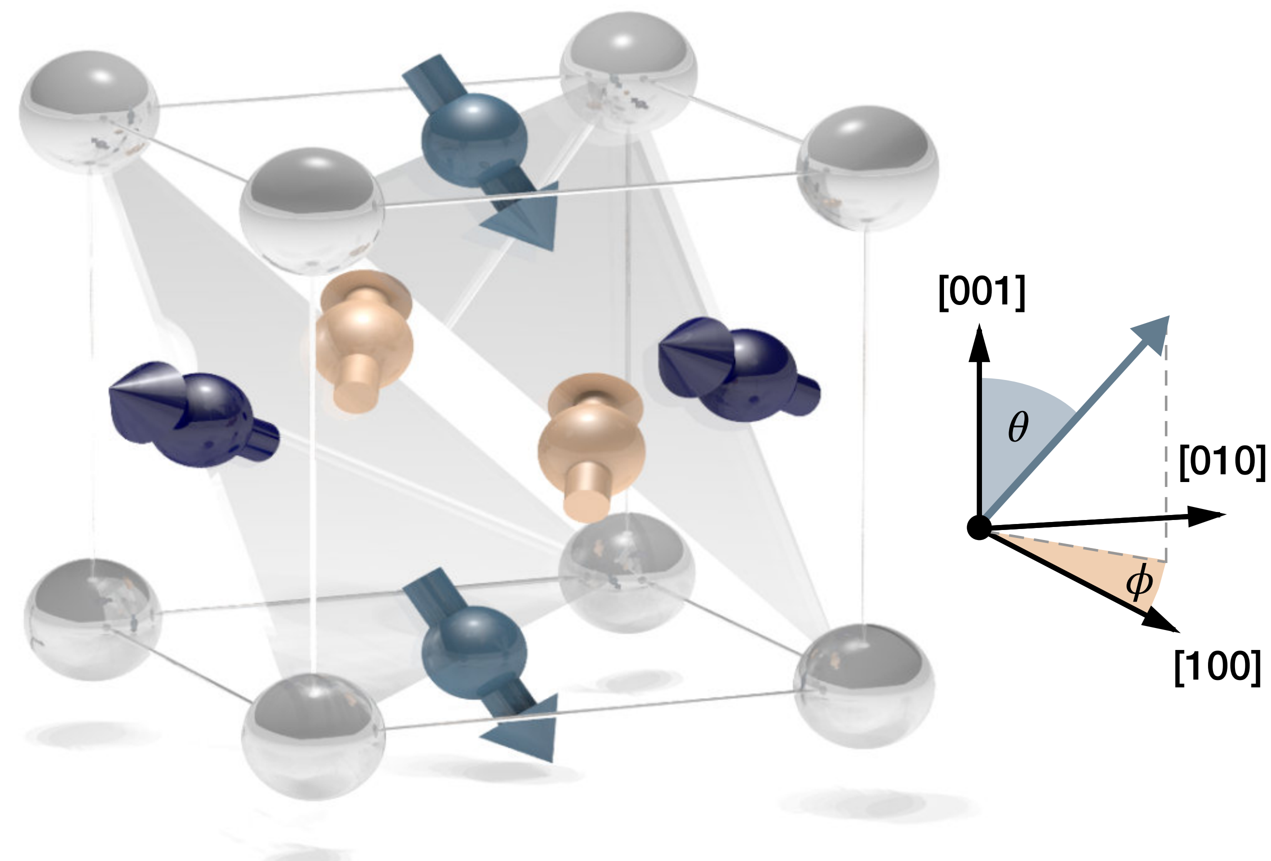
Calculating the anisotropy of IrMn: Constrained Monte Carlo

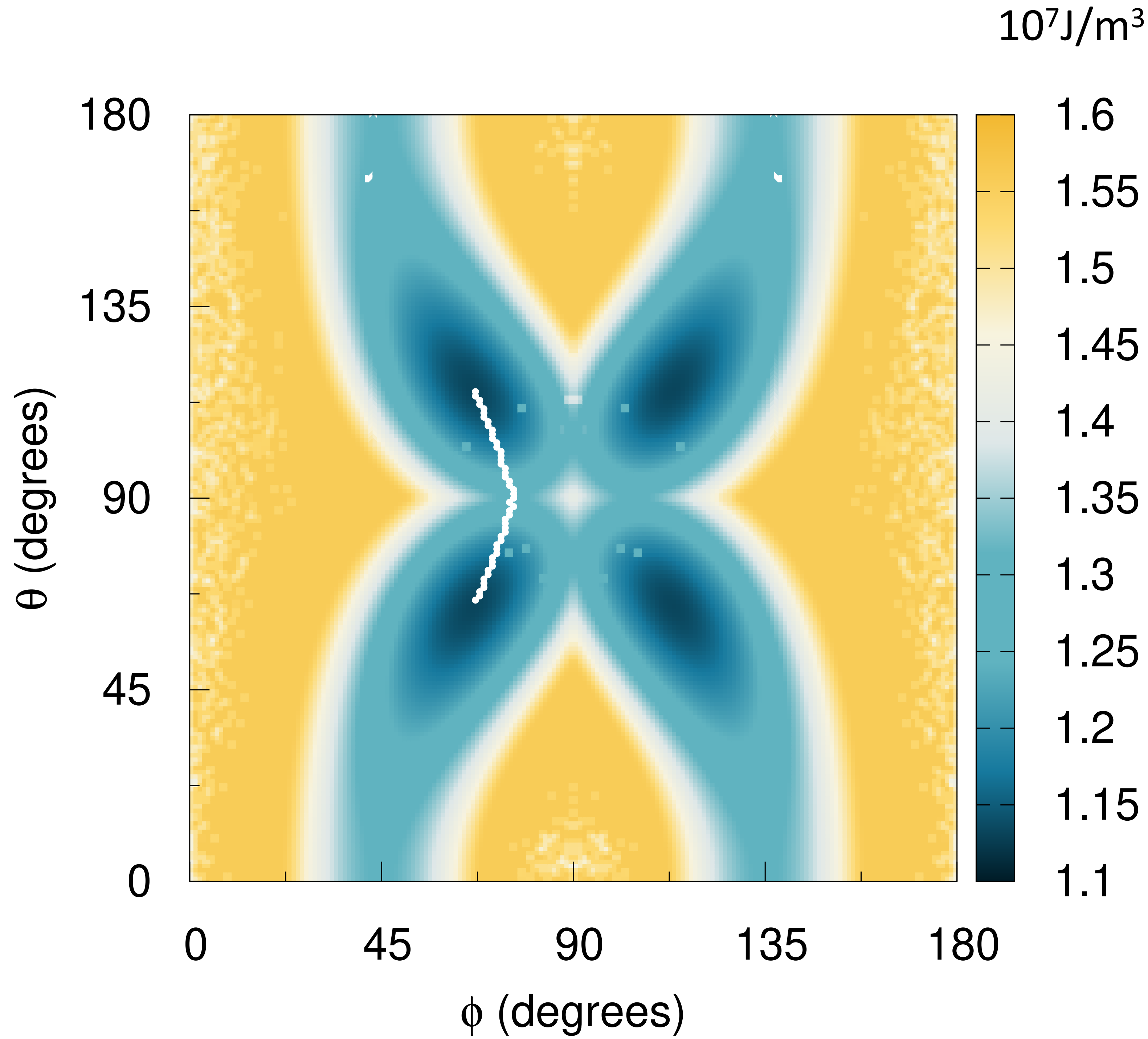
The magnetisation of one Mn sublattice is constrained along a direction (θ, ϕ)

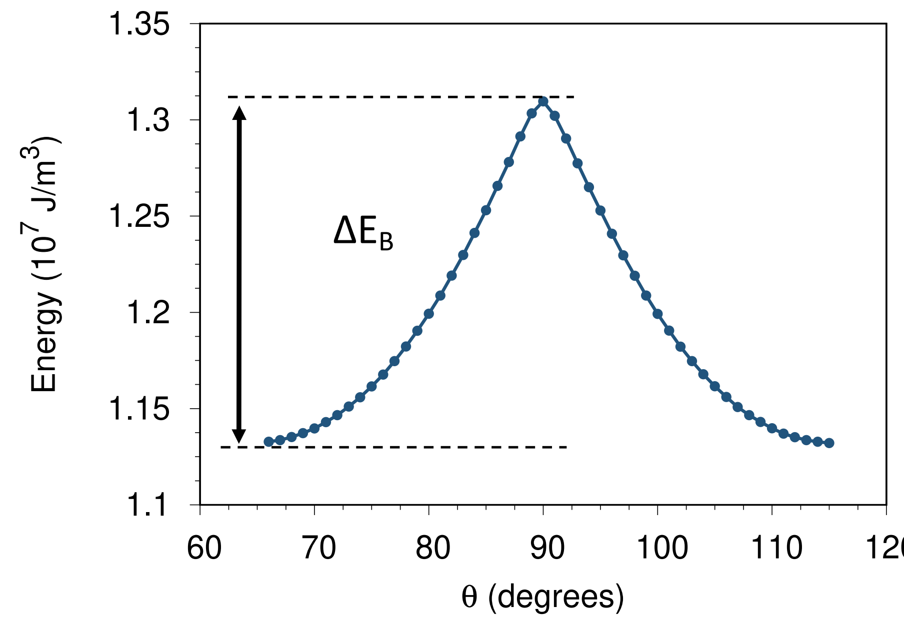
$$\mathcal{F}(\hat{\mathbf{M}}) = \mathcal{F}(\hat{\mathbf{M}}_0) + \int_{\hat{\mathbf{M}}_0}^{\hat{\mathbf{M}}} (\hat{\mathbf{M}}' \times \mathbf{T}') \cdot d\hat{\mathbf{M}}'$$



P Asselin et al, Physical Review B **82** 054415 (2010)

Calculated anisotropy energy surface of L12 IrMn3

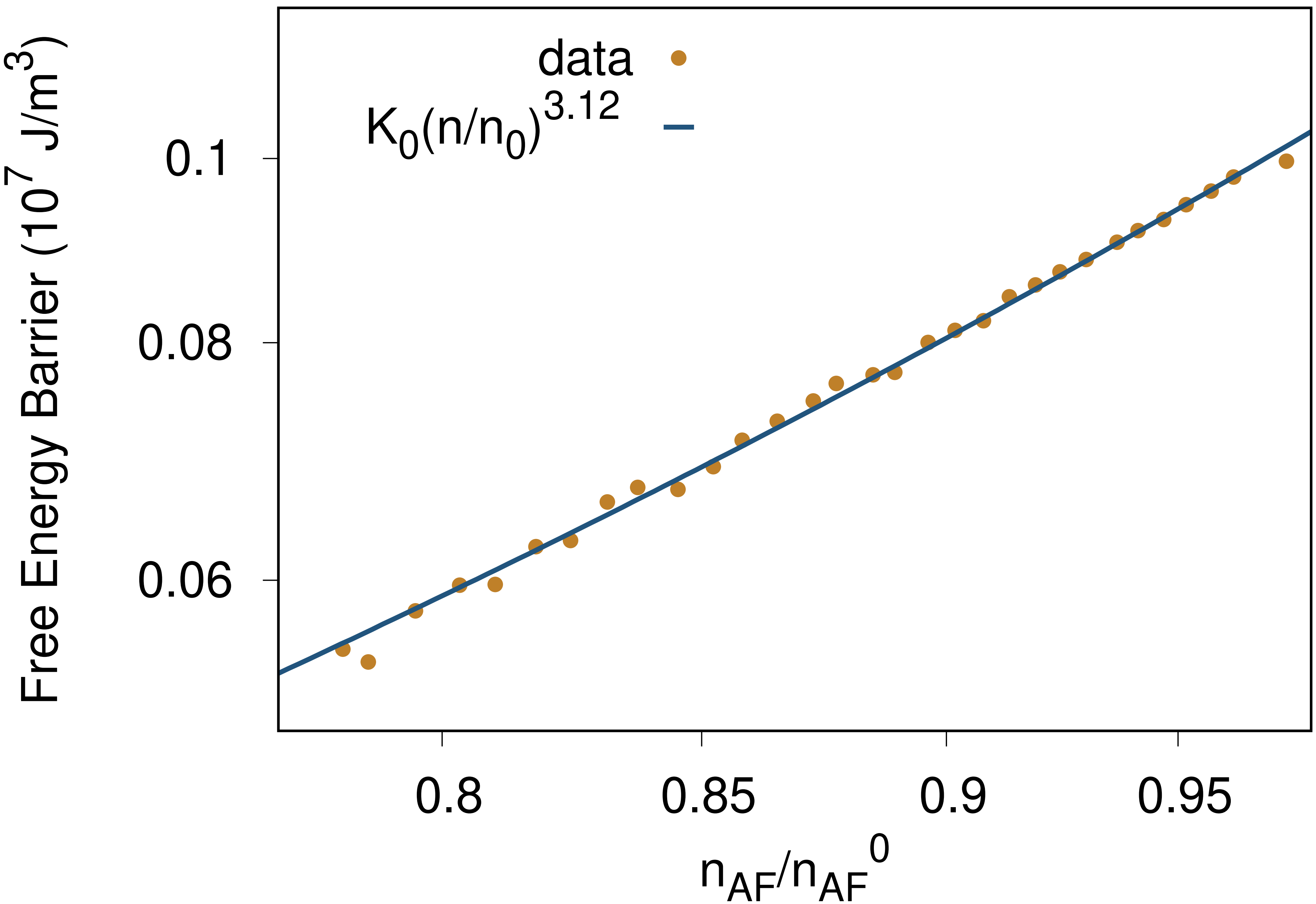




$\Delta E = 17 \times 10^5\ J/m^3$

Sarah Jenkins *et al*, Physical Review B (R) **100** 220405(R) (2019)

Temperature dependence of the anisotropy energy



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Sarah Jenkins *et al*, Physical Review B (R) **100** 220405(R) (2019)

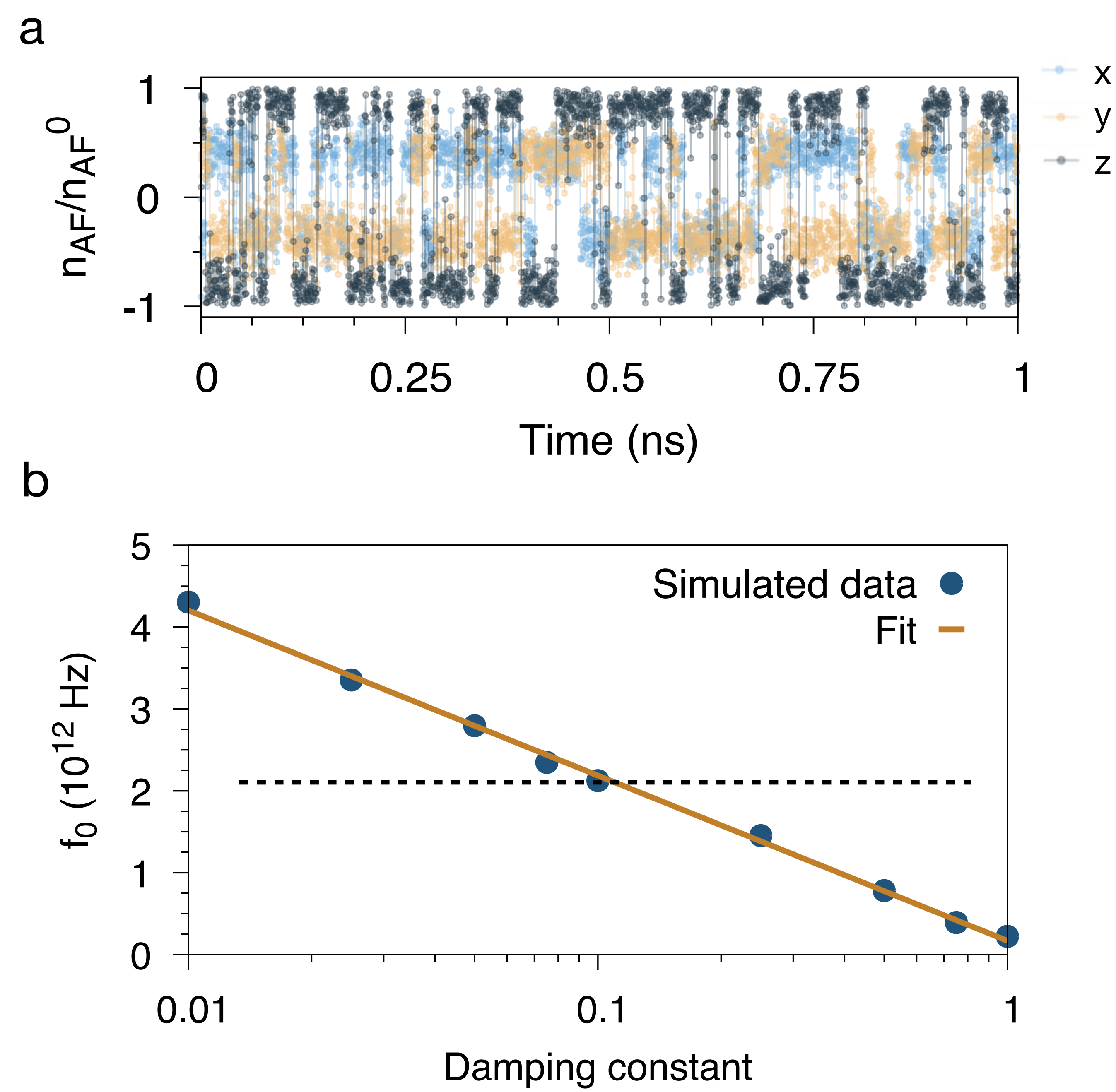
Calculation of the damping parameter in IrMn3

$$f = f_0 \exp\left(\frac{-\Delta E}{k_B T}\right)$$

Fitted f_0 from experimental data

$$f_0 = 2.1 \times 10^{12}\mathrm{s}^{-1}$$

$$\alpha_\mathrm{G} \approx 0.1$$



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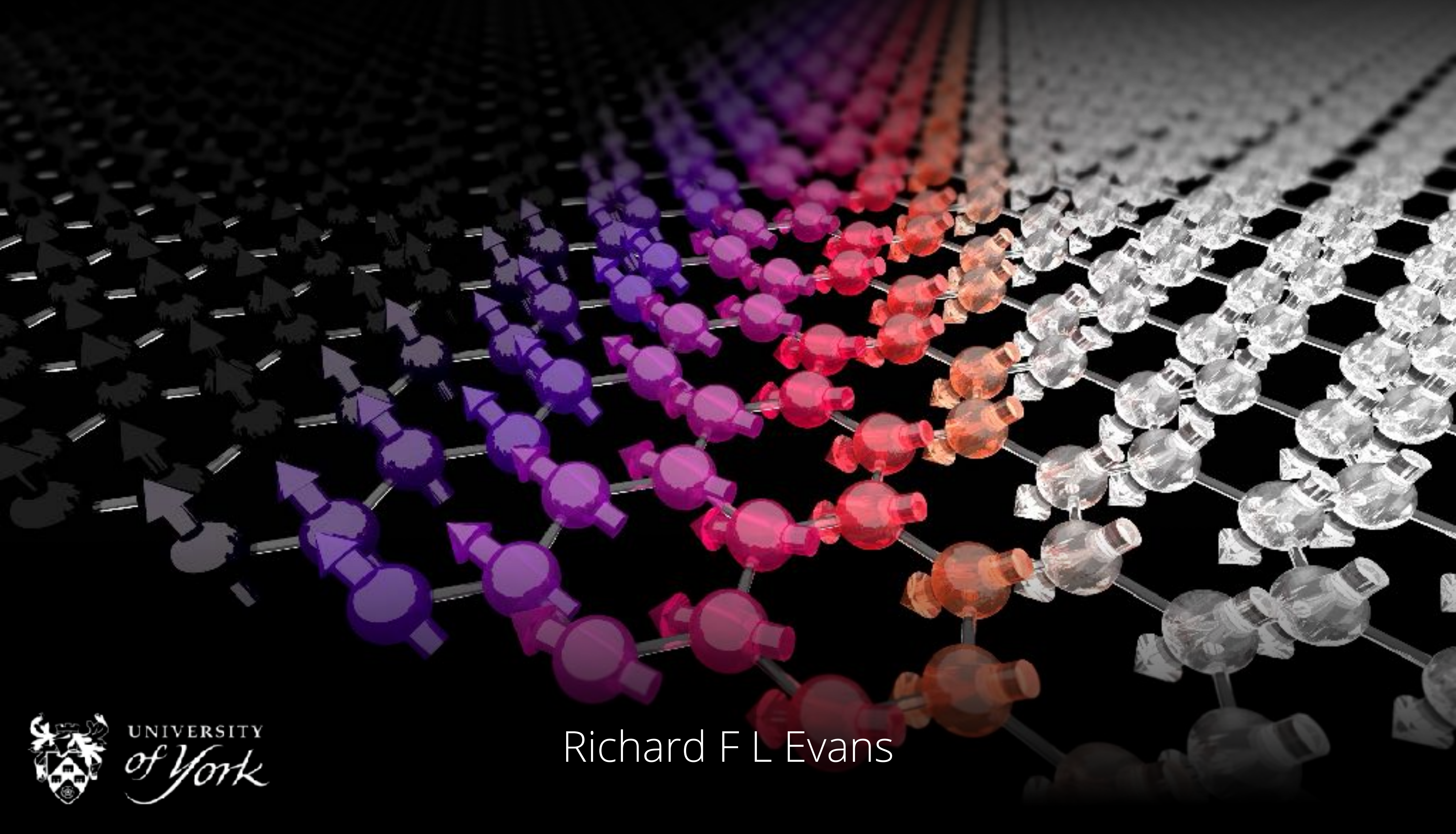
G. Vallejo-Fernandez *et al*, Appl. Phys. Lett. **97**, 222505 (2010)

Atomistic simulations of thermal properties and dynamics of 2D magnets



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Hohenberg-Mermin-Wagner theorem

Absence of Ferromagnetism or Antiferromagnetism in One- or Two-Dimensional Isotropic Heisenberg Models

N. D. Mermin and H. Wagner

Phys. Rev. Lett. **17**, 1133 – Published 28 November 1966; Erratum Phys. Rev. Lett. **17**, 1307 (1966)

An article within the collection: Letters from the Past - A PRL Retrospective

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ABSTRACT

It is rigorously proved that at any nonzero temperature, a one- or two-dimensional isotropic spin-S Heisenberg model with finite-range exchange interaction can be neither ferromagnetic nor antiferromagnetic. The method of proof is capable of excluding a variety of types of ordering in one and two dimensions.

Received 17 October 1966

DOI: https://doi.org/10.1103/PhysRevLett.17.1133

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Common belief - magnetism in 2D isotropic film not possible

5.4.3 Mermin–Wagner theorem

The derivation of the spin-wave dispersion has been based on the existence of a ferromagnetic state in an isotropic chain, or a three-dimensional lattice. The assumptions warrant scrutiny. The number of magnons excited at a temperature T is given by

$$\mathfrak{n}_{\mathfrak{m}} = \int_0^\infty \frac{\mathcal{N}(\omega_q)\mathrm{d}\omega_q}{\mathrm{e}^{\hbar\omega_q/kT} - 1},$$

where the density of states for magnons $\mathcal{N}(\omega_q)$ in one, two and three dimensions varies as $\omega_q^{-1/2}$, $\omega_q^o =$ constant and $\omega_q^{1/2}$, respectively. The argument is similar to that for the electron gas, given in §3.2.5, which has similar dispersion relations. Setting $x = \hbar\omega_q/k_B T$, the integral in three dimensions varies as $(k_B T/\hbar)^{3/2}\int_0^\infty x^{1/2}\mathrm{d}^3x/(\mathrm{e}^x - 1)$, whence comes the Bloch $T^{3/2}$ law (5.61). However, the integrals *diverge* at finite temperature in one and two dimensions. The ferromagnetically ordered state should be unstable in dimensions lower than 3. This is the Mermin–Wagner theorem. Magnetic order is possible in the Heisenberg model in three dimensions, but not in one or two. The linear chain, our example of spin-wave dispersion, cannot order except at $T = 0$ K.

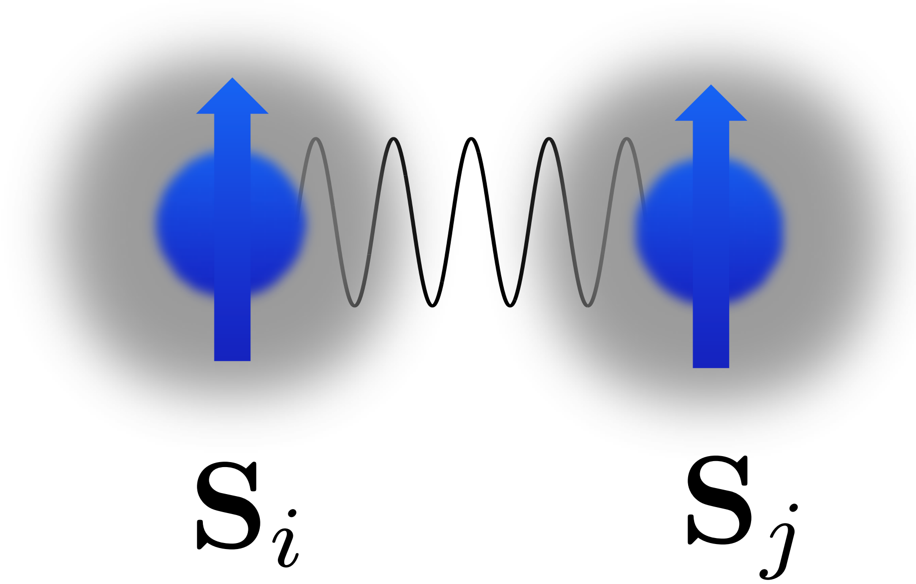
The consequences of this theorem are not as catastrophic as they seem at first sight. The divergence is avoided if there is some anisotropy in the system, which creates a gap in the spin-wave spectrum at $\boldsymbol{q} = 0$; the lower limit of integration is then greater than zero and the divergence is avoided. Some anisotropy is always caused by crystal field or dipolar interactions. Two-dimensional ferromagnetic layers do exist in reality, thanks to anisotropy (§8.1).

JMD Coey, Magnetism and magnetic materials

Breaking through the Mermin-Wagner limit in 2D van der Waals magnets

Sarah Jenkins, Levente Rozsa, Unai Atxitia, Richard F. L. Evans,
Kostya S. Novoselov, Elton J. G. Santos

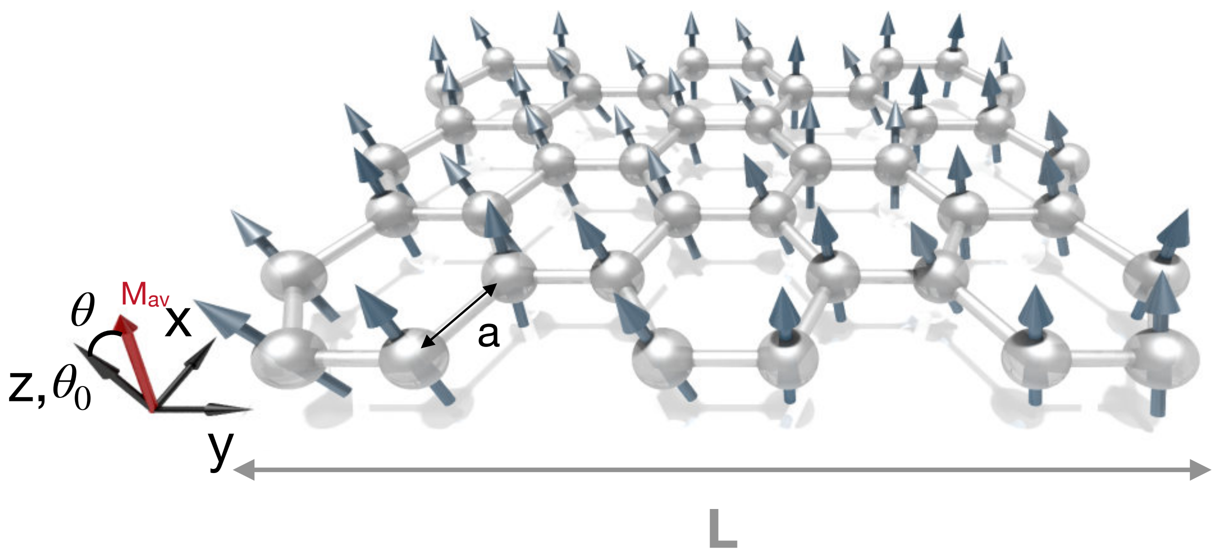
Foundation of the atomistic model is Heisenberg exchange

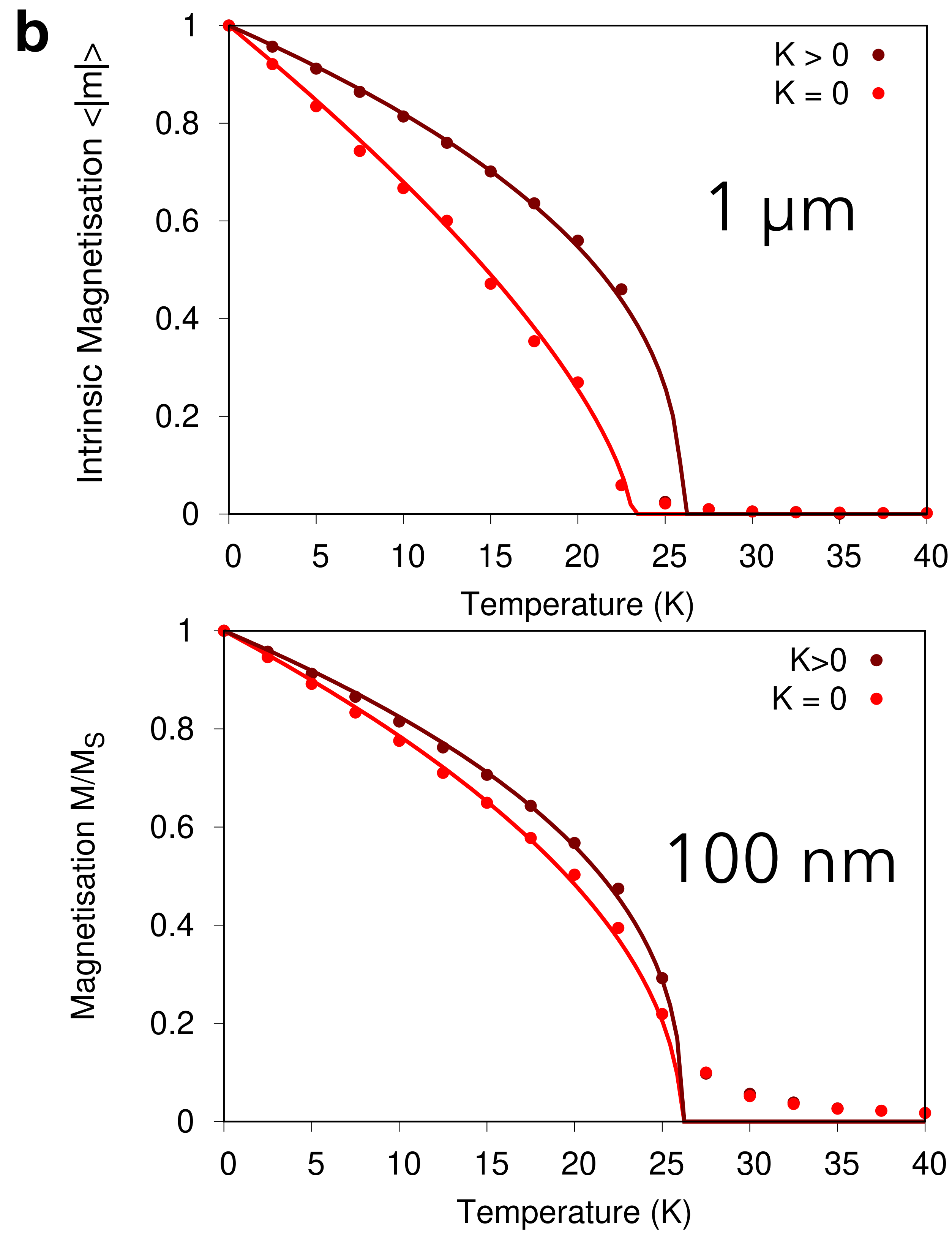


$$\mathcal{H}_{\mathrm{exc}} = -\frac{1}{2}\sum_{i,j} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j$$

Natural discrete limit of magnetization

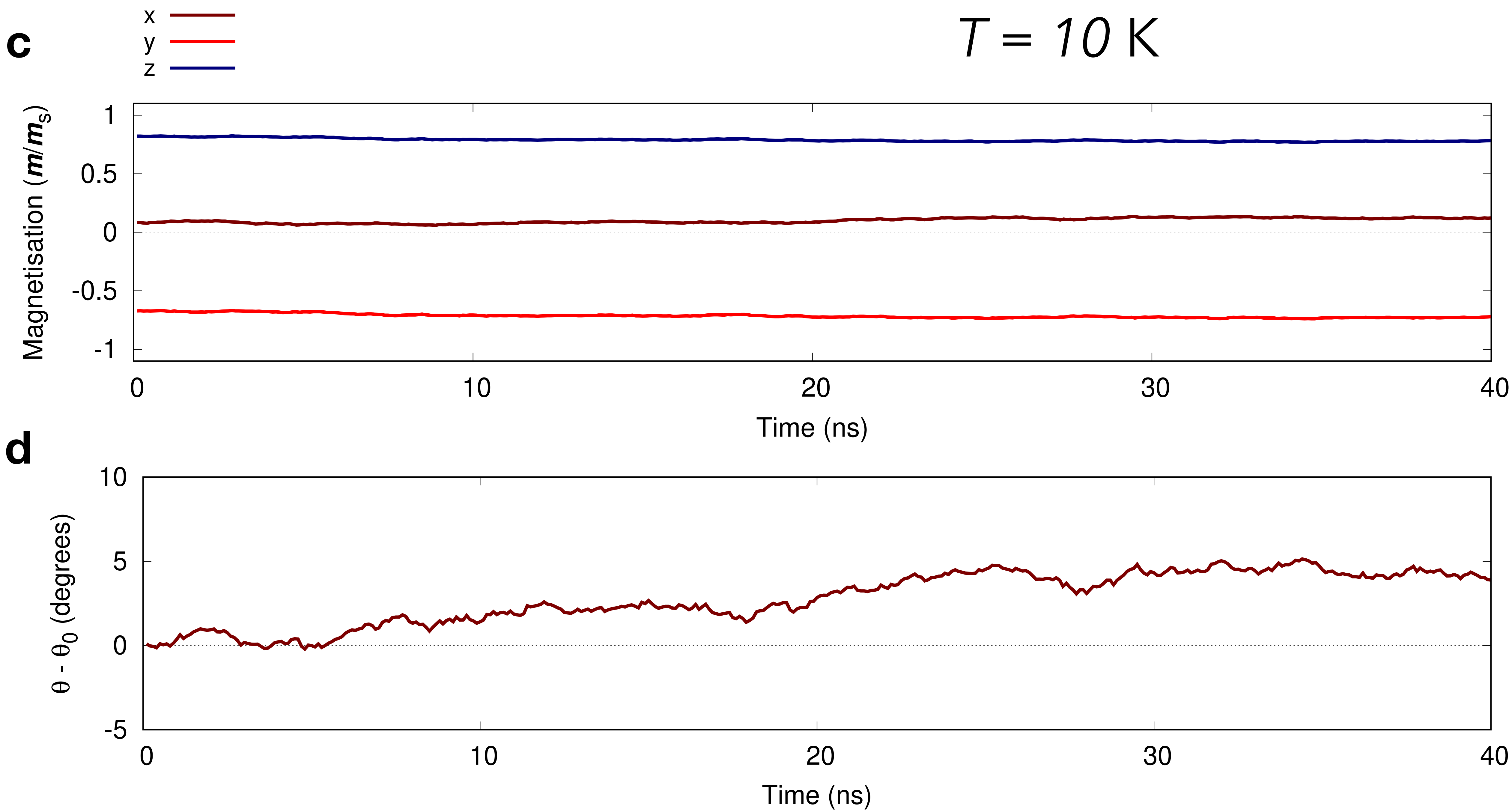
Atomistic model L = 1 μm $M_S(T)$ *(Monte Carlo)*





$$\langle|\mathbf{m}|\rangle(T) = \left(1 - \frac{T}{T_x}\right)^{\beta}$$

Evolution in time for *L* = *1* μm *(sLLG)*



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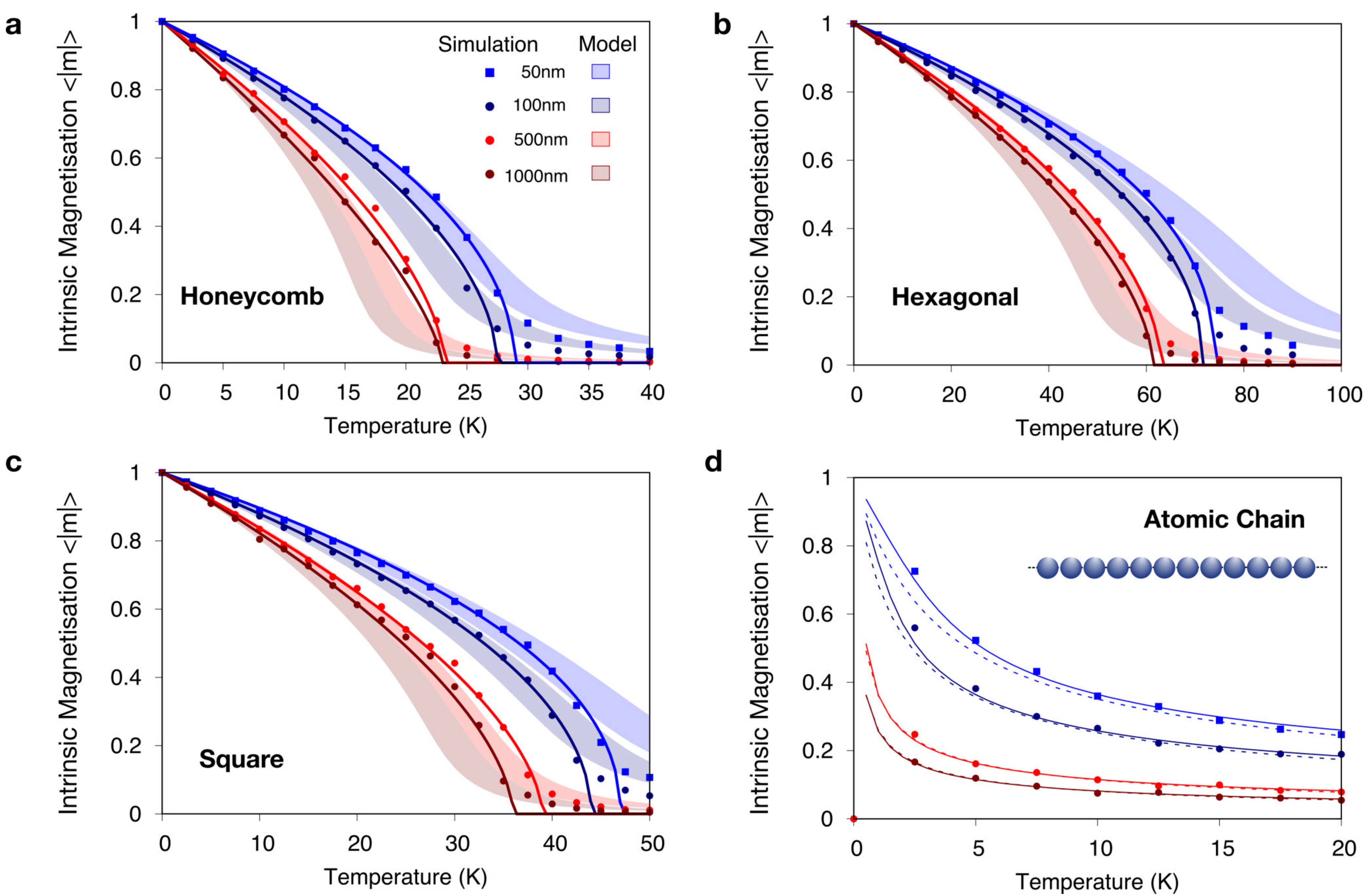
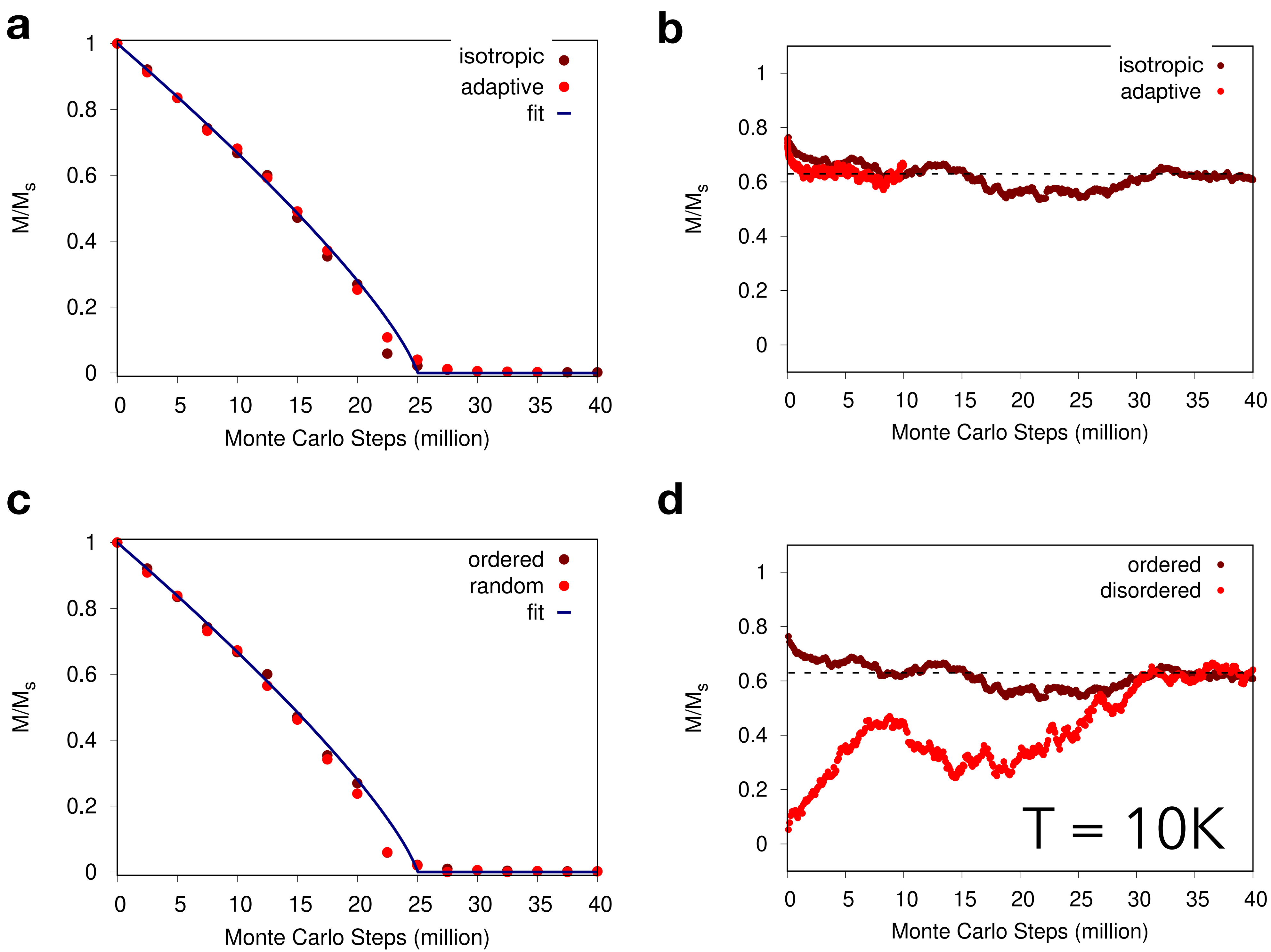
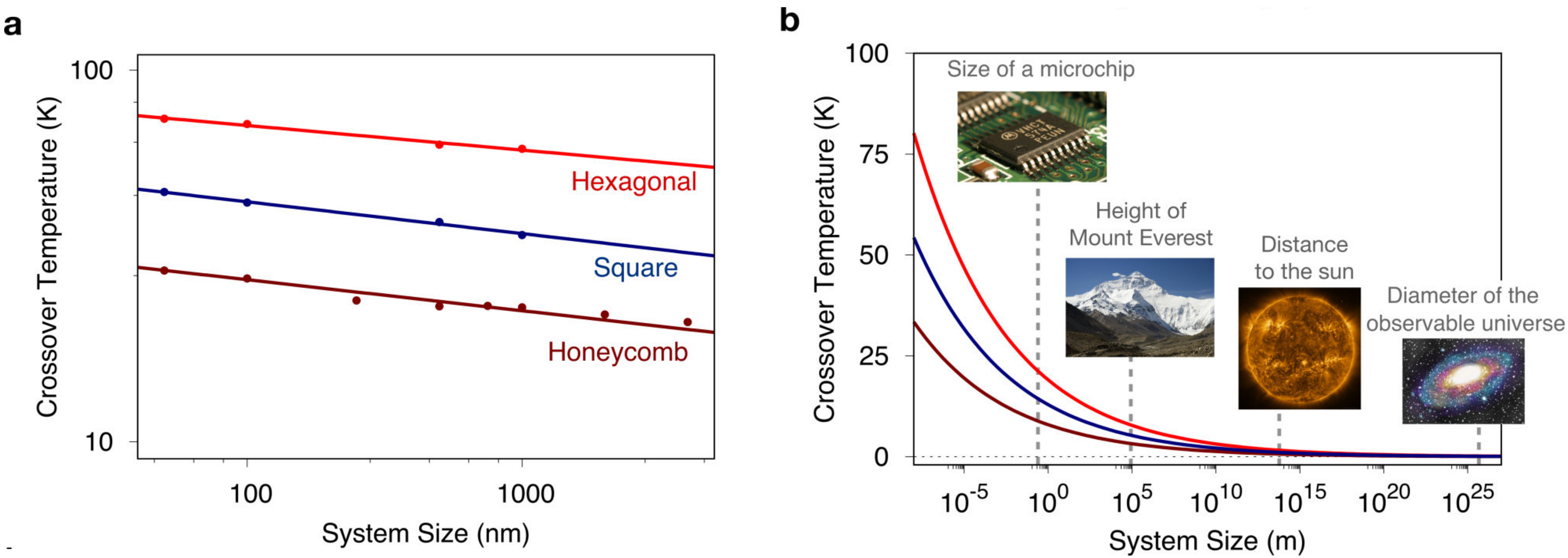
Size-dependent magnetisation



Magnetisation vs temperature from ordered and disordered starting states



Size-scaling of the crossover temperature



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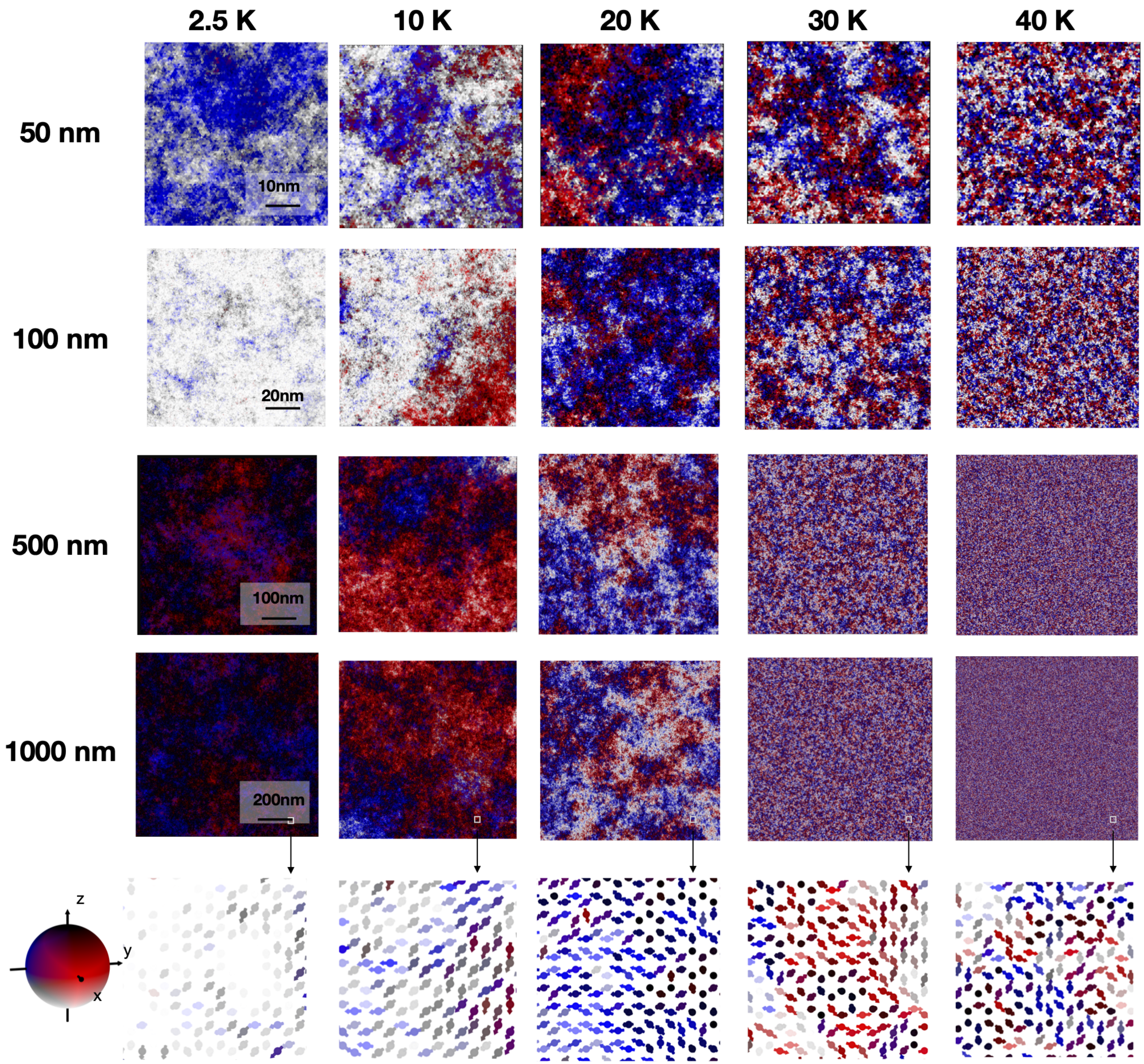
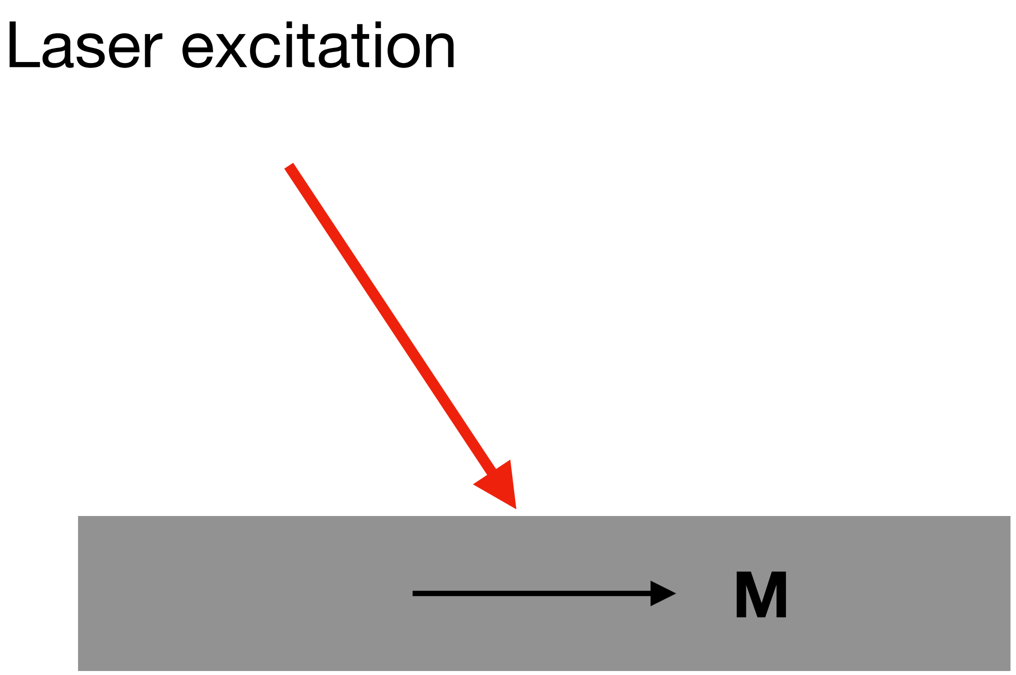
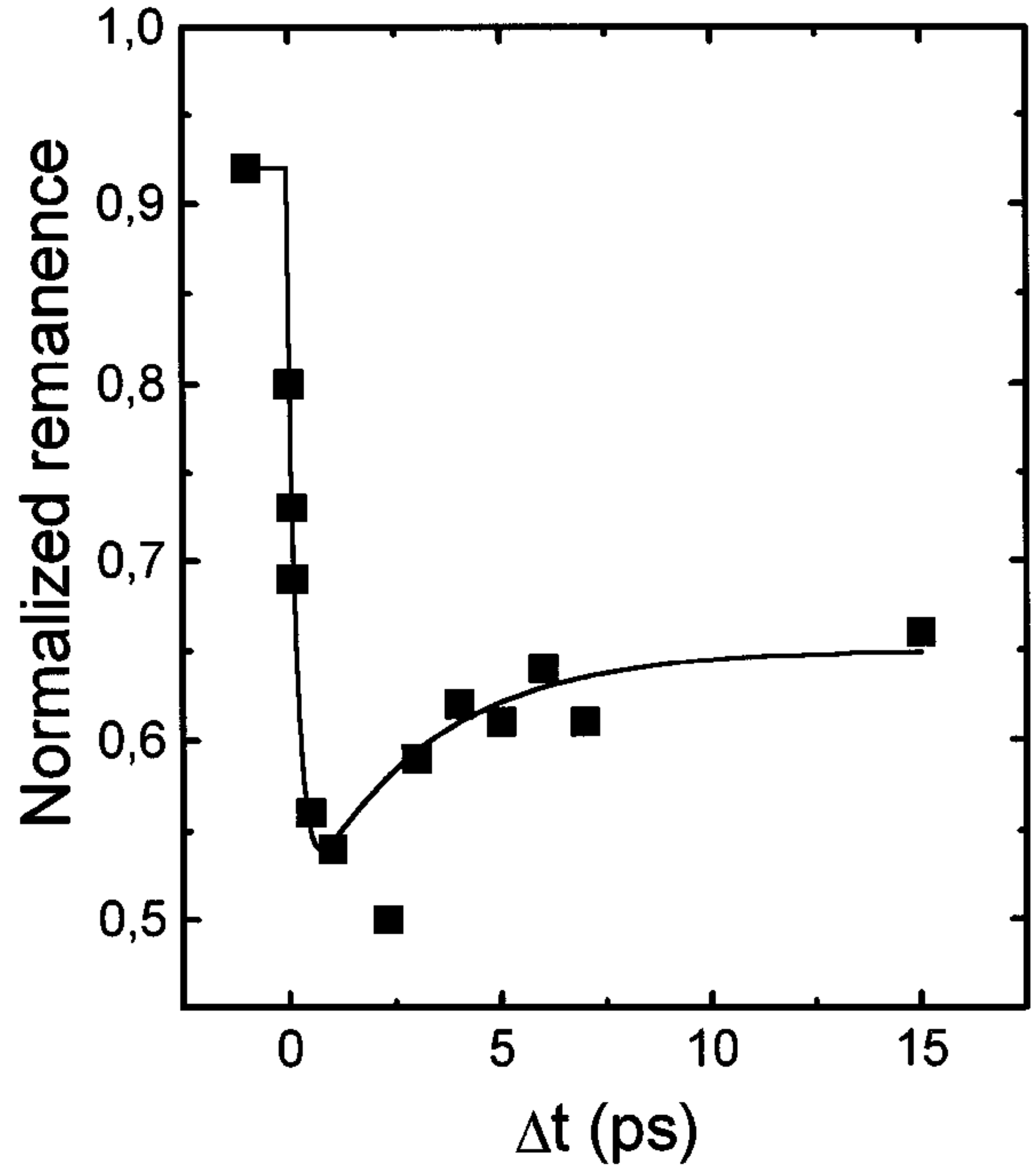
Evolution of magnetic structure with size, temperature



Thermodynamics of ultrafast magnetization processes

Ultrafast demagnetization in Ni

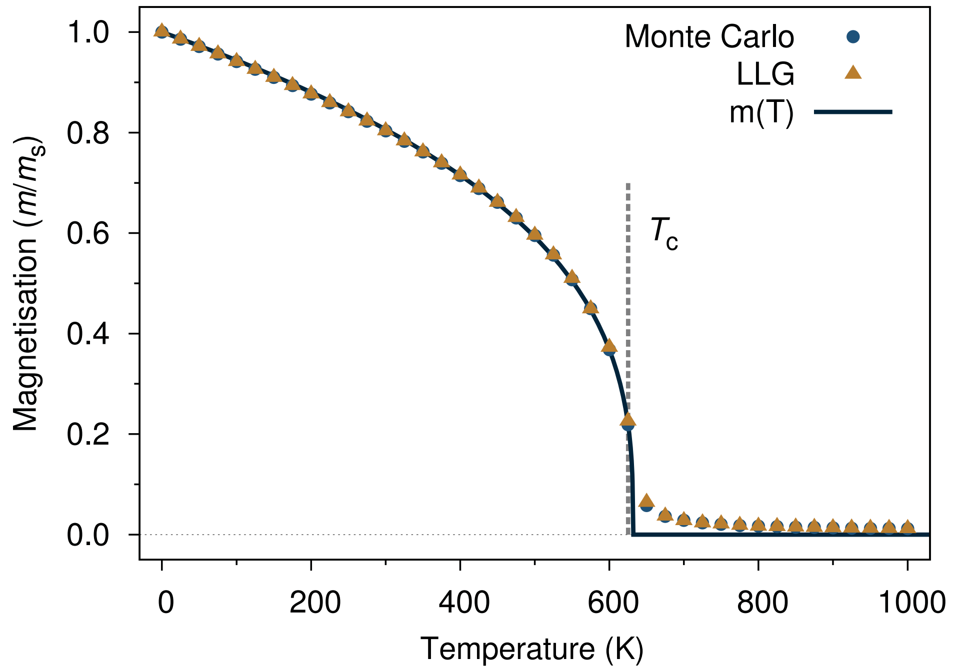


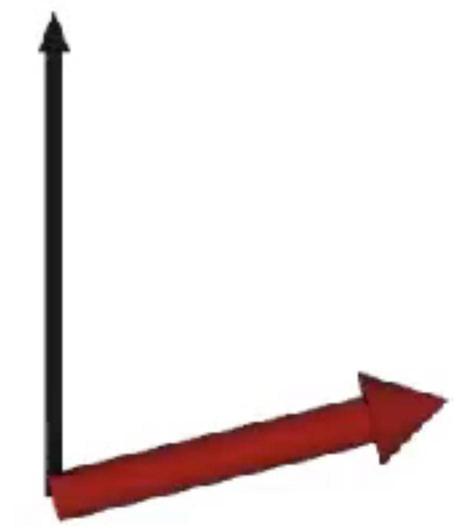


E. Beaurepaire et al, Phys. Rev. Lett. **76** 4250 (1996)

Origin of thermal fluctuations in the atomistic model

- Lets go back to the thermal fluctuations in the atomic model



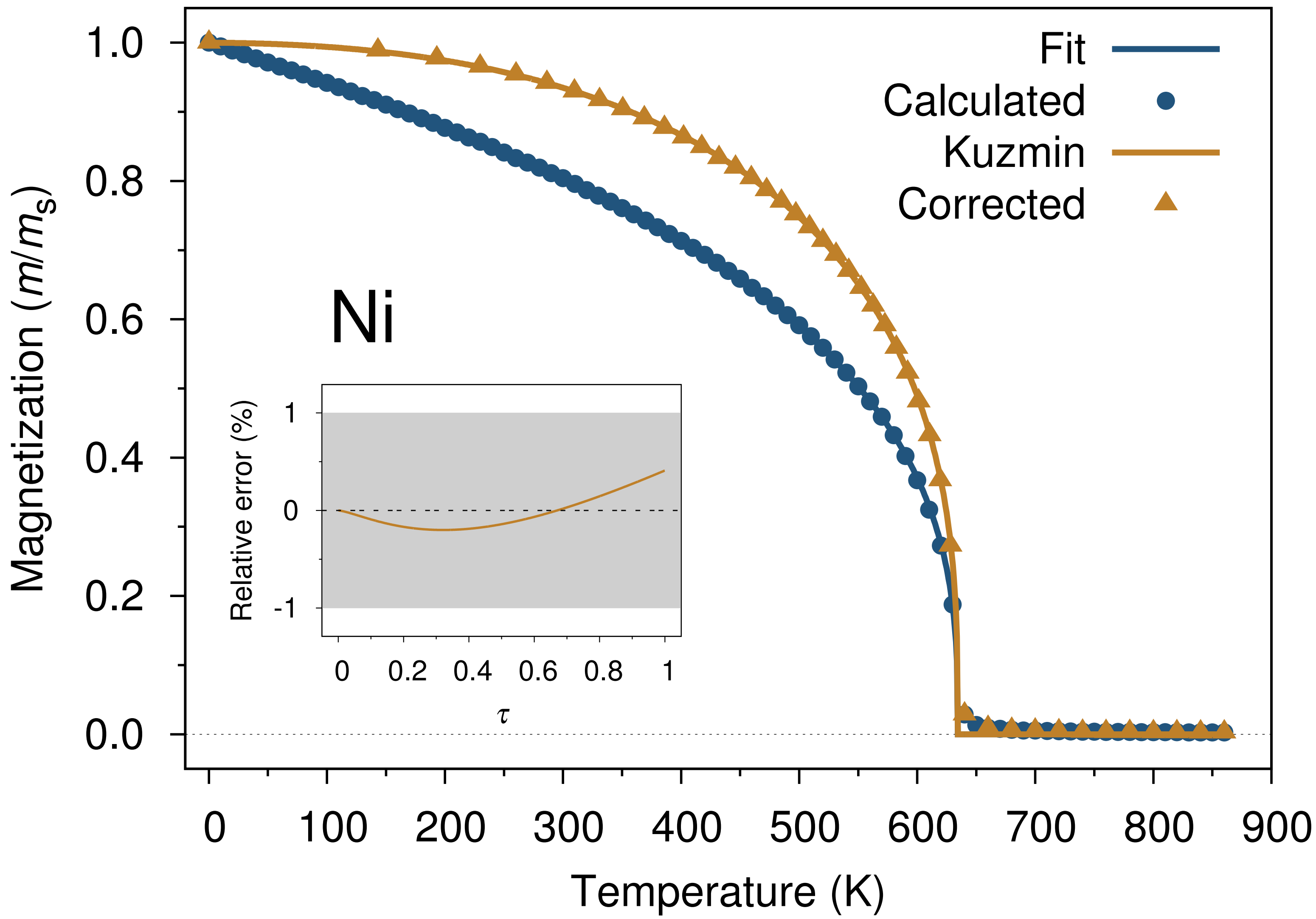


- Physically caused by spin scattering phenomena
  - electron-spin, spin-phonon, spin-photon

- Laser interaction causes heating of the electrons and more scattering events -> fast increase in the effective temperature in the material

Equilibrium properties of Ni

- Use spin temperature rescaling to accurately reproduce temperature dependent magnetization



Evans *et al*, Phys. Rev. B **91**, 144425 (2015)

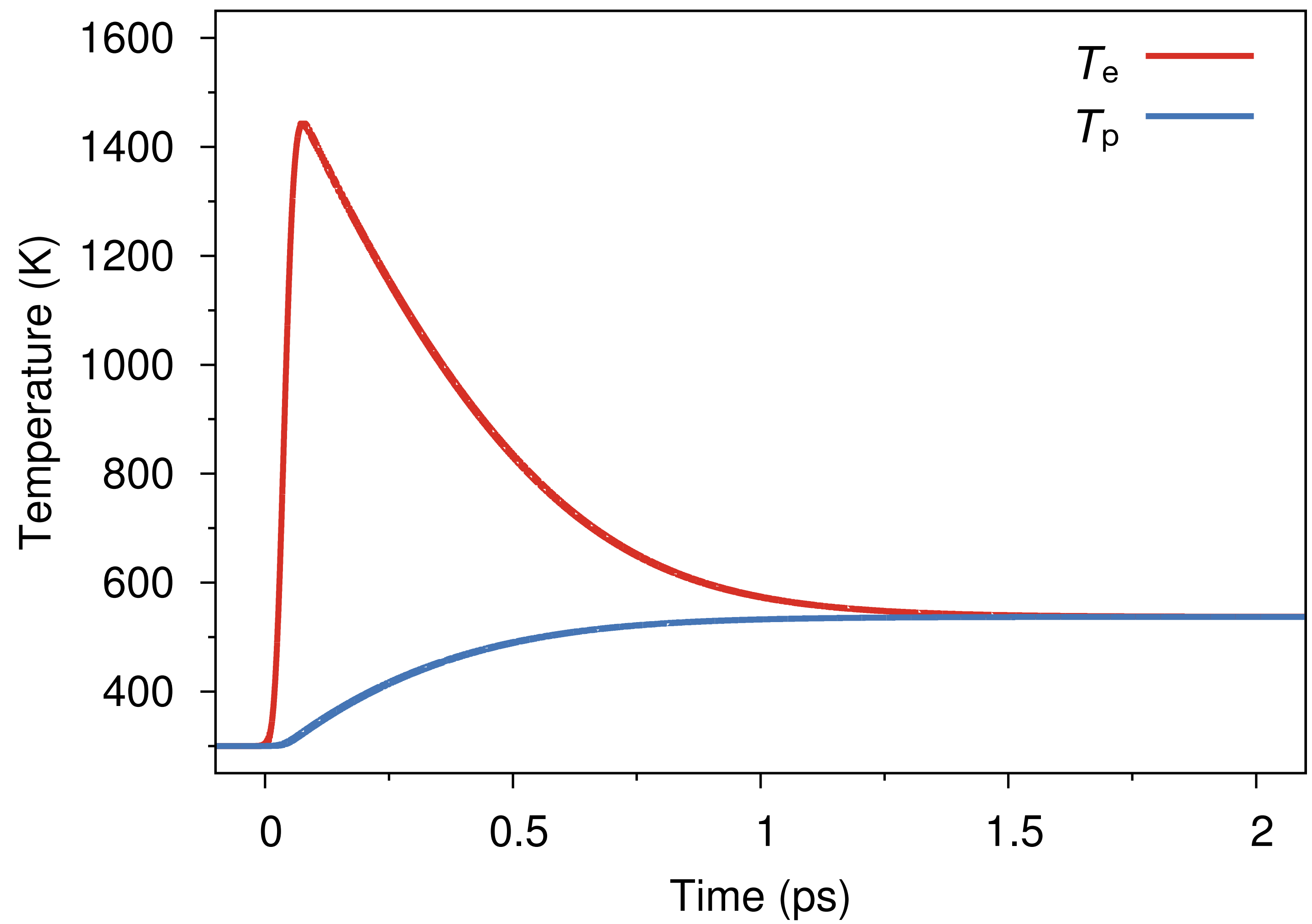
Simulating a laser pulse: two temperature model

$$C_e \frac{\partial T_e}{\partial t} = -G(T_e - T_p) + S(t)$$

$$C_p \frac{\partial T_p}{\partial t} = -G(T_p - T_e)$$

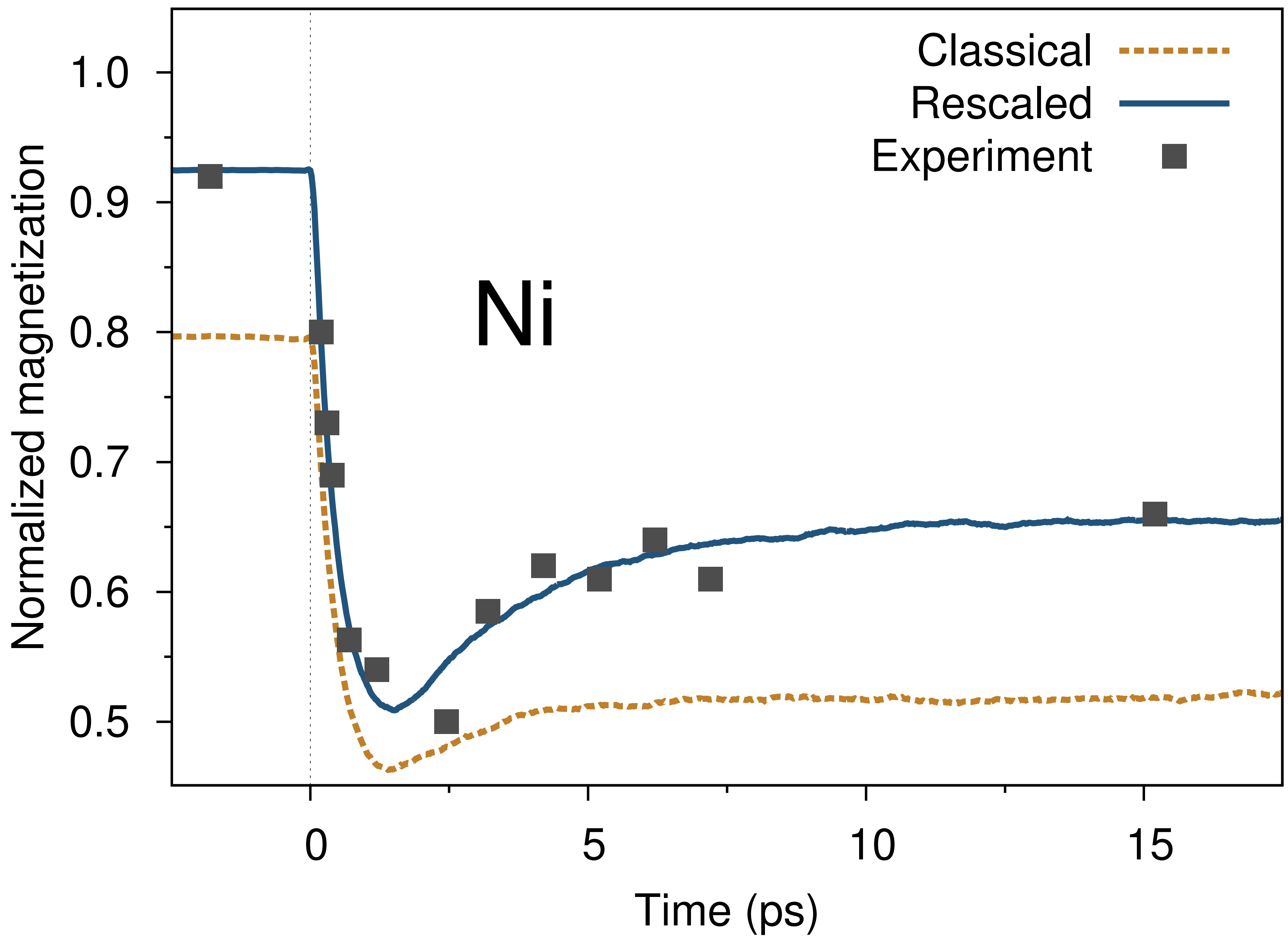
Free electron approximation

$$C_e = C_0 T_e$$



S. Anisimov, B. Kapeliovich, and T. Perelman, Sov. Phys. JETP **39**, 375 (1974).

Ultrafast demagnetization in Ni

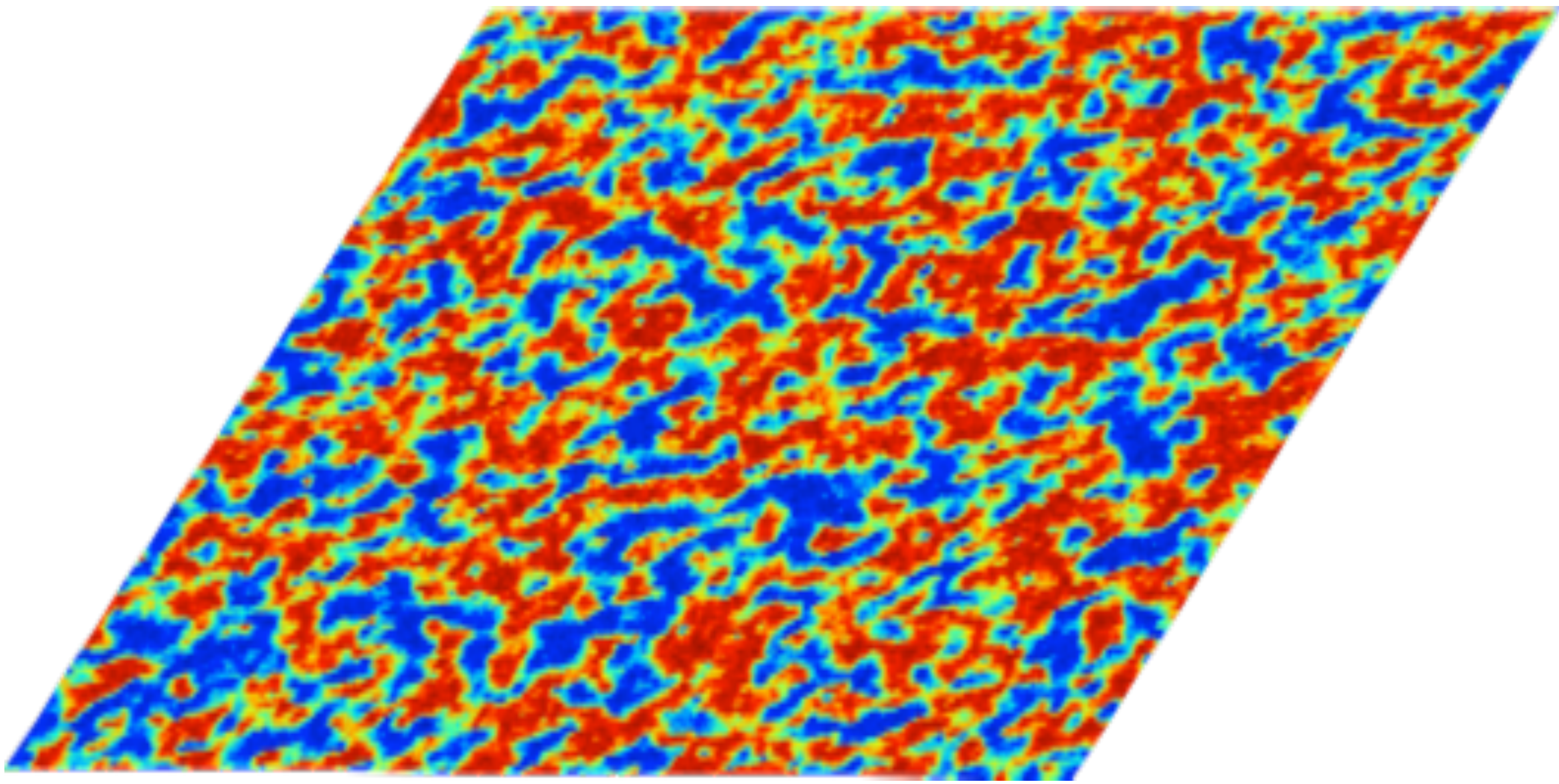


damping-constant = 0.001

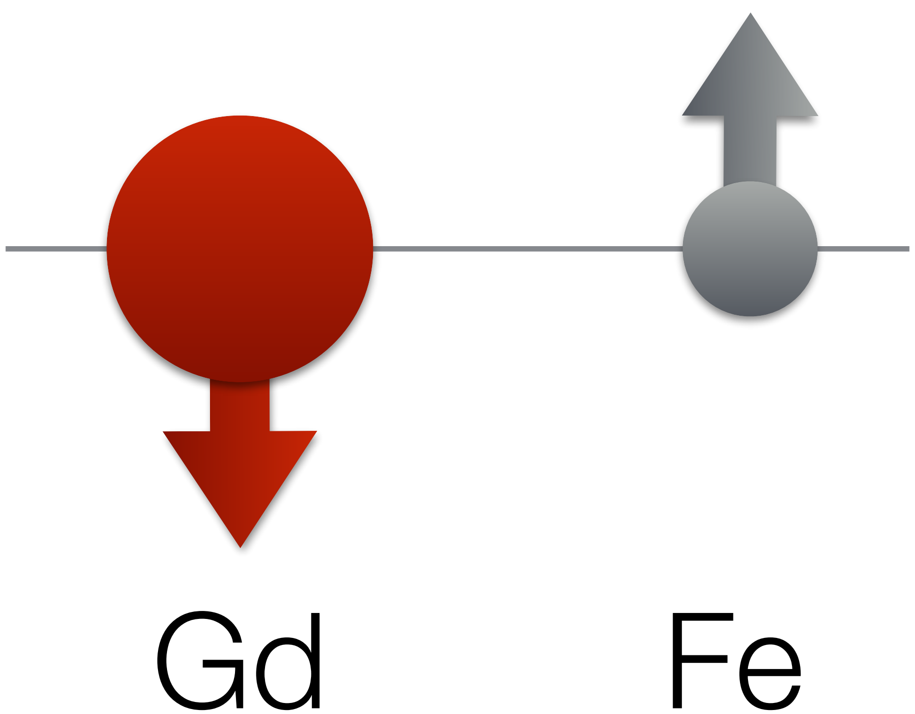
E. Beaurepaire *et al*, Phys. Rev. Lett. **76**, 4250 (1996)

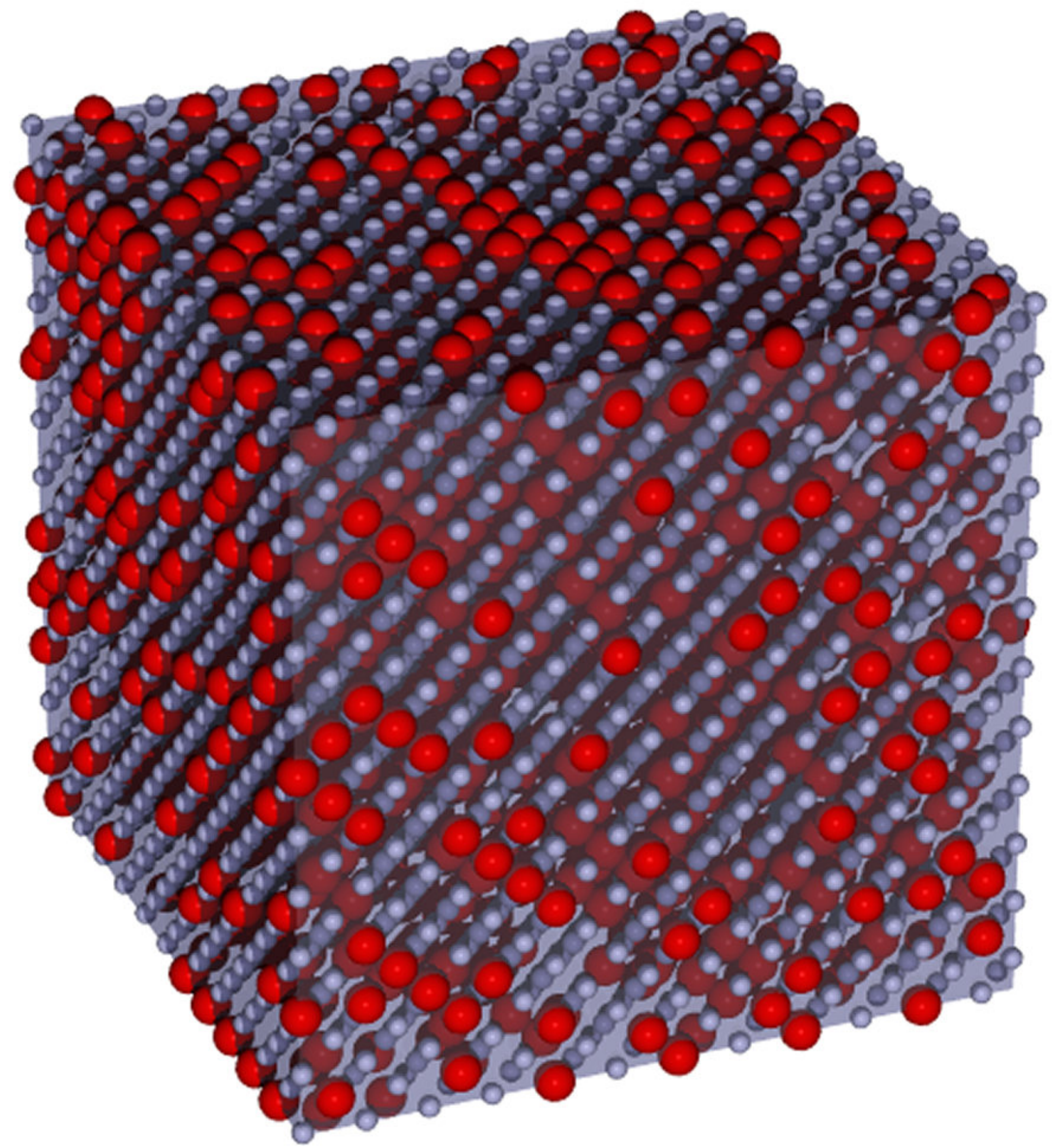
R. F. L. Evans *et al*, Phys. Rev. B **91**, 144425 (2015)

Ultrafast heat-induced switching of GdFeCo



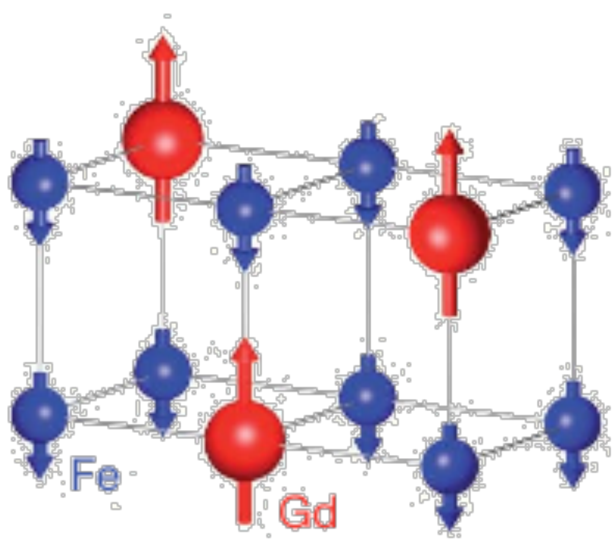
GdFe ferrimagnet

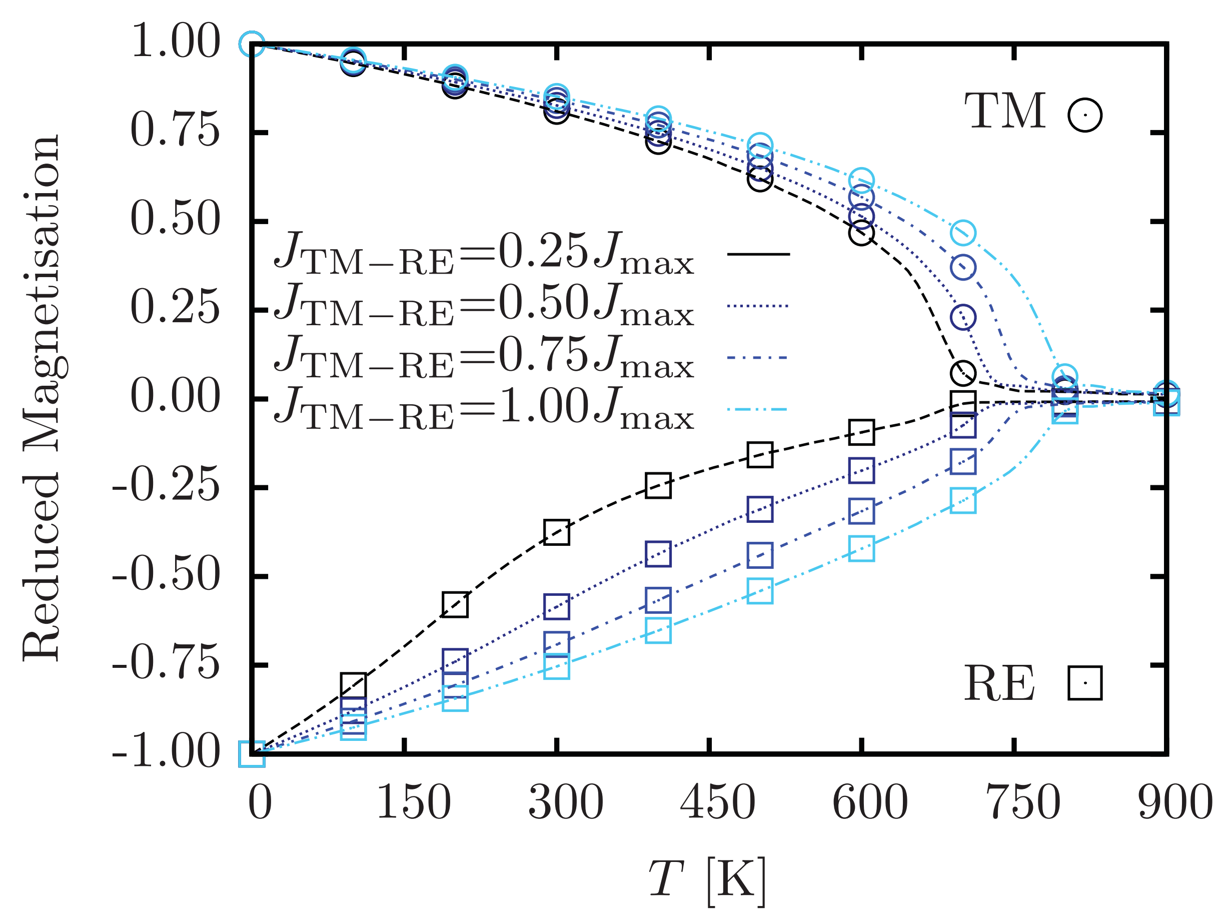




Ferrimagnetic nature of GdFe(Co) and spin models

$$\mathcal{H} = -\frac{1}{2}\sum_{\langle i,j\rangle} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j - \sum_{i=1}^{\mathcal{N}} D_i(\mathbf{S}_i \cdot \mathbf{n}_i)^2 - \sum_{i=1}^{\mathcal{N}} \mu_i \mathbf{B} \cdot \mathbf{S}_i$$

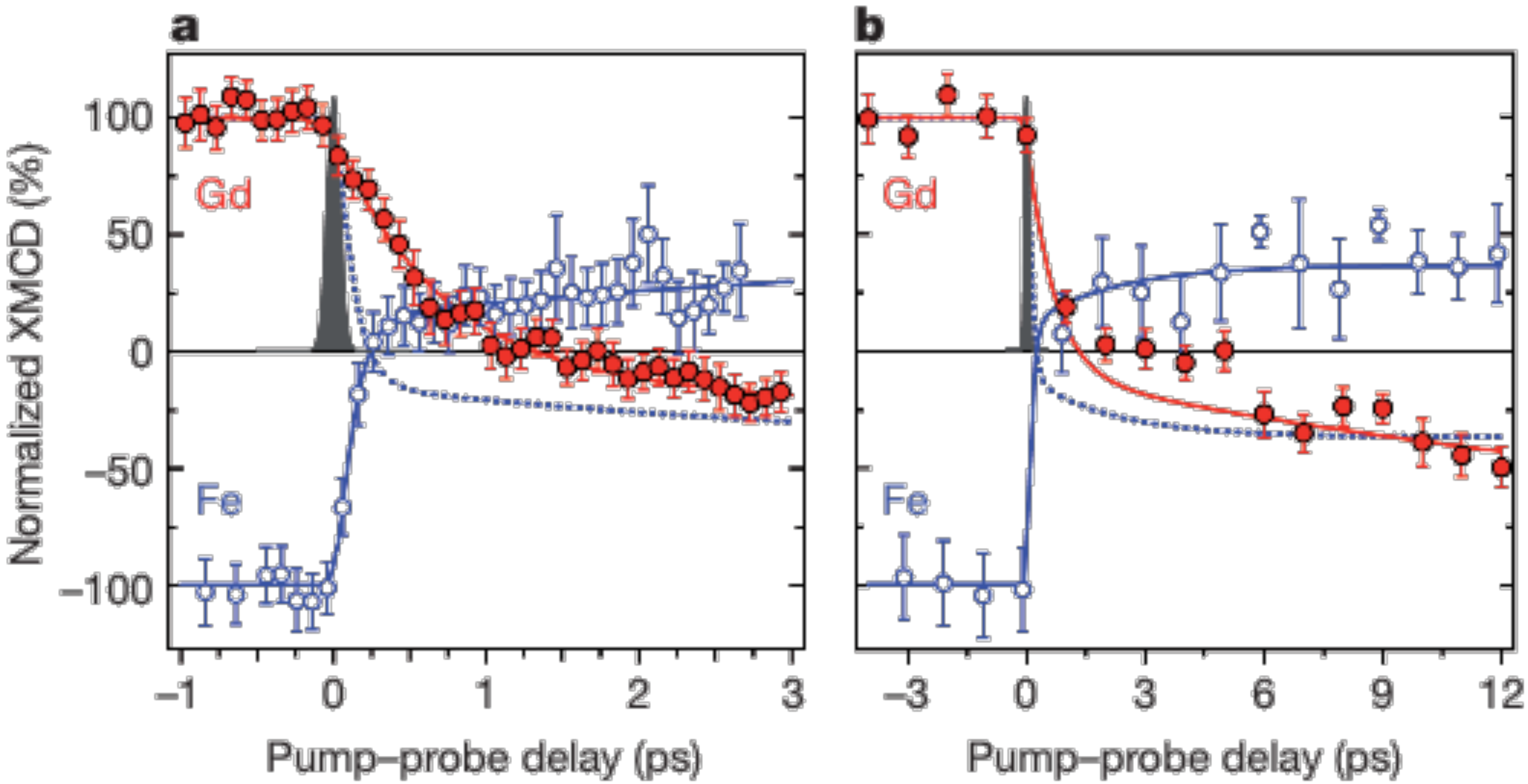




T Ostler *et al*, Phys. Rev. B **84**, 024407 (2011)

UNIVERSITY of York

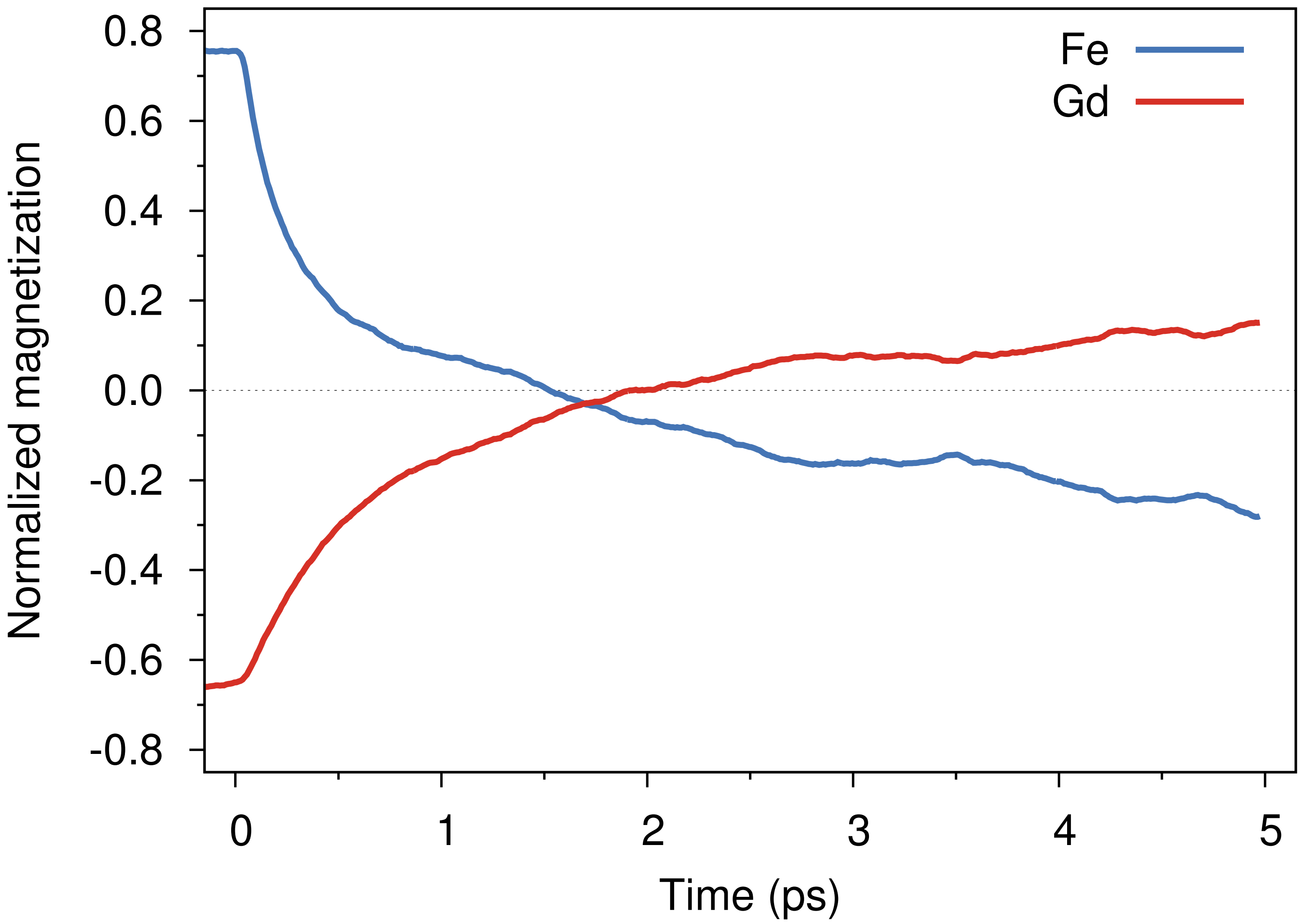
Ultrafast magnetization dynamics measured with XMCD



Complex reversal mechanism owing to different sub lattice magnetization dynamics

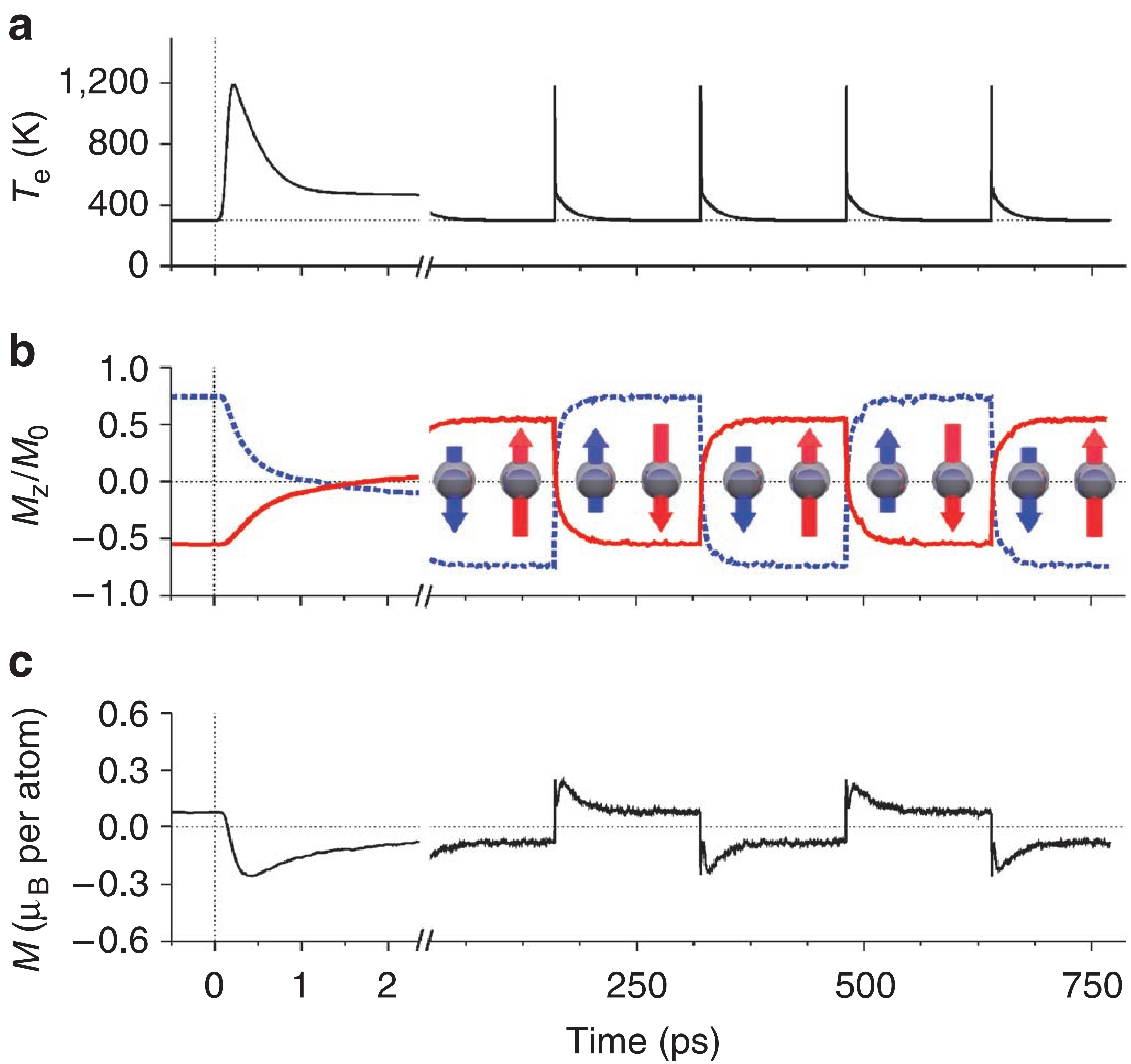
I. Radu *et al*, *Nature* **472**, 205–208 (2011)

Ultrafast magnetization dynamics simulated with atomistic spin model



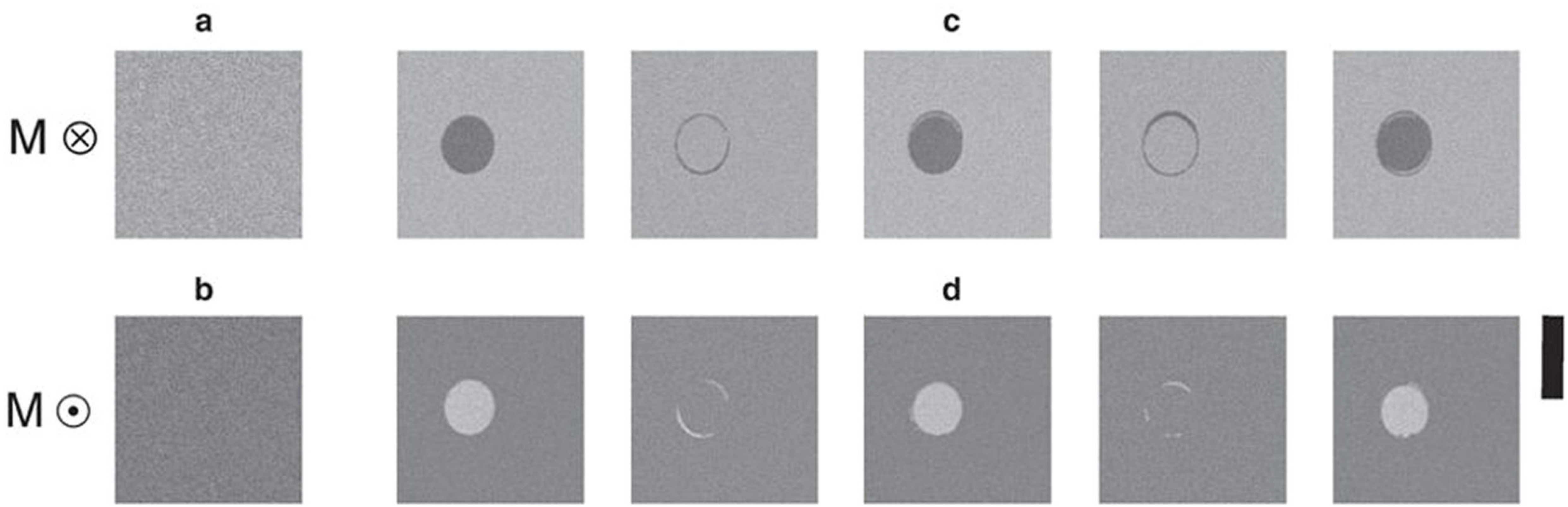
I. Radu *et al*, *Nature* **472**, 205–208 (2011)

Atomistic prediction of heat induced switching



T. Ostler *et al*, *Nat. Commun.* ***3***, *666* (2012)

Experimental confirmation of heat-induced switching

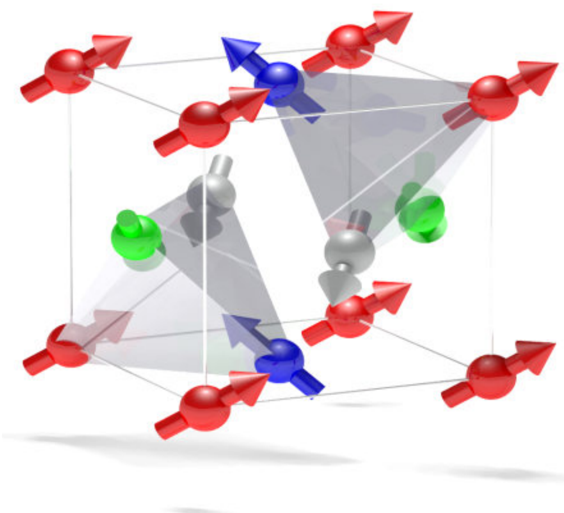


T. Ostler *et al*, *Nat. Commun.* ***3***, *666* (2012)

Summary

- Introduced the basic background of Landau-Lifshitz-Bloch micromagnetics

- Presented simulations of the static and dynamic properties of more complex magnets



- Thermodynamics is a significant and important contribution to ultrafast magnetic processes