Introduction to microscopic three temperature models for the description of the ultrafast magnetization dynamics

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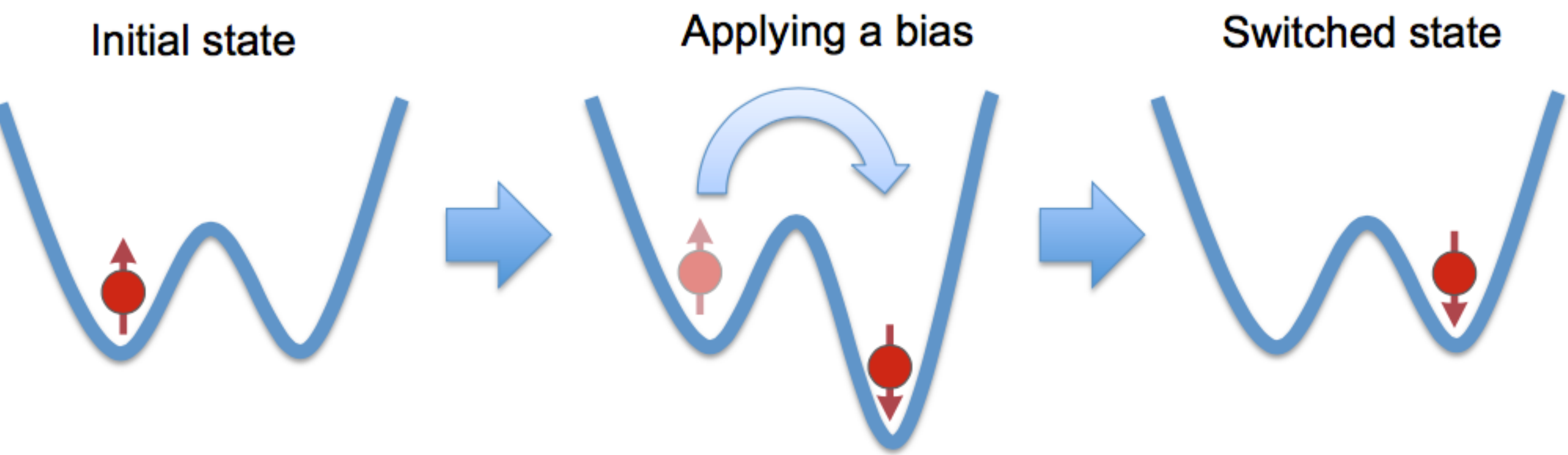
CRC / TRR 227
Ultrafast Spin Dynamics

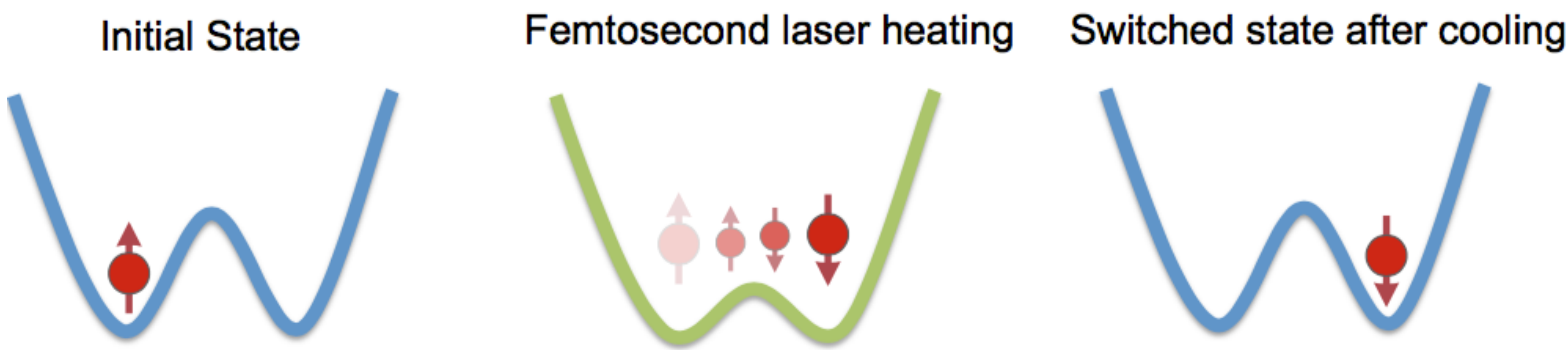
How do we switch a magnet?

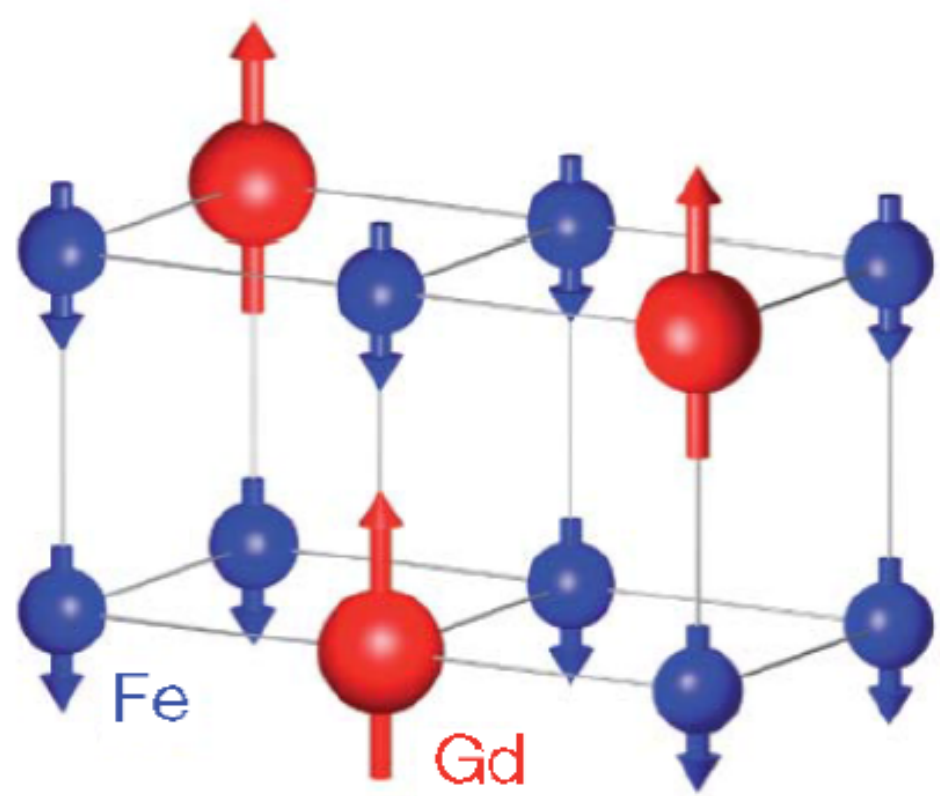


We apply a bias to break the symmetry of the system.

- Magnetic or electric fields
- spin injection/ spin transfer torque

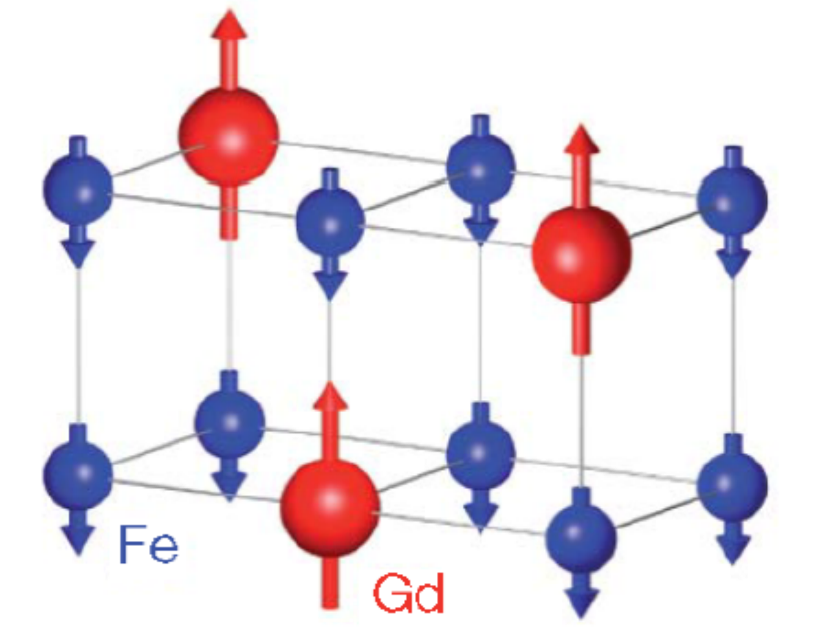
Ultrafast switching using heating

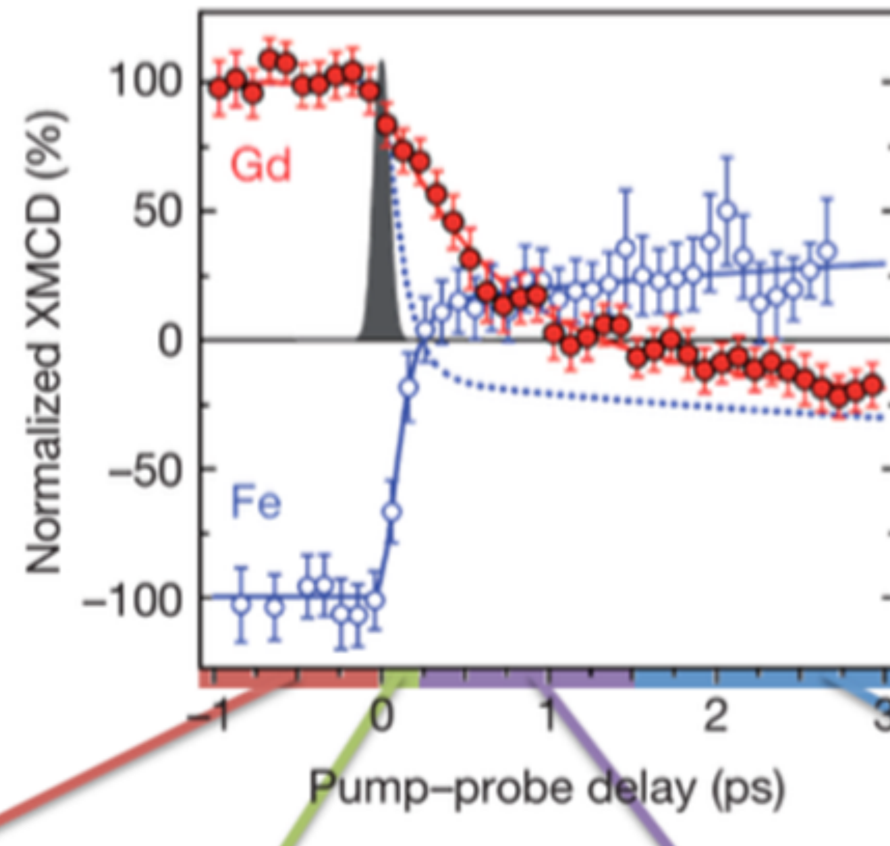




T. Ostler, et al, Nat. Comm. **3,** 666 (2012)

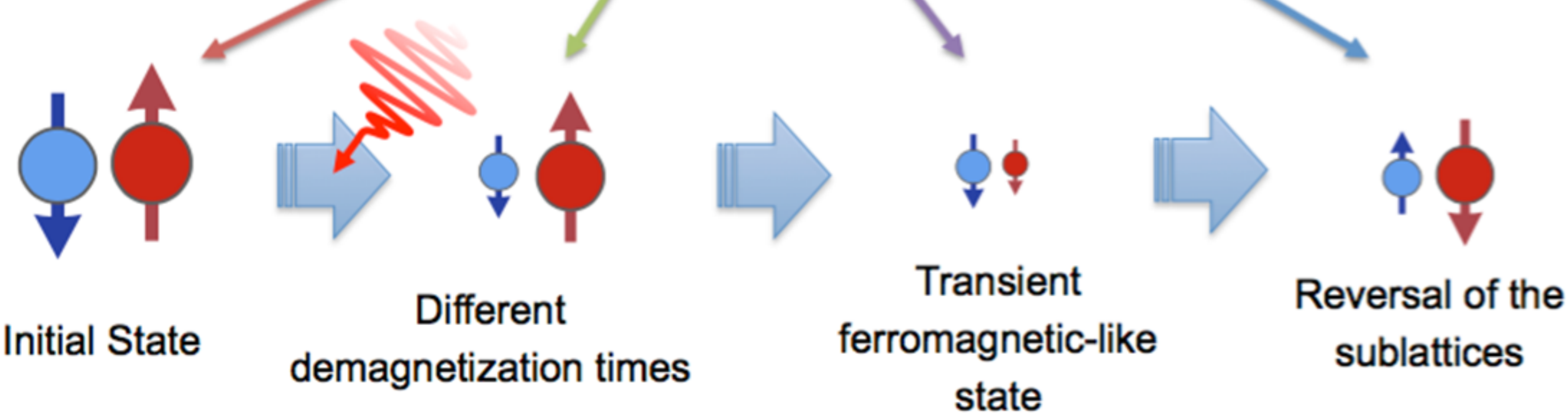
Element-specific demagnetization times



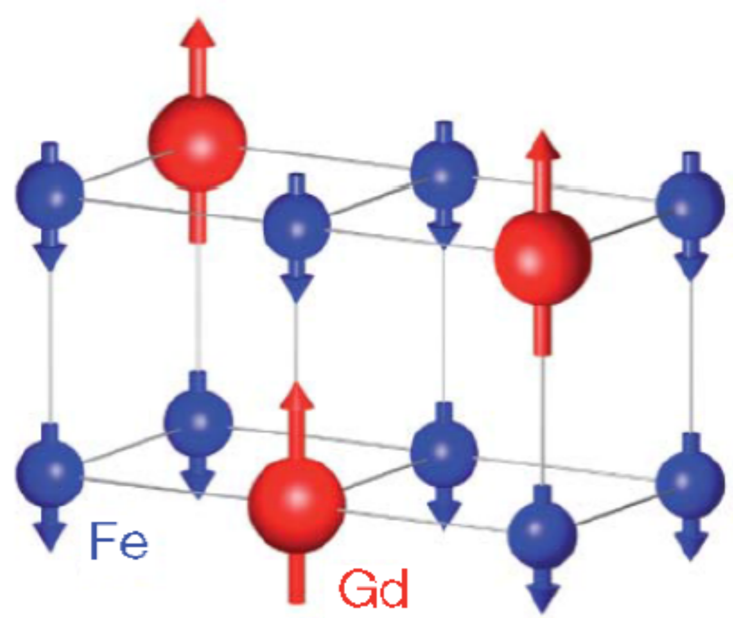


I. Radu et al., Nature **472** 205 (2011)

Element-resolved dynamics of the Fe and Gd magnetic moments measured by time-resolved XMCD with femtosecond time-resolution.



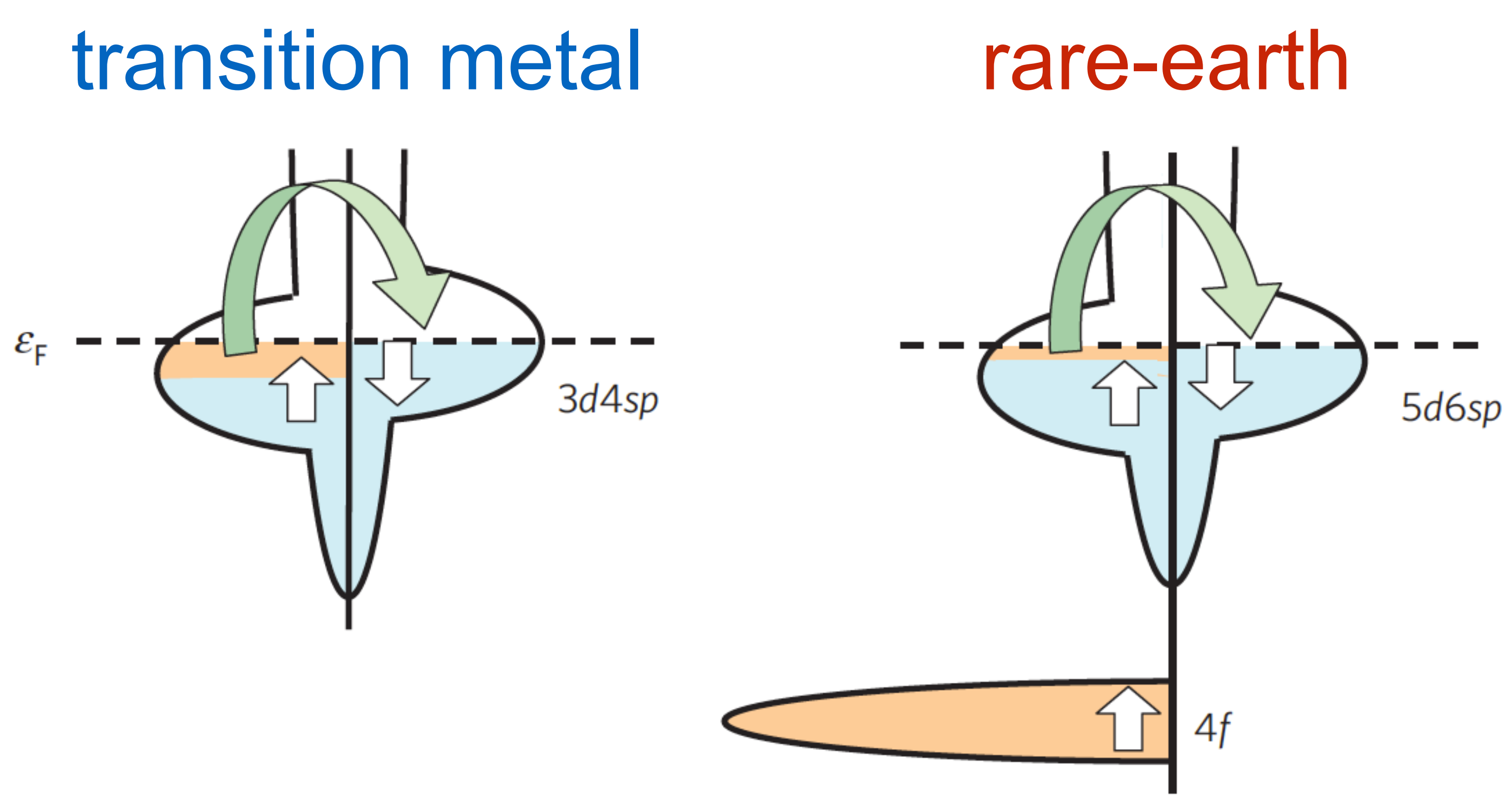
Ultrafast switching of the magnetic order



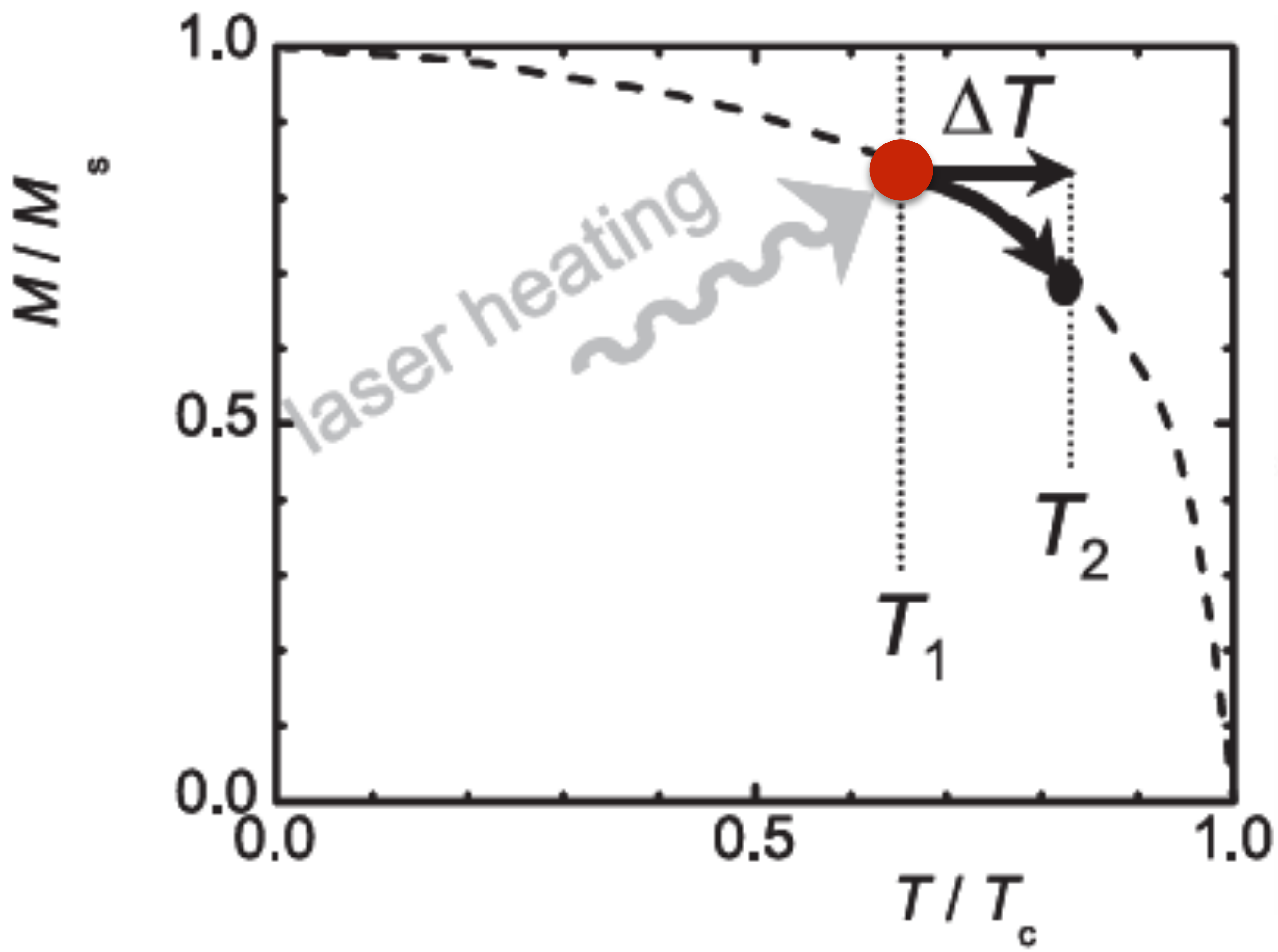
Gd rare-earth metal

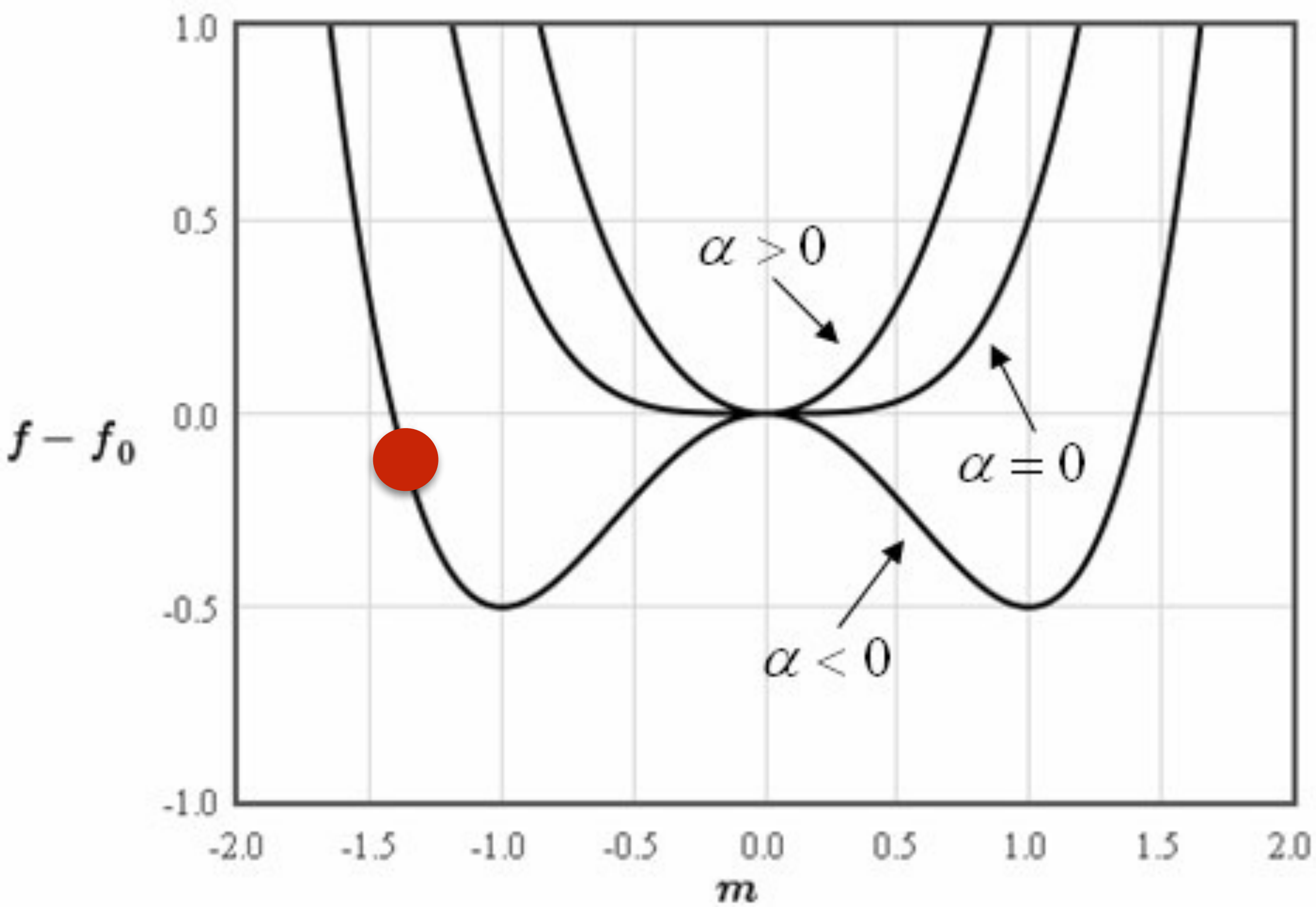
Fe transition metal

Response of the magnetic order to heat pulses

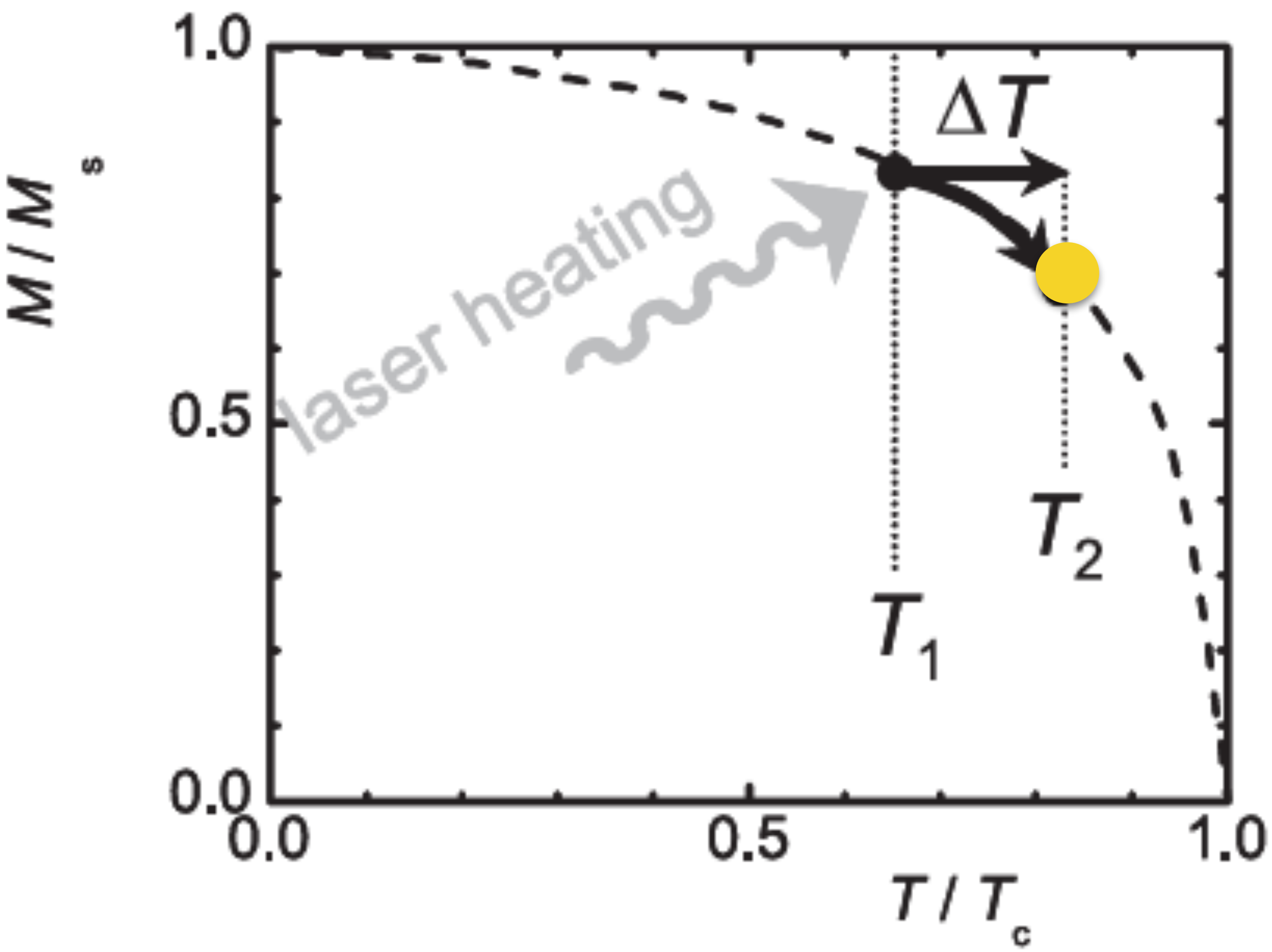


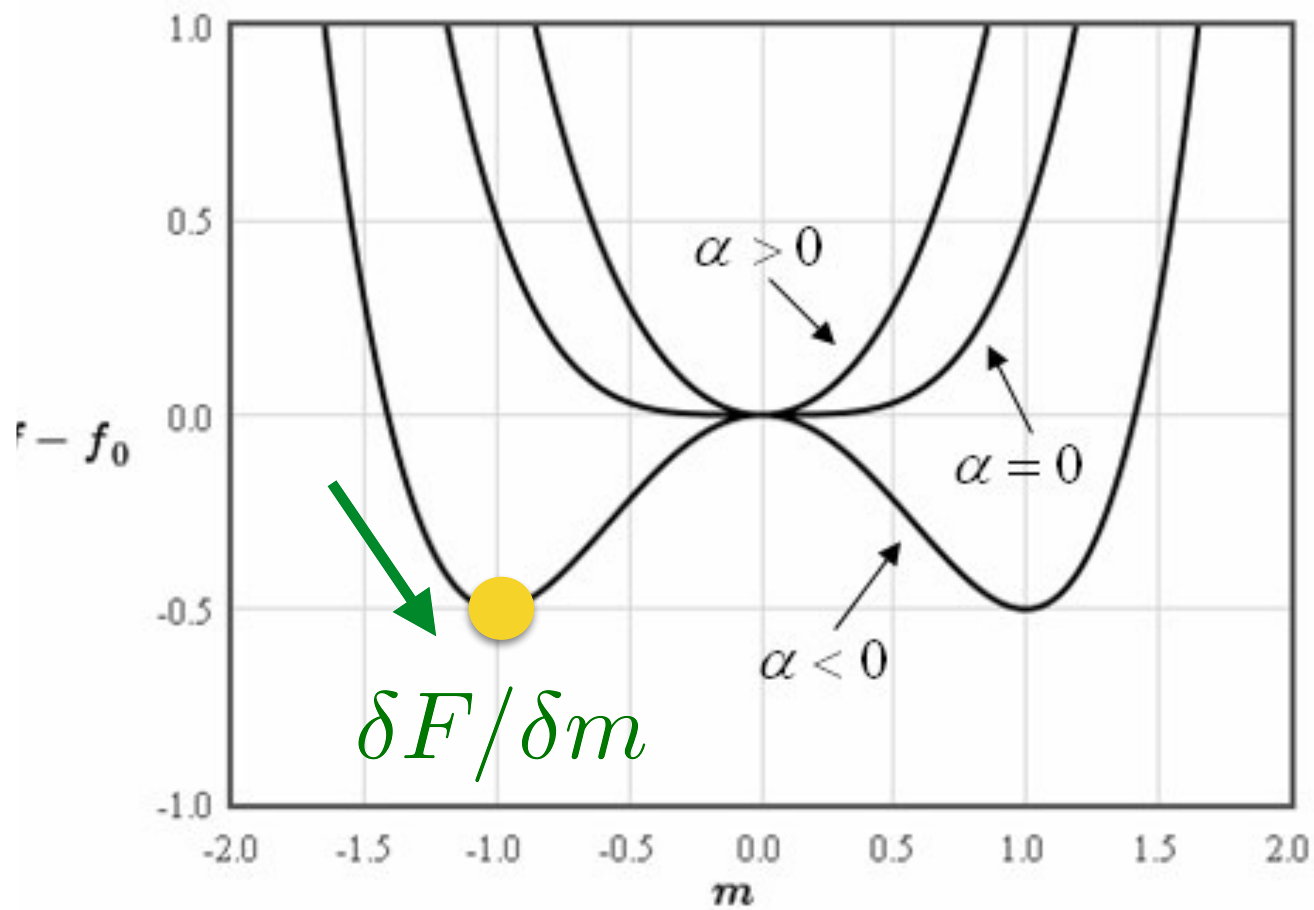
Magnetization dynamics driven by heating





Magnetization dynamics driven by heating





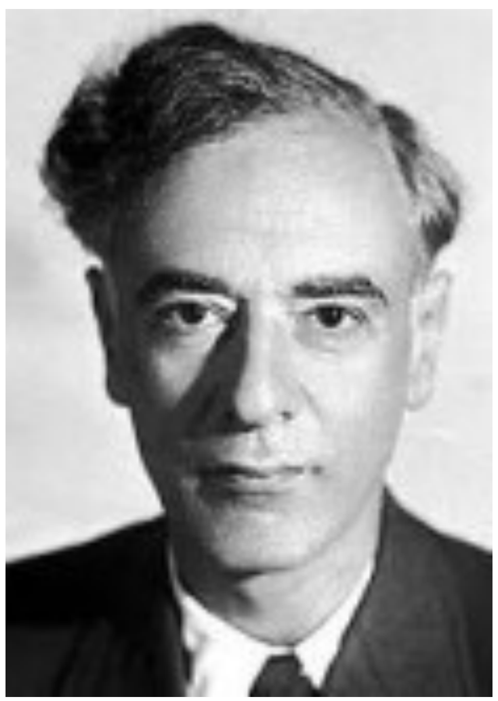
Magnetization dynamics

$$\boxed{\frac{dm}{dt} = \lambda(-\frac{\delta F}{\delta m})}$$

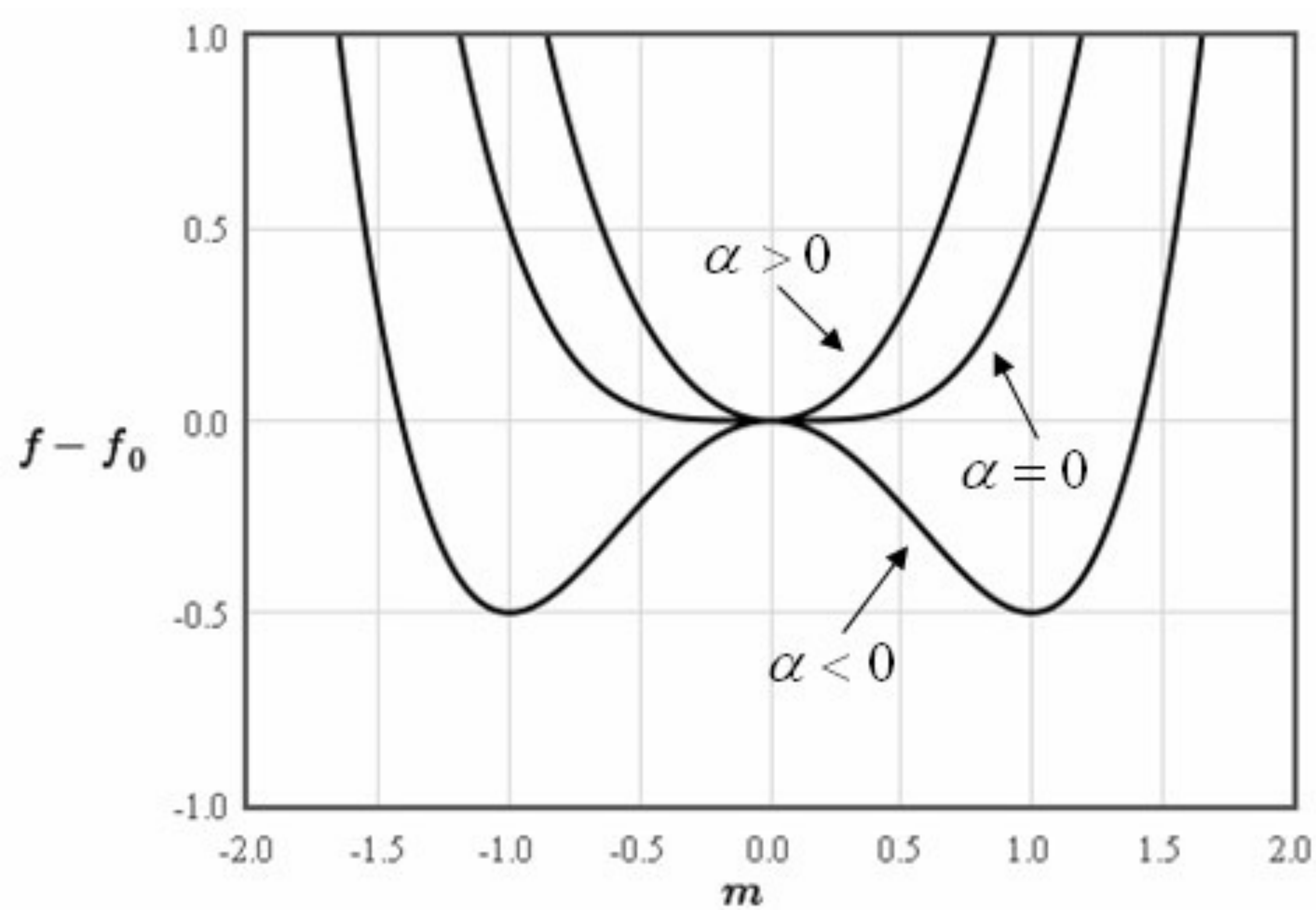
Magnetic free energy: Landau approach

Landau realised that near a phase transition:

- an approximate form for the free energy
- can be constructed
- without first calculating the microscopic states.



$$f(T) = f_0 + \alpha m^2 + \frac{1}{2}\beta m^4$$



Magnetic free energy: Landau approach

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- an approximate form for the free energy
- can be constructed
- without first calculating the microscopic states.

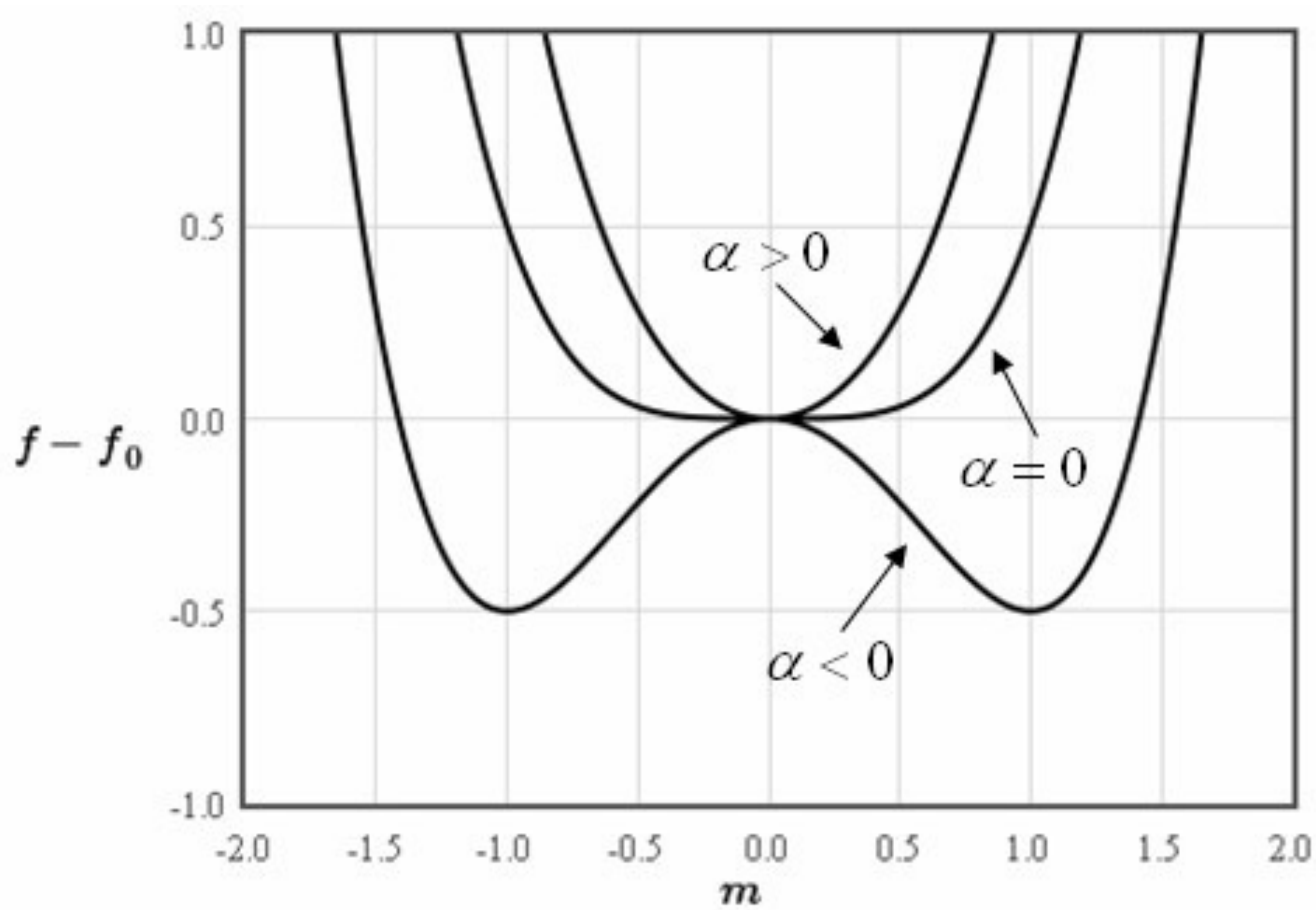
$$f(T) = f_0 + \alpha m^2 + \frac{1}{2}\beta m^4$$

<u>transition:</u>

$$\alpha = \alpha_0 (T - T_C)$$

<u>finite minimum</u>

$$\beta > 0$$



$$f(T) = f_0 + \alpha_0 (T - T_C) m^2 + \frac{1}{2}\beta m^4$$

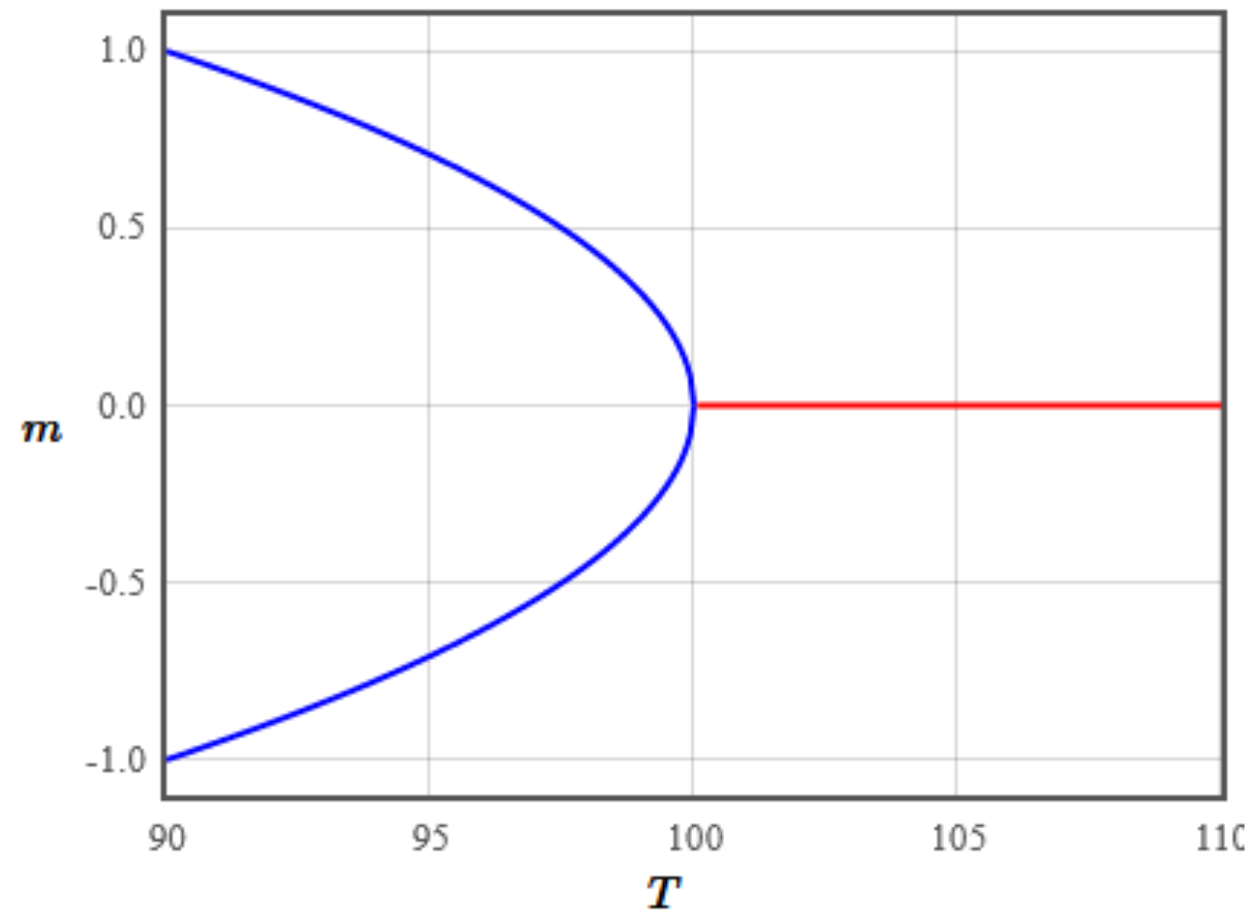
Magnetic free energy: Landau approach

from the free energy

$$\boxed{f(T) = f_0 + \alpha_0 (T - T_C) m^2 + \frac{1}{2}\beta m^4}$$

spontaneous magnetisation

$$\frac{df}{dm} = 0 \qquad m_e = \pm\sqrt{\frac{\alpha_0 (T_C - T)}{\beta}}$$



we can express one parameter in terms of the others, for example:

$$\beta = \frac{\alpha_0 (T_C - T)}{m_e^2}$$

we rewrite the free energy as

$$\boxed{f(T) = f_0 + \alpha_0 (T - T_C) m^2 - \frac{1}{2}\frac{\alpha_0 (T - T_C)}{m_e^2} m^4}$$

Magnetic free energy: Landau approach

One can include the effect of an external magnetic field *B*

$$f(T) = f_0 + \boxed{\alpha_0(T - T_C)}\,m^2 - \frac{1}{2}\frac{\boxed{\alpha_0(T - T_C)}}{m_e^2}m^4 - mB$$

susceptibility *m/B* for B —> 0:

$$\frac{df}{dm} = 0 \quad \Rightarrow \quad 2\alpha_0(T - T_C)m + 2\beta m^3 - B = 0$$

$$\Rightarrow \quad 2\alpha(T)\frac{dm}{dB} + 6\beta m^2\frac{dm}{dB} - 1 = 0$$

$$\Rightarrow \quad \boxed{\begin{aligned}\chi = \left(\frac{dm}{dB}\right)_{B=0} &= \frac{1}{2\alpha(T) + 6\beta m^2} \\ &= \frac{1}{2\alpha(T) + 6\beta(-\alpha(T)/\beta)} = -\frac{1}{4\alpha(T)}\end{aligned}}$$

$$\boxed{f(T) = f_0 - \frac{1}{4\chi}m^2 + \frac{1}{4\chi m_e^2}m^4 - mB}$$

Magnetic free energy: Landau approach

One can include the effect of an external magnetic field *B*

$$f(T) = f_0 - \frac{1}{4\chi}m^2 + \frac{1}{8\chi m_e^2}m^4 - mB$$

second term multiplied and divided: $2m_e^2$

$$f(T) = f_0 - 2m_e^2\frac{1}{4\chi 2m_e^2}m^2 + \frac{1}{8\chi m_e^2}m^4 - mB$$

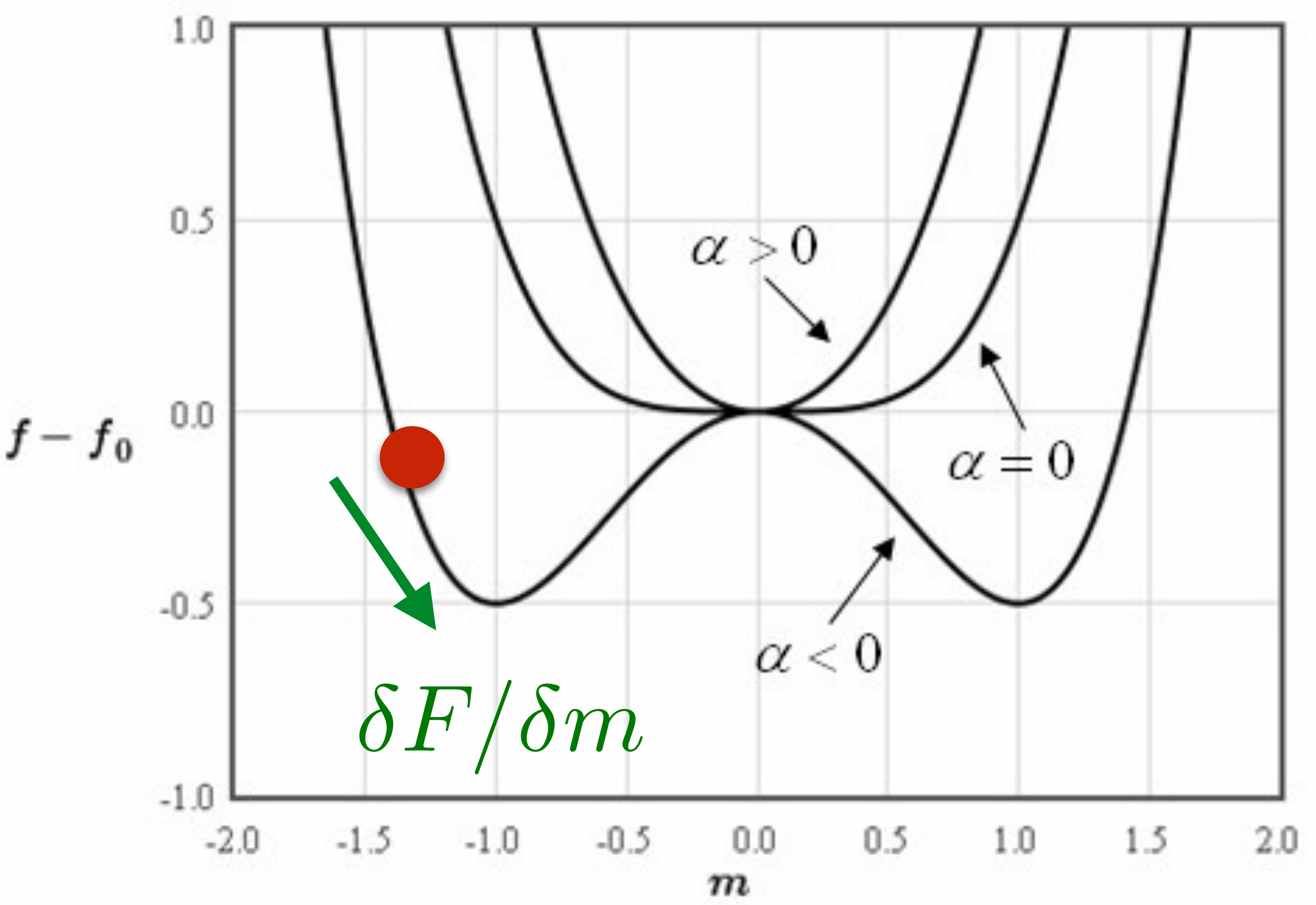
we use the identity:

$$(m^2 - m_e^2)^2 = m^4 - 2m_e m^2 + m_e^4$$

$$\Rightarrow \quad \boxed{f(T) = \tilde{f}_0 + \frac{1}{8\chi m_e^2}(m^2 - m_e^2)^2 - mB}$$

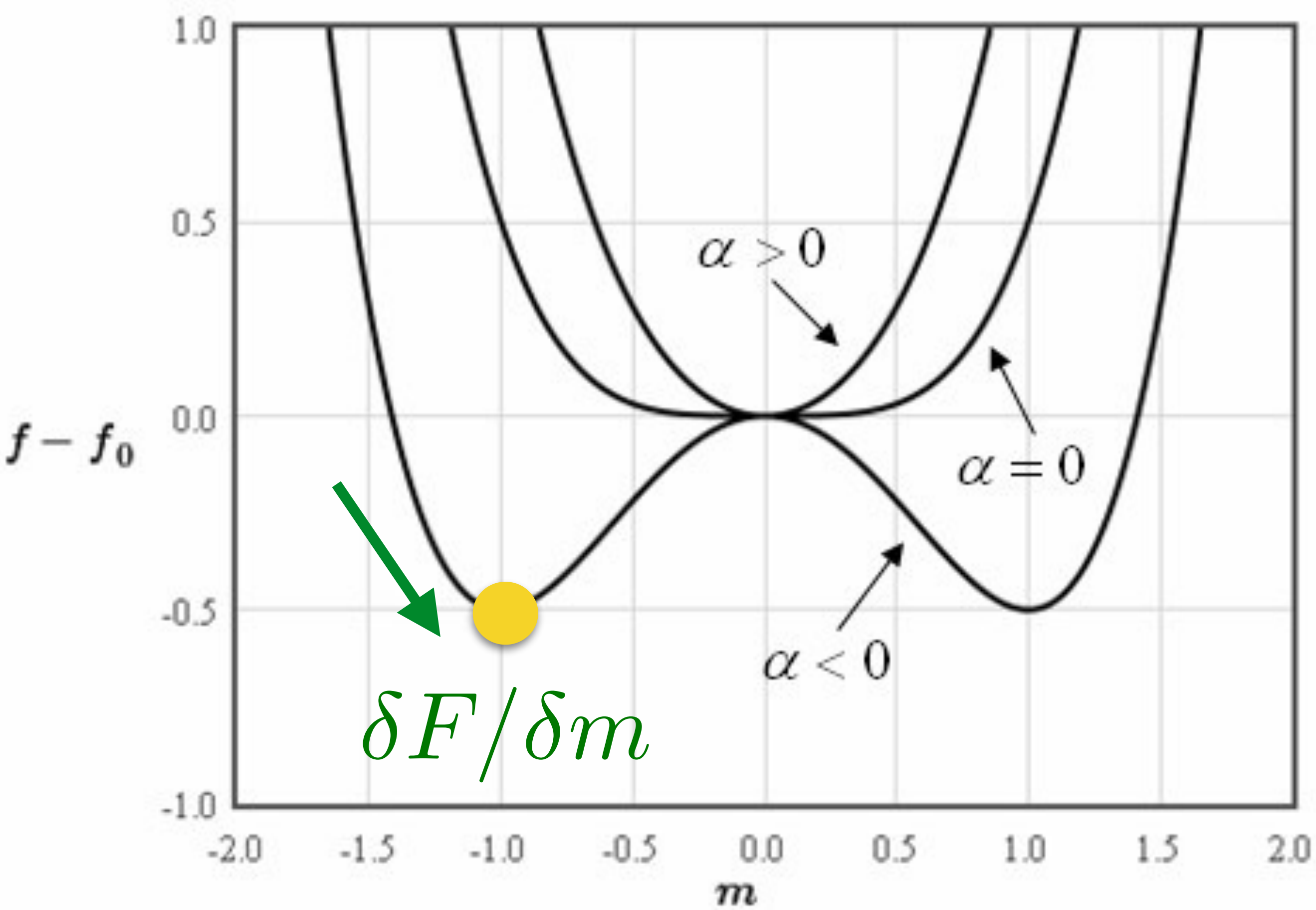
Magnetization dynamics

$$f(T) = \widetilde{f}_0 + \frac{1}{8\chi m_e^2}\left(m^2 - m_e^2\right)^2$$



Magnetization dynamics

$$\boxed{f(T) = \widetilde{f}_0 + \frac{1}{8\chi m_e^2}\left(m^2 - m_e^2\right)^2}$$



$$\frac{\delta f(T)}{\delta m} = \frac{2(2m)}{8\chi m_e^2}(m^2 - m_e^2)$$
$$= \frac{1}{2\chi}\left(\frac{m^2}{m_e^2} - 1\right)m$$

Magnetization dynamics

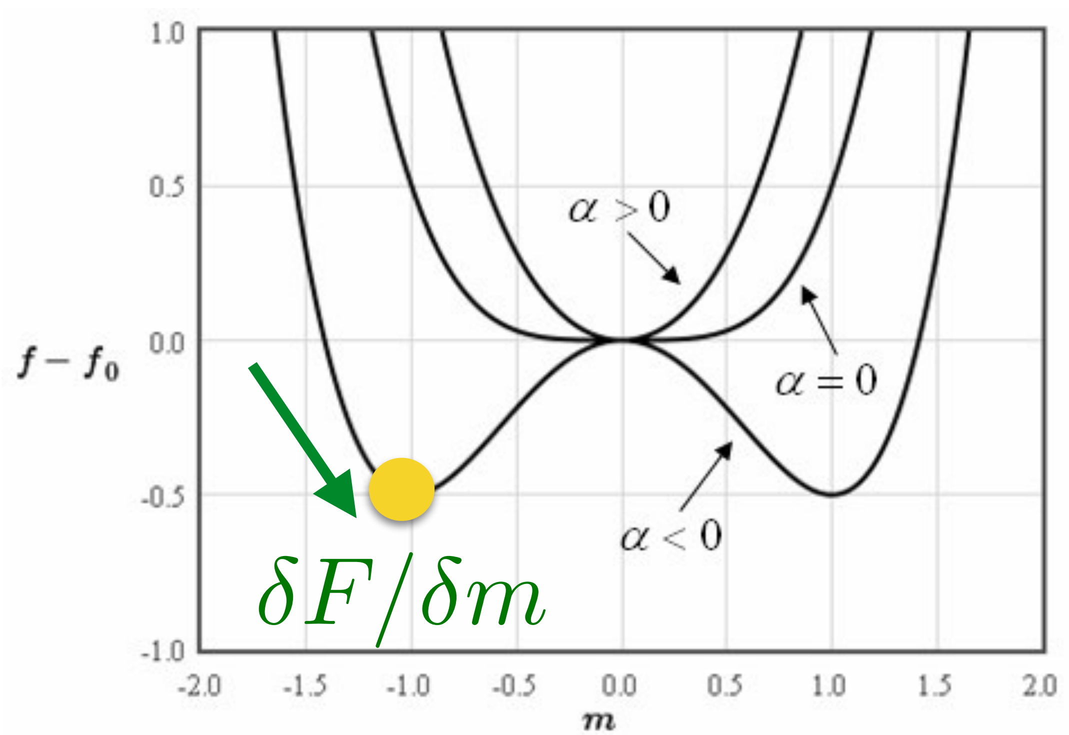
$$\boxed{\frac{dm}{dt} = \lambda(-\frac{\delta F}{\delta m})} \Rightarrow \boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right)m}$$

Magnetization dynamics

Magnetization dynamics

$$\boxed{\frac{dm}{dt} = \lambda(-\frac{\delta F}{\delta m})}$$

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right) m}$$



in the linear regime:

$$\delta m = m - m_e \ll m_e$$

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{\chi}(m_e - m)}$$

solution:

$$\frac{\Delta m(t)}{\Delta m(0)} = \frac{m(t) - m_e}{m(0) - m_e} = \exp(-t/\tau_{\rm de})$$

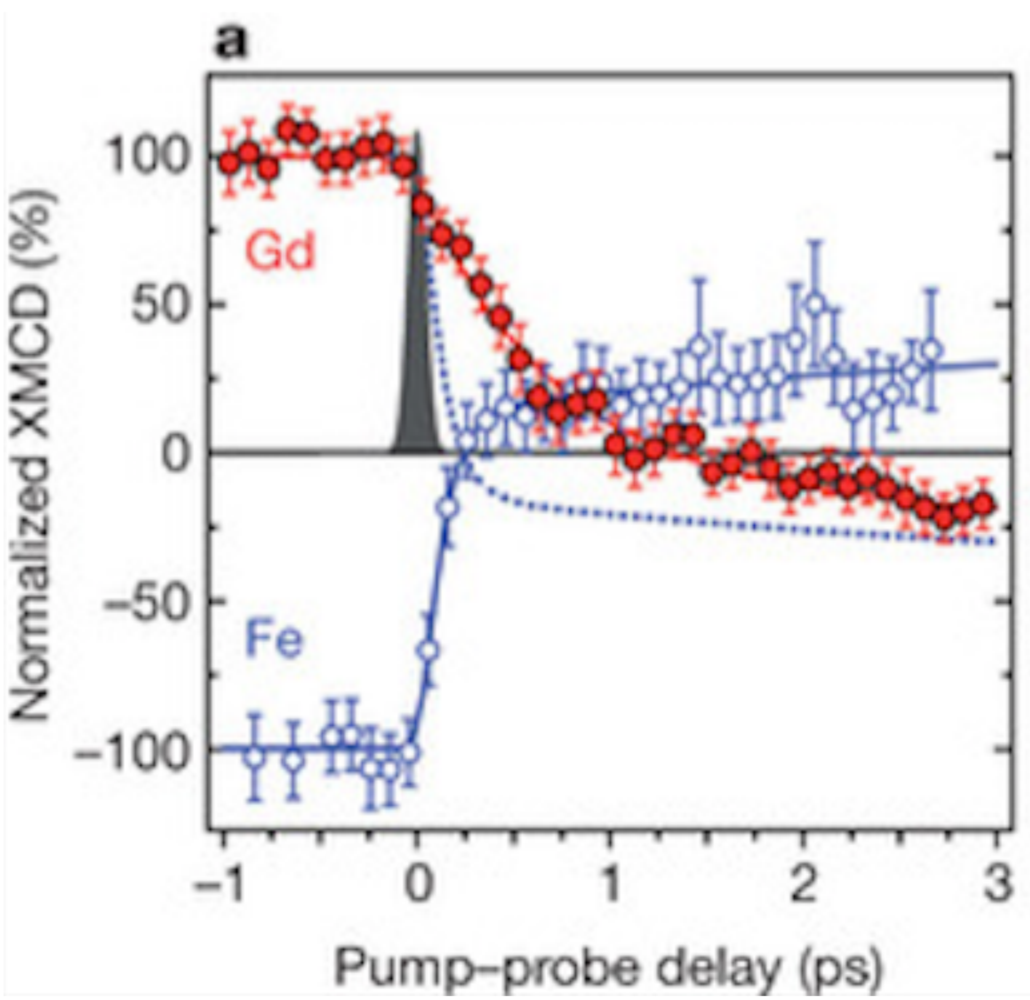
relax time:

$$\boxed{\tau_{\rm de} = \chi/\lambda}$$

Relaxation time

relax time:

$$\boxed{\tau_{\mathrm{de}} = \chi/\lambda}$$



Curie-Weiss law

$$\chi \sim \frac{\mu_{\mathrm{at}}}{T - T_c} = \left(\frac{\mu_{\mathrm{at}}}{T_c}\right)\left(\frac{1}{1 - T/T_c}\right)$$

Magnetic properties

Critical slowing down

Fe	$\mu_{\mathrm{at}} = 2.20\mu_{\mathrm{B}}, T_c = 1049\ K$
Gd	$\mu_{\mathrm{at}} = 7.5\mu_B, T_c = 293\ \mathrm{K}$

Relaxation time

relax time:

$$\boxed{\tau_{\mathrm{de}} = \chi/\lambda}$$

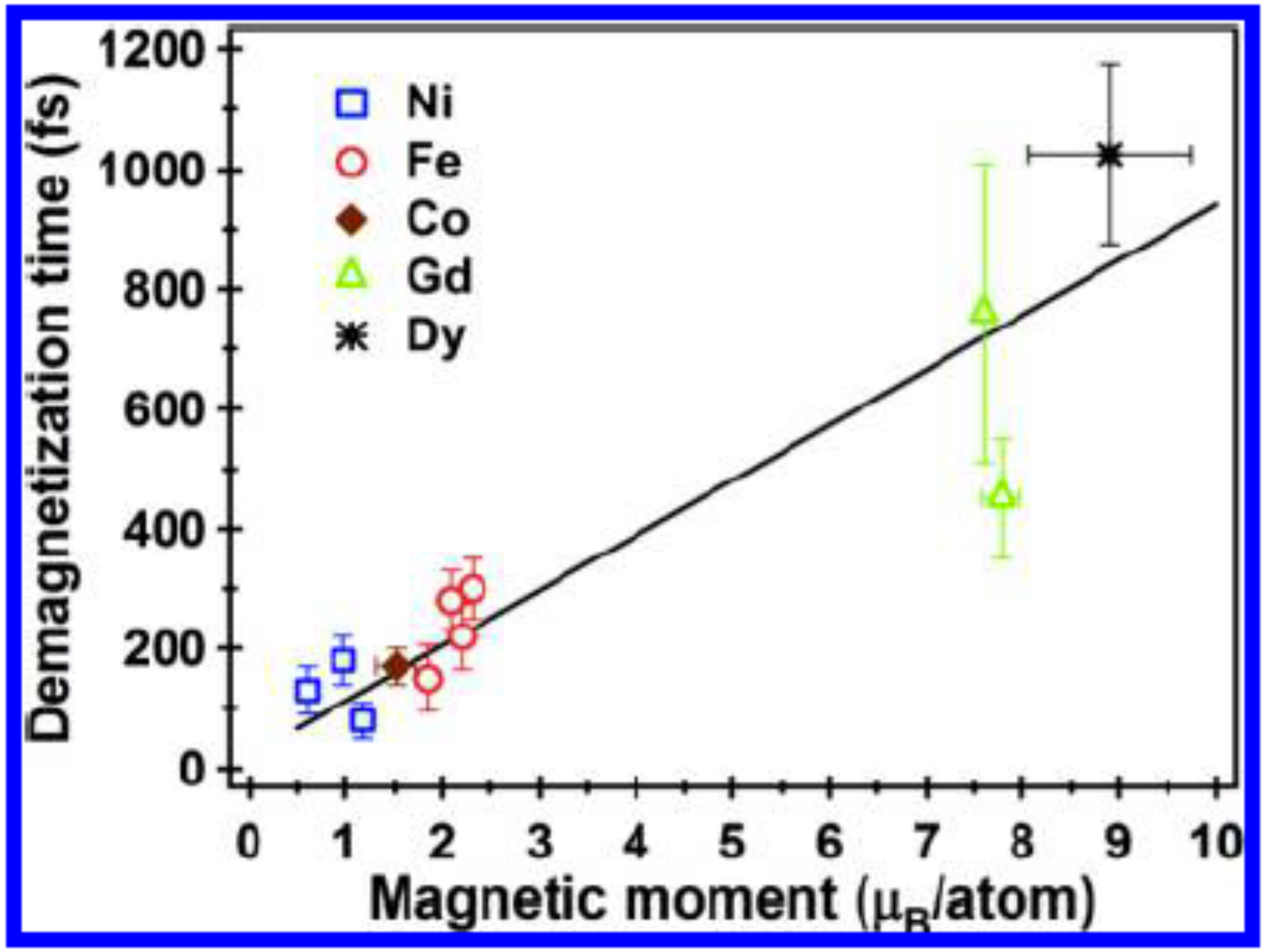
Curie-Weiss law

$$\chi \sim \frac{\mu_{\mathrm{at}}}{T - T_c} = \left(\frac{\mu_{\mathrm{at}}}{T_c}\right)\left(\frac{1}{1 - T/T_c}\right)$$

low-intermediate temperature limit:

$$\boxed{\tau_{\mathrm{de}} = \frac{\chi}{\lambda} \sim \frac{\mu_{\mathrm{at}}}{T_c}\frac{1}{\lambda}}$$

related to the microscopic details of angular momentum dissipation process



I. Radu et al. **5** 1550004 SPIN (2015)

Summary

Magnetization dynamics from Landau

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right)m}$$

- parameters: **experimental** values.
- parameters **mean-field theory**.

One-spin problem

Let us consider one spin ***S*** in a magnetic field ***H***

$$E = -\mathbf{H} \cdot \mathbf{S}$$

Partition function: $\mathcal{Z} = \sum_i \exp(\beta \mu_0 H S)$

for S=1/2

$$\mathcal{Z} = \exp(\beta \mu_0 H/2) + \exp(-\beta \mu_0 H/2)$$

S = 1/2

$$n_\uparrow = \frac{\exp(\beta \mu_0 H/2)}{\exp(\beta \mu_0 H/2) + \exp(-\beta \mu_0 H/2)}$$

S = - 1/2

$$n_\downarrow = \frac{\exp(-\beta \mu_0 H/2)}{\exp(\beta \mu_0 H/2) + \exp(-\beta \mu_0 H/2)}$$

induced magnetization:

$$\boxed{m = n_\uparrow - n_\downarrow = \tanh(\beta \mu_0 H/2)}$$

Mean-field theory

Heisenberg Hamiltonian

$$\mathcal{H} = -J \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i \mathbf{S}_i \cdot \mathbf{H}$$

mean field approximation (MFA)

$$\mathcal{H}_{\mathrm{MFA,i}} = -\mathbf{S}_i \left(J \sum_j \langle S \rangle + H \right) = -\mathbf{H}_{\mathrm{MFA}} \cdot \mathbf{S}_i$$

one-spin problem with effective MFA field

$$\mathrm{H}^{\mathrm{MFA}} = \frac{zJ}{\mu_0} m + H_z$$

where $m = \langle S \rangle$

z number of neighbours (6 in sc lattice)

$$\boxed{m = \tanh(\beta(zJm + \mu_0 H_z))}$$

Mean-field theory

$$\boxed{m = \tanh(\beta(zJm + \mu_0 H_z))}$$

Magnetization dynamics

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right) m}$$

Equilibrium magnetization

$$m_e = \tanh(\beta(zJm_e + \mu_0 H_z))$$

close to Curie temperature: $m_e \approx \beta_c z J m_e \rightarrow k_B T_c = zJ$

$$\boxed{m_e = \tanh\left(\frac{T_c}{T} m_e\right)}$$

exercise: Zero-field susceptibility

$$\boxed{\chi = \frac{dm}{dH_z} = \frac{\mu_0}{zJ}\frac{\beta zJ \tanh'(x)}{1 - \beta zJ \tanh'(x)}}$$

Mean-field theory

Magnetization dynamics

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right) m}$$

$$\boxed{m_e = \tanh\left(\frac{T_c}{T} m_e\right)}$$

small deviations: $\delta m = m - m_e \ll m_e$

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{\chi}(m_e - m)}$$

to account for non-equilibrium situations we rather consider $m_e \to \widetilde{m}$

$$\widetilde{m} = \tanh\left(\frac{T_c}{T} m\right)$$

the equation of motion:

$$\begin{aligned} \frac{dm}{dt} &= \frac{\lambda}{\chi}(\widetilde{m} - m) \\ &= \frac{\lambda}{\chi}\widetilde{m}(1 - \frac{m}{\widetilde{m}}) \\ &\approx \frac{\lambda}{\chi} m(1 - \frac{m}{\widetilde{m}}) \end{aligned}$$

$$\Rightarrow \quad \boxed{\frac{dm}{dt} = \frac{\lambda}{\chi} m(1 - m\coth\left(\frac{T_c}{T} m\right))}$$

Summary

Magnetization dynamics from Landau

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right) m}$$

- parameters: **experimental** values.
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Magnetization dynamics S=1/2

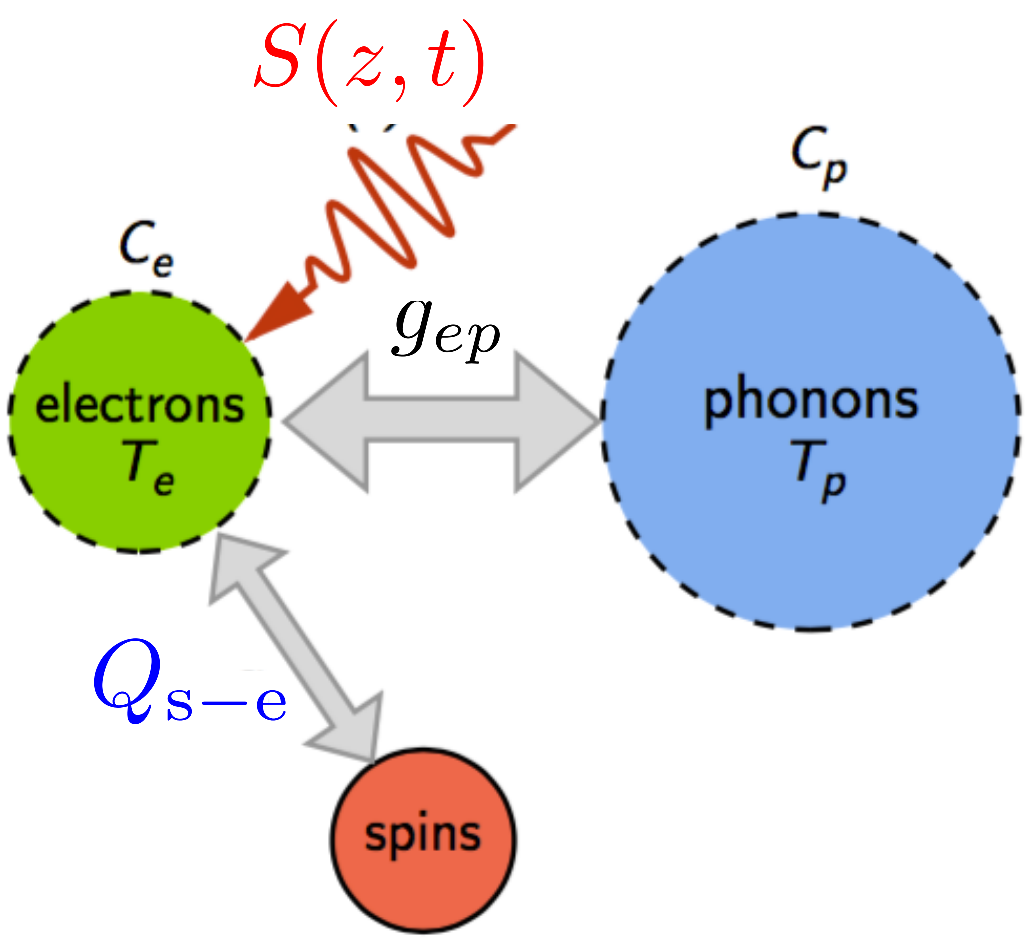
$$\boxed{\frac{dm}{dt} = \frac{\lambda}{\chi} m(1 - m\coth\left(\frac{T_c}{T} m\right))}$$

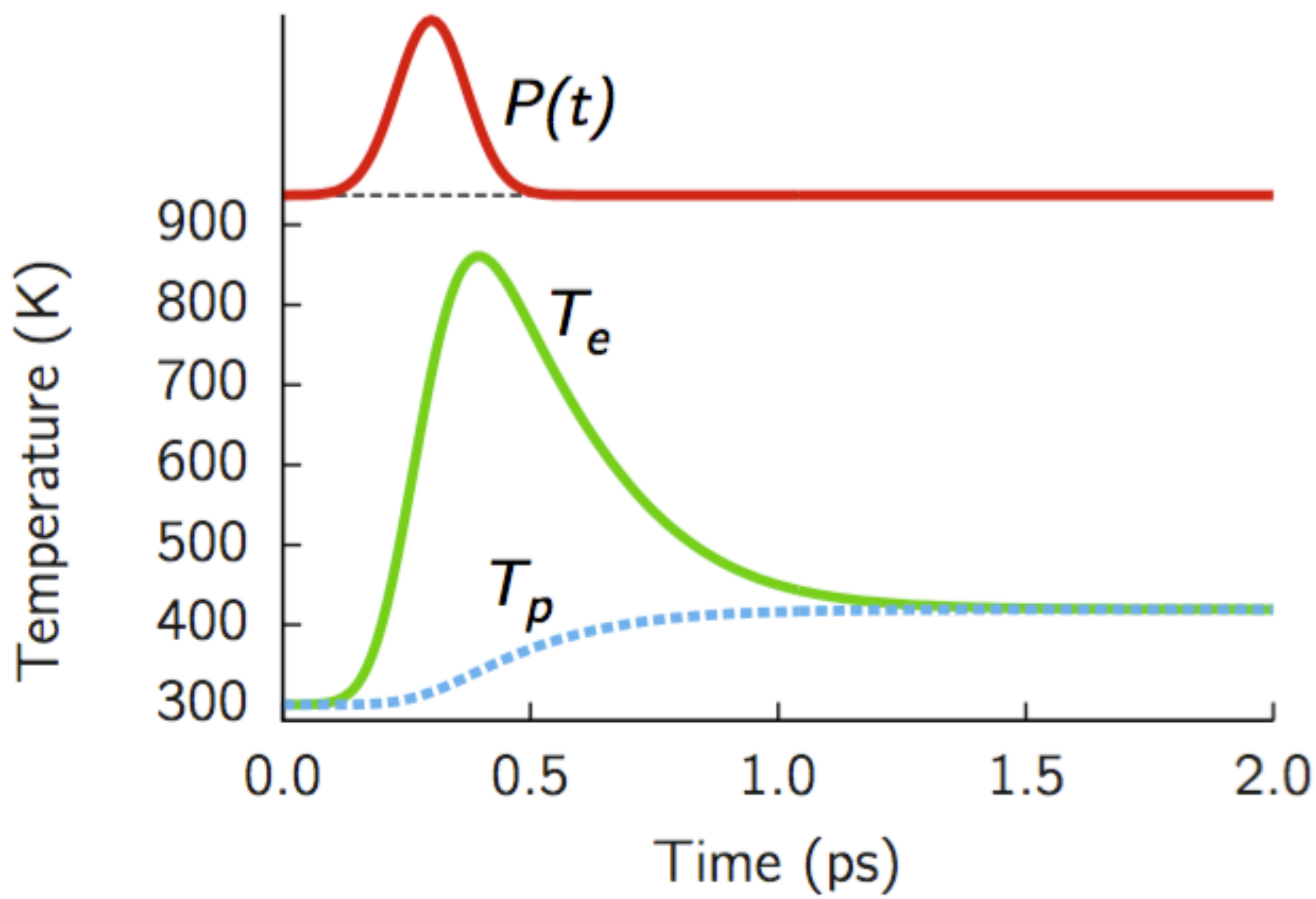
Microscopic three temperature model (M3TM)

non-equilibrium between electrons and phonons: 2TM

$$C_e \frac{dT_e}{dt} = g_{ep}(T_e - T_p) + \kappa \nabla T_e + S(z,t) + Q_{\mathrm{e-s}}$$

$$C_p \frac{dT_p}{dt} = g_{ep}(T_p - T_e)$$



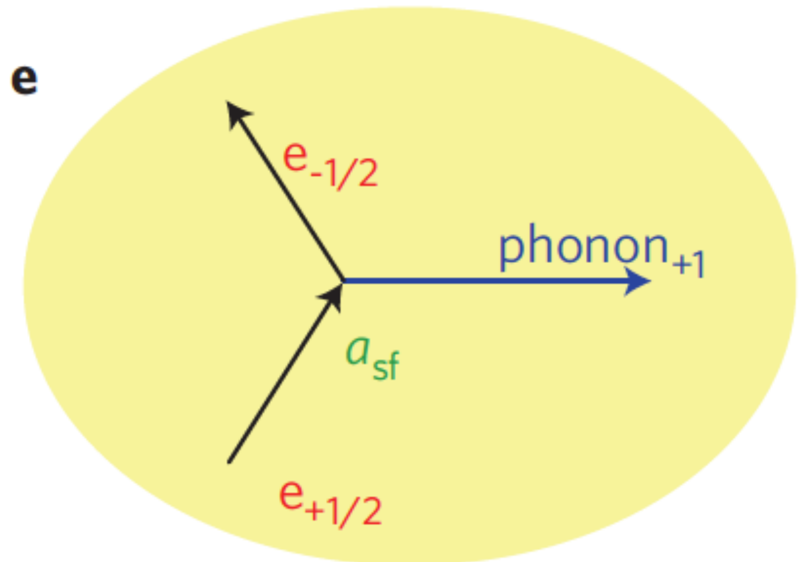


U. Bovensiepen **19**, JCMP (2007)

Microscopic three temperature model (M3TM)

Spin-electron-phonon coupling potential

$$\mathcal{H}_{e-s} = \sqrt{\frac{a_{sf}}{D_s}} \frac{\lambda_{ep}}{N^{3/2}} \sum_{k,k',q,j} c_k^\dagger c_{k'} \left(S_+ + S_-\right) \left(a^\dagger + a_q\right)$$



relaxation rate:

$$R = \frac{8 a_{\mathrm{sf}} g_{\mathrm{ep}} k_B T_C^2}{\mu_{\mathrm{at}} E_D^2}$$

E_{D} energy of phonons

a_{sf} spin-flip probability

g_{ep} electron-phonon coupling

$$\frac{dm}{dt} = R \frac{T_p}{T_c} m \left(1 - m \coth\left(\frac{T_c}{T_e} m\right)\right)$$

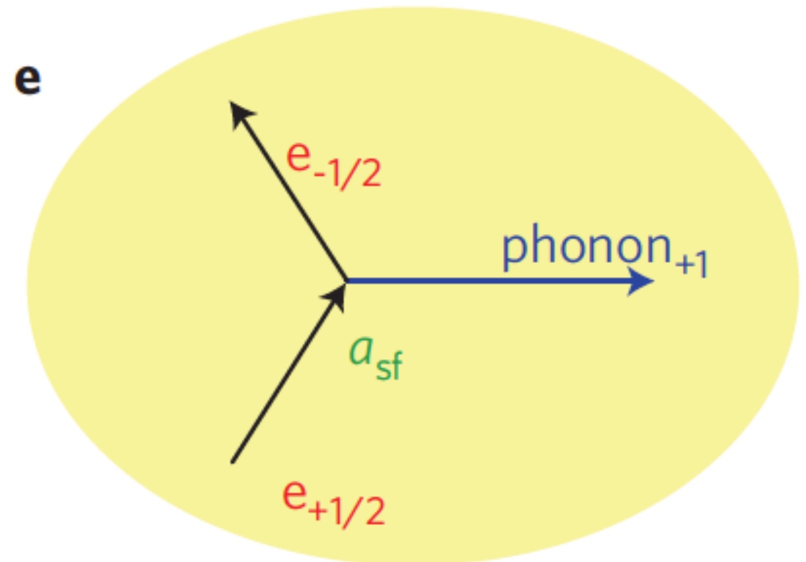
B. Koopmans et al.
Nature Materials **9** 259, (2010)

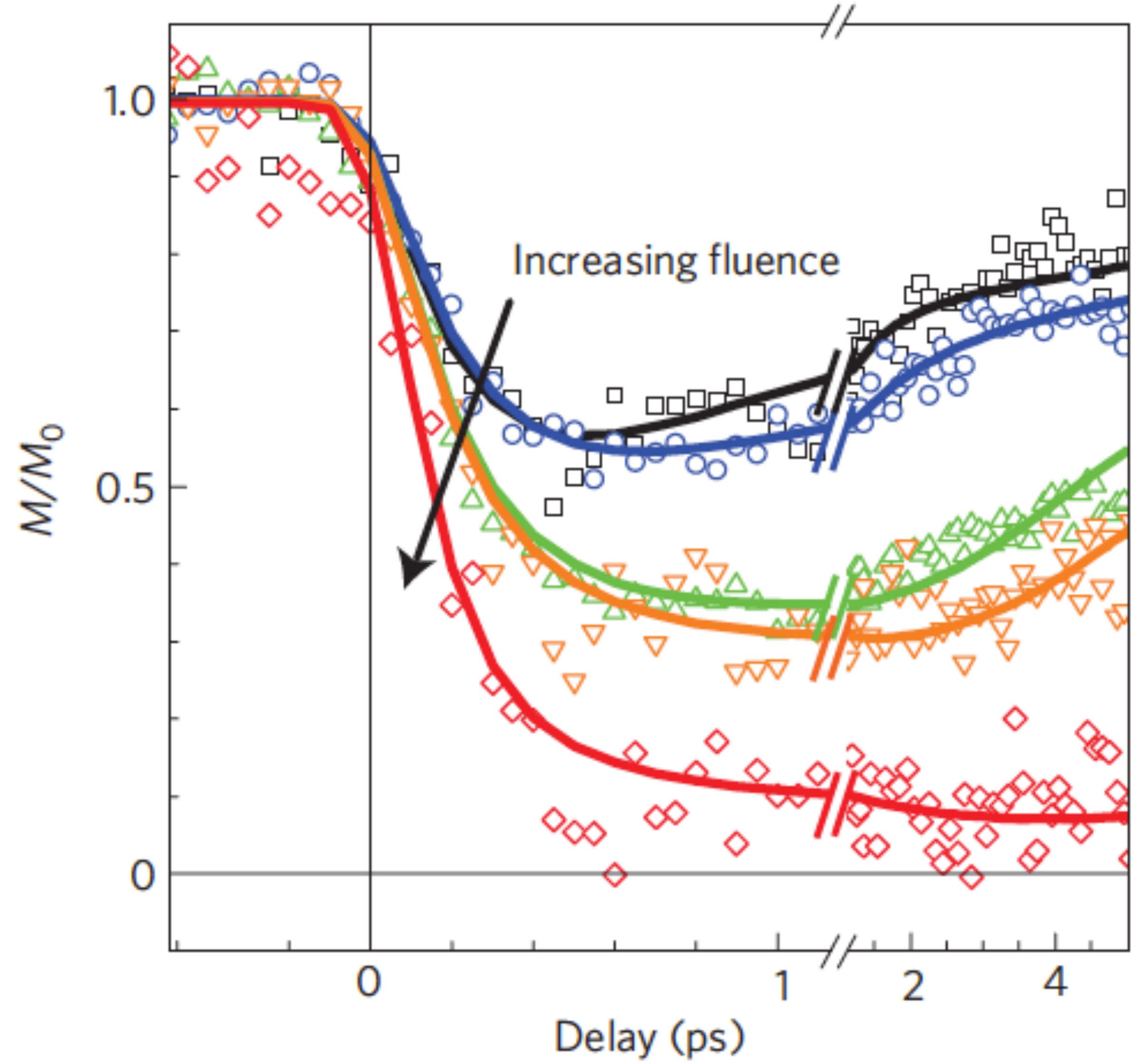
Project 1: Derivation of M3TM

Microscopic three temperature model (M3TM)

Spin-electron-phonon coupling potential

$$\boxed{\mathcal{H}_{e-s} = \sqrt{\frac{a_{sf}}{D_s}} \frac{\lambda_{ep}}{N^{3/2}} \sum_{k,k',q,j} c_k^\dagger c_{k'} \left(S_+ + S_-\right)\left(a^\dagger + a_q\right)}$$





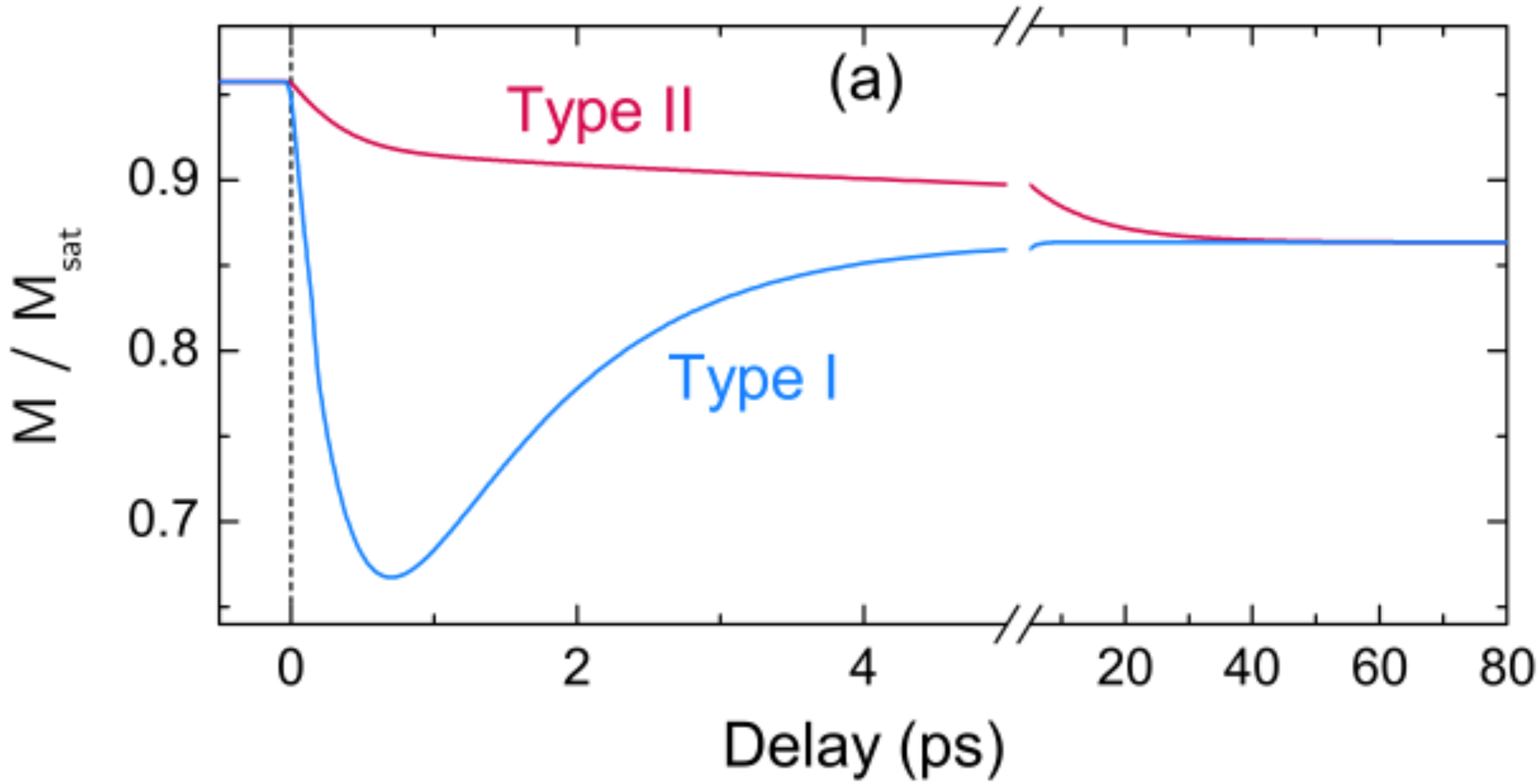
$$\boxed{\frac{dm}{dt} = R\frac{T_p}{T_c} m\left(1 - m\coth\left(\frac{T_c}{T_e} m\right)\right)}$$

B. Koopmans et al.
Nature Materials **9** 259, (2010)

transition from type I to type II dynamics

type I to type II dynamics

Roth et al. PRX **2**, 021006 (2012)



type I

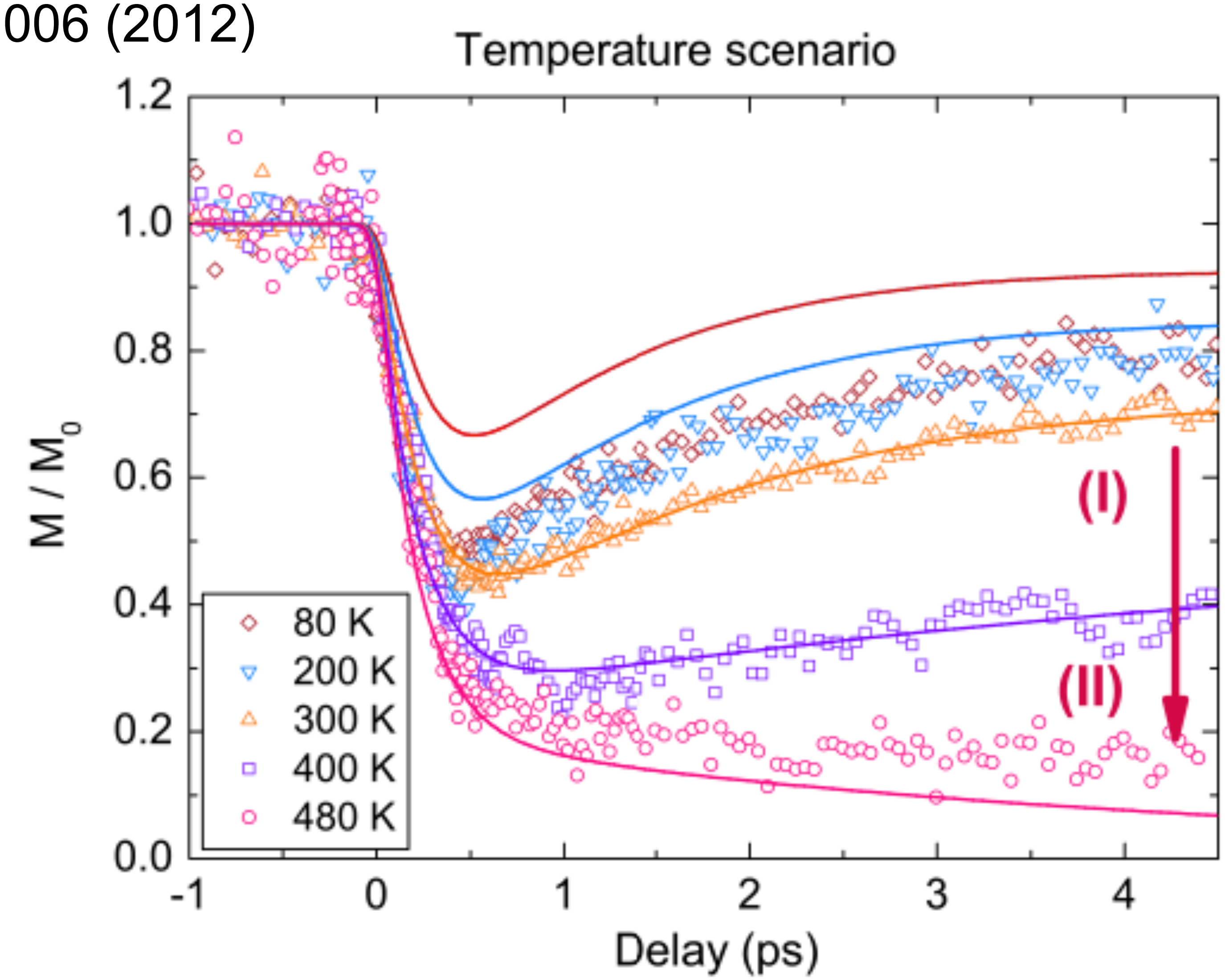
- fast demagnetization dynamics, 100s femtoseconds
- recovery of the magnetization, 1-10s picoseconds

type II

- first demagnetization dynamics, picoseconds
- second demagnetization dynamics, 10s picoseconds

type I to type II dynamics

Roth et al. PRX **2**, 021006 (2012)



type I

- magnetization dynamics faster than electron-phonon eq.

type II

- **slowing down** of the magnetization dynamics as ambient temperature increases

Summary

Magnetization dynamics from Landau

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right) m}$$

Magnetization dynamics S=1/2

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{\chi} m(1 - m \coth\left(\frac{T_c}{T} m\right))}$$

microscopic three temperature model

$$\boxed{\frac{dm}{dt} = R\frac{T_p}{T_c} m(1 - m \coth\left(\frac{T_c}{T_e} m\right))}$$

$$\boxed{R = \frac{8 a_{\mathrm{sf}} g_{\mathrm{ep}} k_B T_C^2}{\mu_{\mathrm{at}} E_D^2}}$$

Summary

Magnetization dynamics from Landau

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right) m}$$

microscopic derivation

$$\lambda(T) = \lambda \frac{2T}{3T_c} \frac{2q}{\sinh(2q)} \qquad q = \frac{T_c}{T} m$$

Quantum-Landau-Lifshitz-Bloch model

$$\boxed{\frac{dm}{dt} = \lambda_0 \frac{2T}{3T_c} \frac{2q}{\sinh(2q)} \frac{1}{2\chi}\left(1 - \frac{m^2}{m_e^2}\right) m}$$

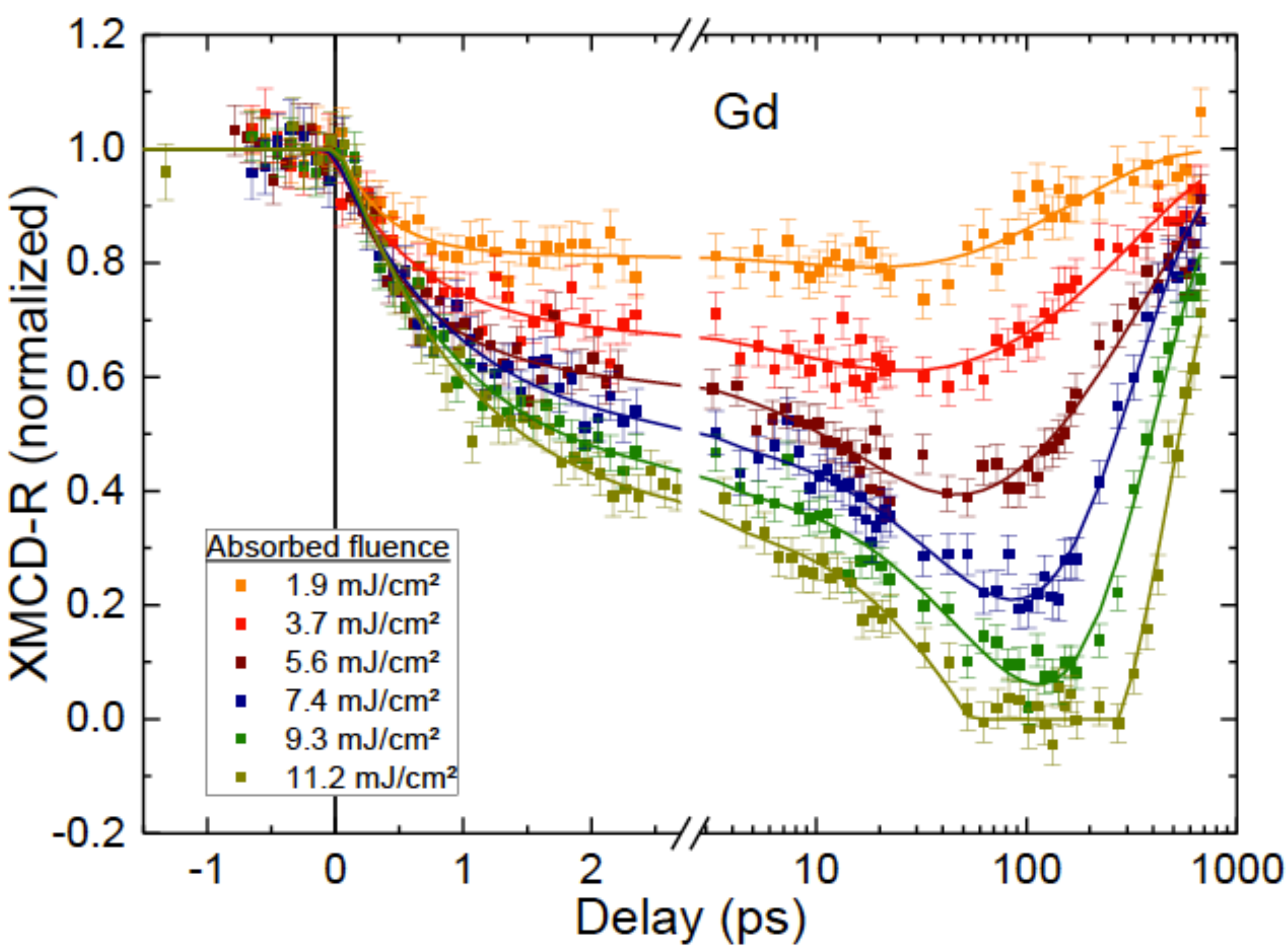
microscopic three temperature model

$$\boxed{\frac{dm}{dt} = R\frac{T_p}{T_c} m\left(1 - m \coth\left(\frac{T_c}{T_e} m\right)\right)} \qquad \boxed{R = \frac{8 a_{\mathrm{sf}} g_{\mathrm{ep}} k_B T_C^2}{\mu_{\mathrm{at}} E_D^2}}$$

Project 0: M3TM into qLLB

Example; Ultrafast magnetization in Gd

A01+ A08 (Weinelt +Atxitia)



Fluence-dependent and time-resolved XMCD scans of single-crystalline Gd measured in reflection geometry at the FEMTOSPEX facility at BESSY II (HZB).

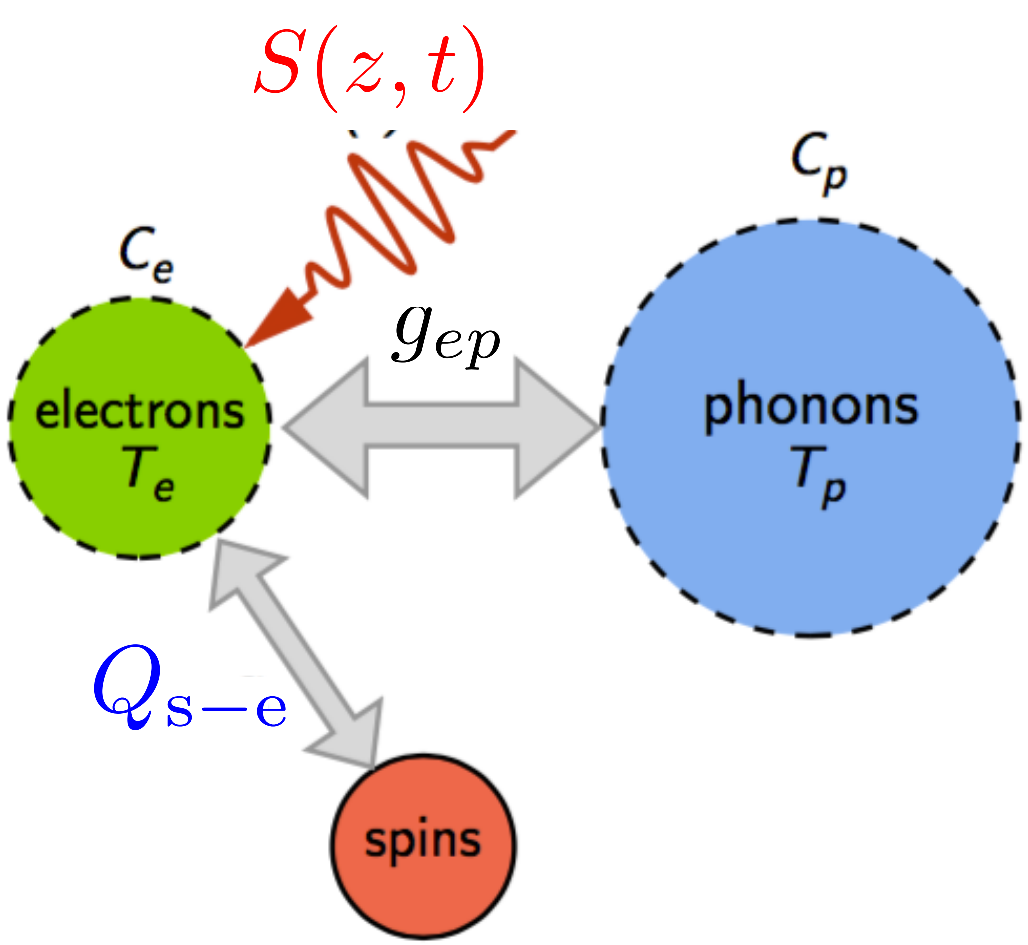
Microscopic three temperature model (M3TM)

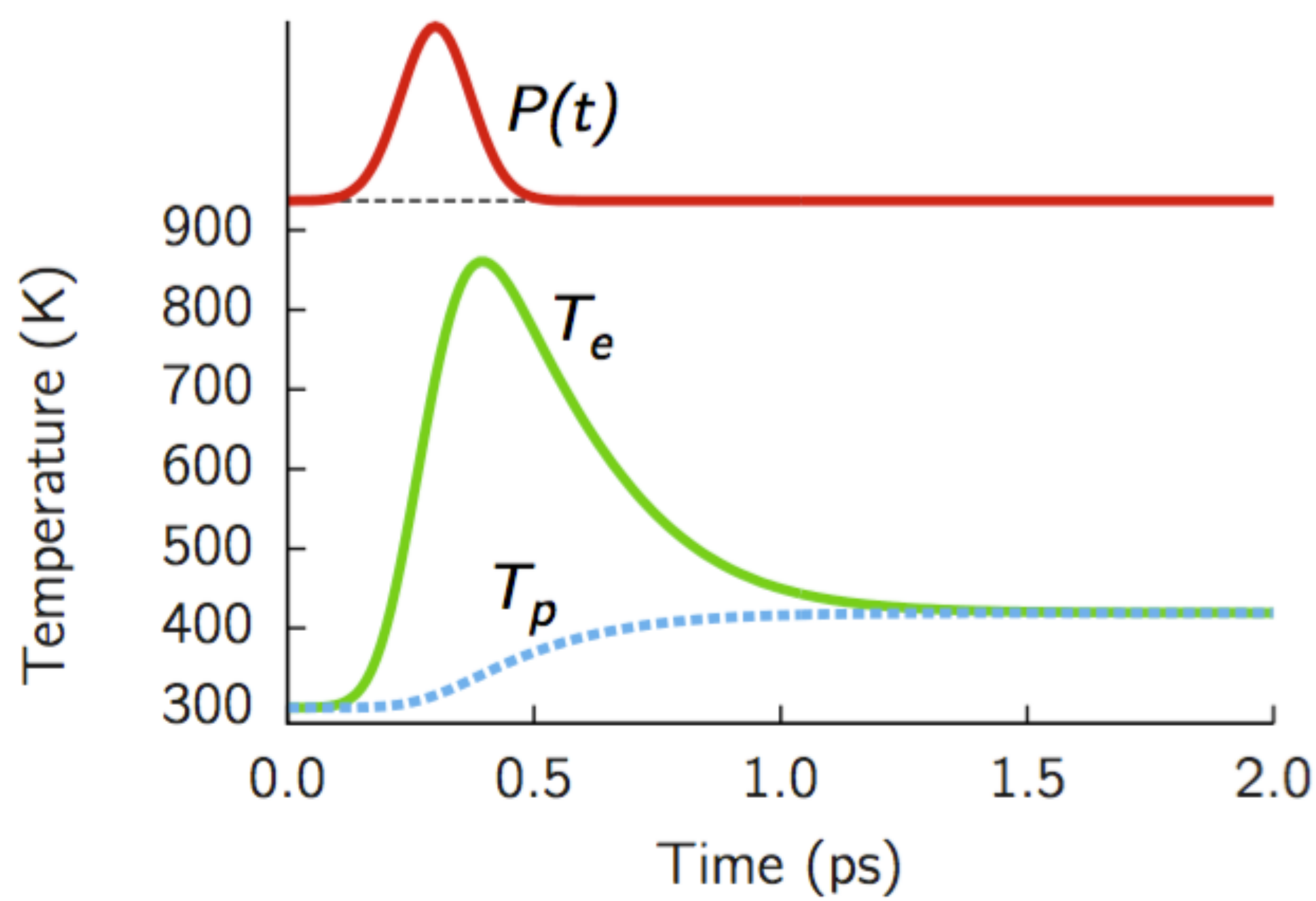
non-equilibrium between electrons and phonons: 2TM

$$C_e \frac{dT_e}{dt} = g_{ep}(T_e - T_p) + \kappa \nabla T_e + S(z,t) + Q_{\mathrm{e-s}}$$

$$C_p \frac{dT_p}{dt} = g_{ep}(T_p - T_e)$$

$$\frac{dm}{dt} = Rm\frac{T_p}{T_c}\left(1 - \frac{m}{B_{S=7/2}(\beta\Delta_{\mathrm{ex}})}\right)$$

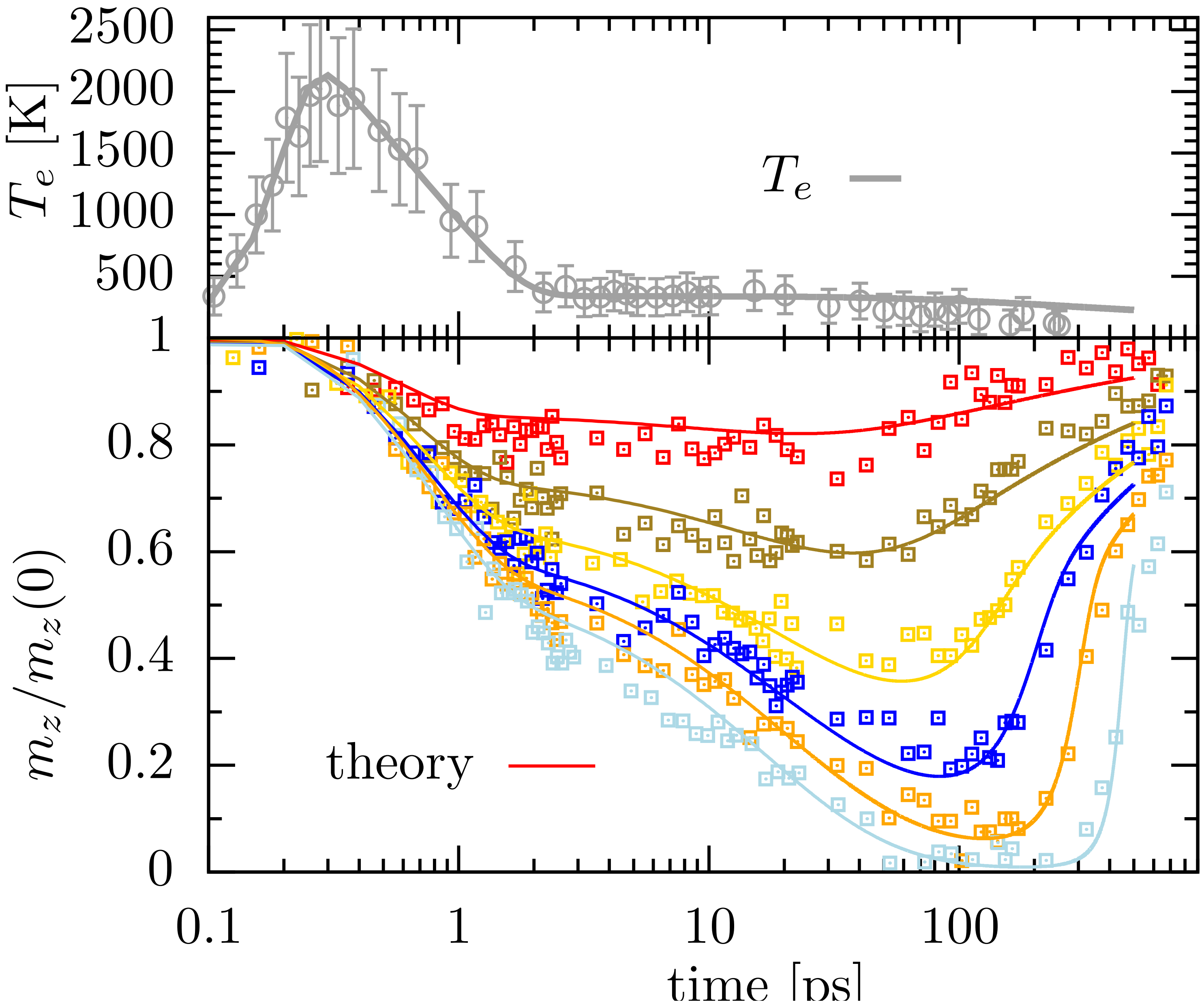


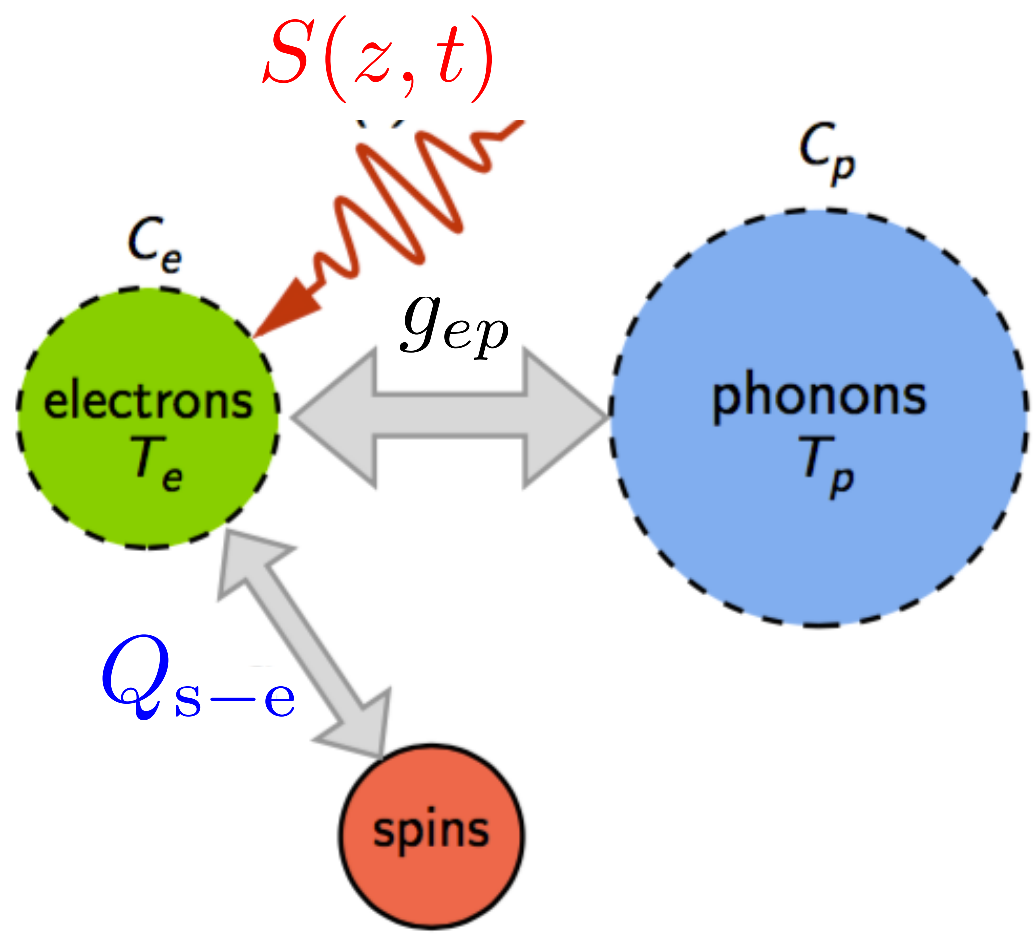


U. Bovensiepen **19**, JCMP (2007)

Magnetization dynamics in Gd

experiments vs model

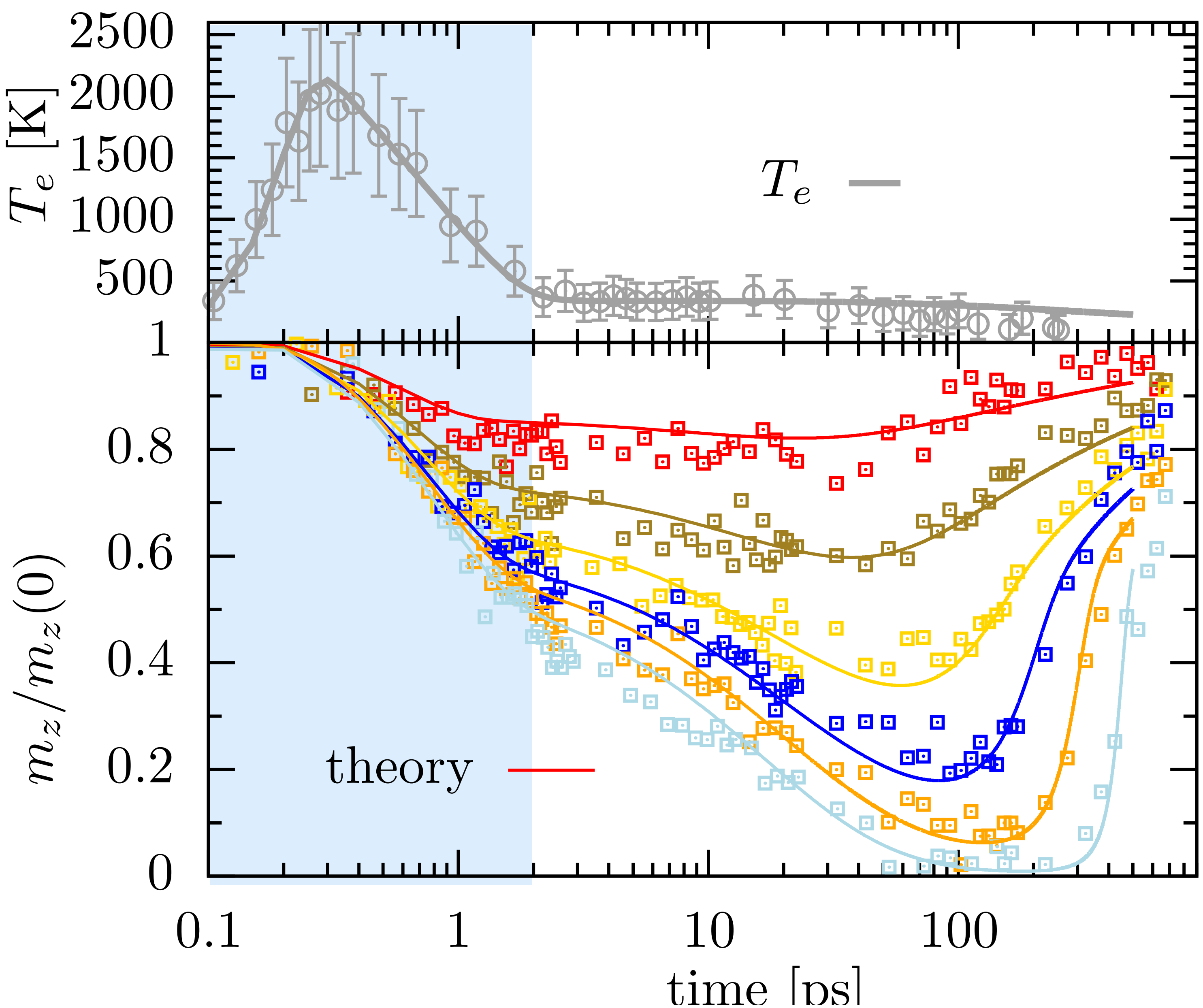


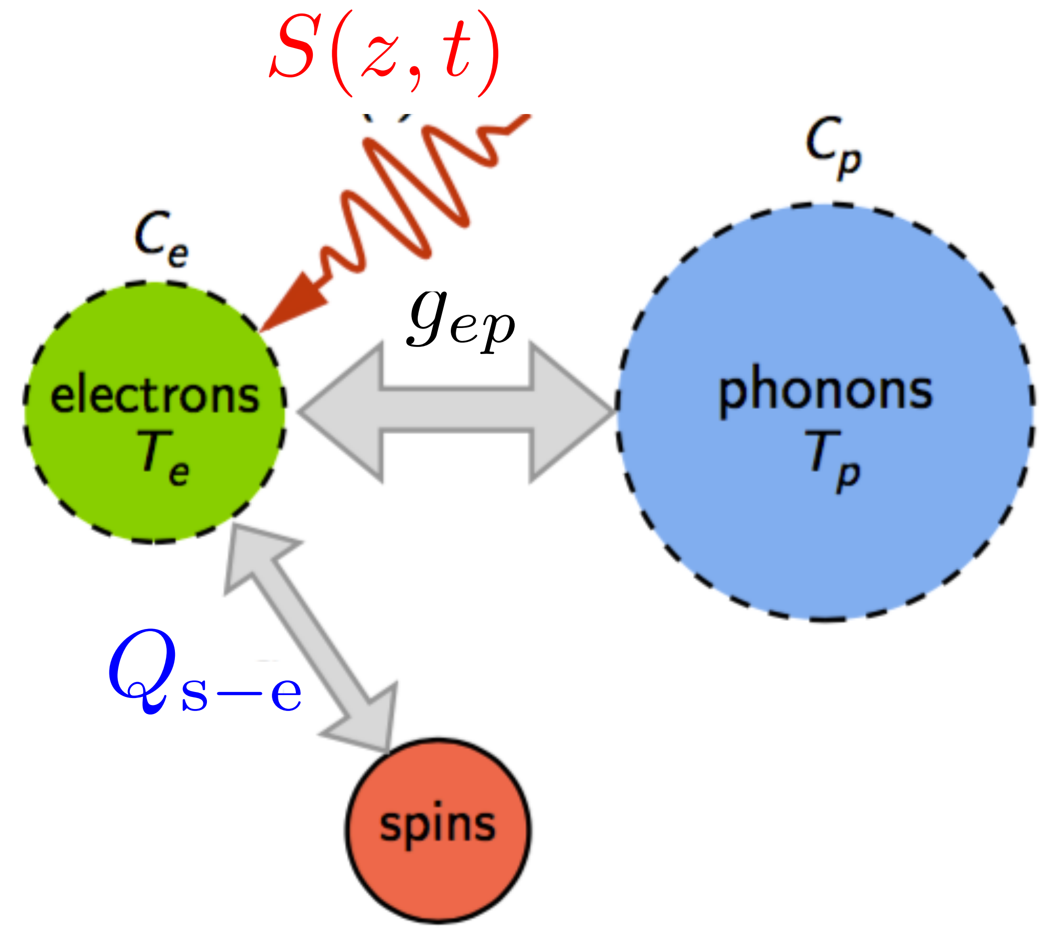


$$\frac{dm}{dt} = Rm\frac{T_p}{T_c}\left(1 - \frac{m}{B_{S=7/2}(\beta\Delta_{\mathrm{ex}})}\right)$$

Magnetization dynamics in G

experiments vs model



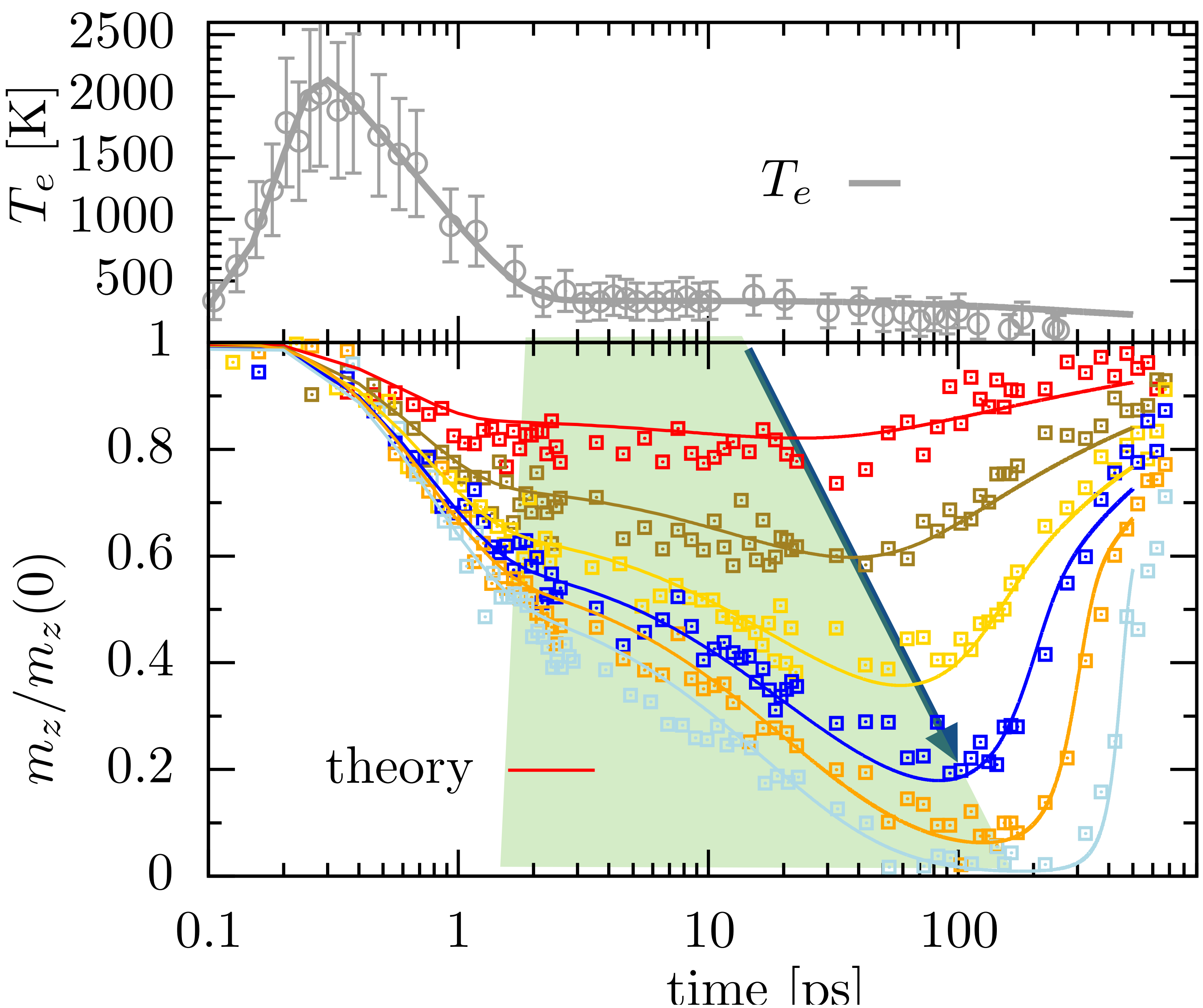


$$\frac{dm}{dt} = Rm\frac{T_p}{T_c}\left(1 - \frac{m}{B_{S=7/2}(\beta\Delta_{\mathrm{ex}})}\right)$$

Magnetization dynamics in Gd

Critical slowing down $\chi \sim \frac{\mu_{\rm at}}{T - T_c} = \left(\frac{\mu_{\rm at}}{T_c}\right)\left(\frac{1}{1 - T/T_c}\right)$

experiments vs model



in the liner regime:

$$\delta m = m - m_e \ll m_e$$

$$\boxed{\frac{dm}{dt} = \frac{\lambda}{\chi}(m_e - m)}$$

demagnetization time:

$$\boxed{\tau_{\rm de} = \chi/\lambda}$$

Origin of the distinct element-specific ultrafast spin dynamics

transition metal (Ni)

$$a_{\mathrm{sf}} = 0.185$$

$$\mu_{\mathrm{at}} = 0.63\mu_B$$

$$T_C = 631K$$

B. Koopmans et al.
Nature Materials ***9*** *259, (2010)*

$$\tau_{\mathrm{de}} \sim \frac{a_{\mathrm{sf}} g_{\mathrm{ep}} T_C}{\mu_{\mathrm{at}} E_D^2}$$

rare-earth metal (Gd)

$$a_{\mathrm{sf}} = 0.15$$

$$\mu_{\mathrm{at}} = 7.55\mu_B$$

$$T_C = 293K$$

A01+ A08 (Weinelt +Atxitia)

$$\tau_{\mathrm{de}} \sim \frac{1}{a_{\mathrm{sf}} \lambda_{ep} g(\epsilon_F)} \frac{\mu_{\mathrm{at}}}{T_C}$$

Conclusion:

- Differences come not only from magnetic, but also from the electronic and lattice parameters
- Microscopic spin-flip probability in Gd and Ni are very similar
- Electron-spin phonon mediated spin-flip mechanism describes well magnetization dynamics for all time scales in Gd

Goal: run a Fortran code in your machine

How: Windows —> install 'Visual Studio' in you machine.
Follow the instructions.
Linux —> install gcc compiler, e.g. 'gfortran'.
type 'make'
./run_code

What: up to you, see list of small projects below:

Project 0: express M3TM equation as a quantum LLB equation.

Project 1: Present the details of the derivation of the M3TM

Project 2: Present simulations of the M3TM for type-I and type-II magnets. Estimate the value of R at the transition.

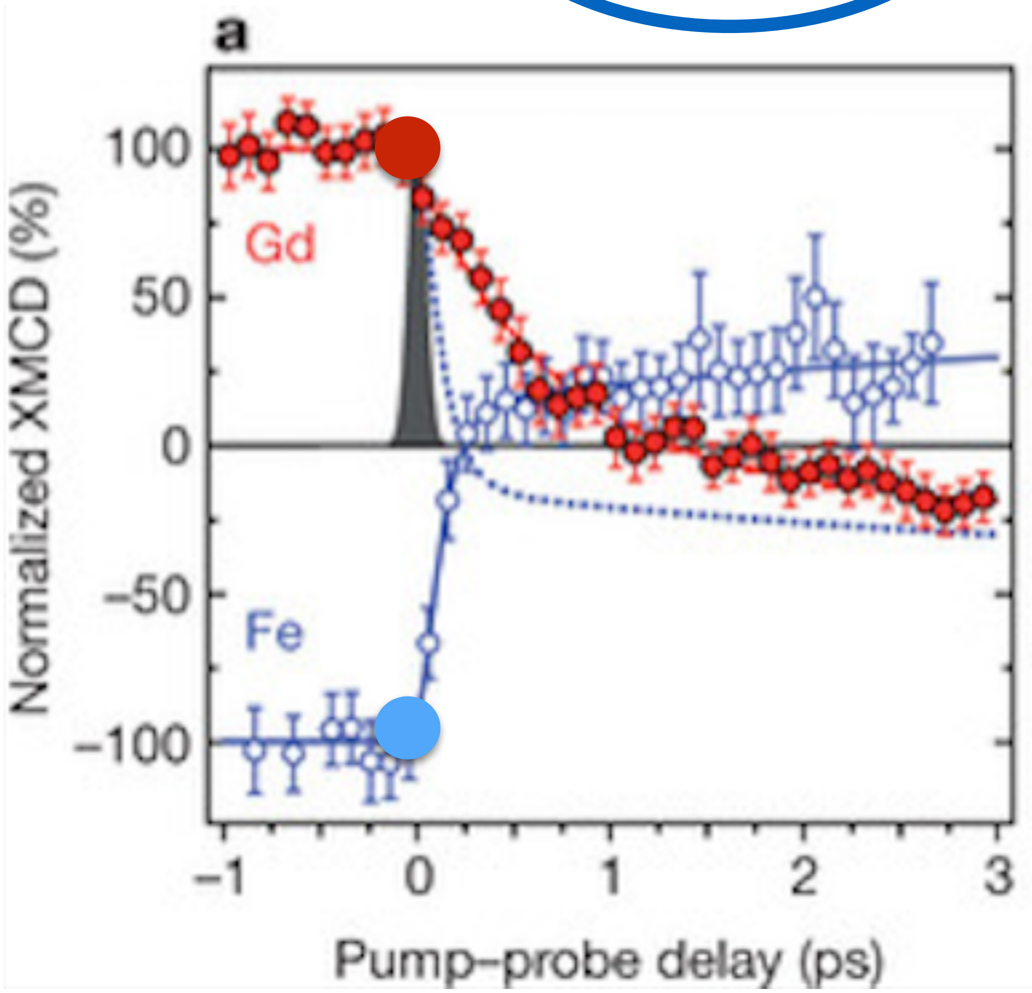
Project 3: Study the effect of thermal transport on the magnetization dynamics, with focus on type-II magnets.

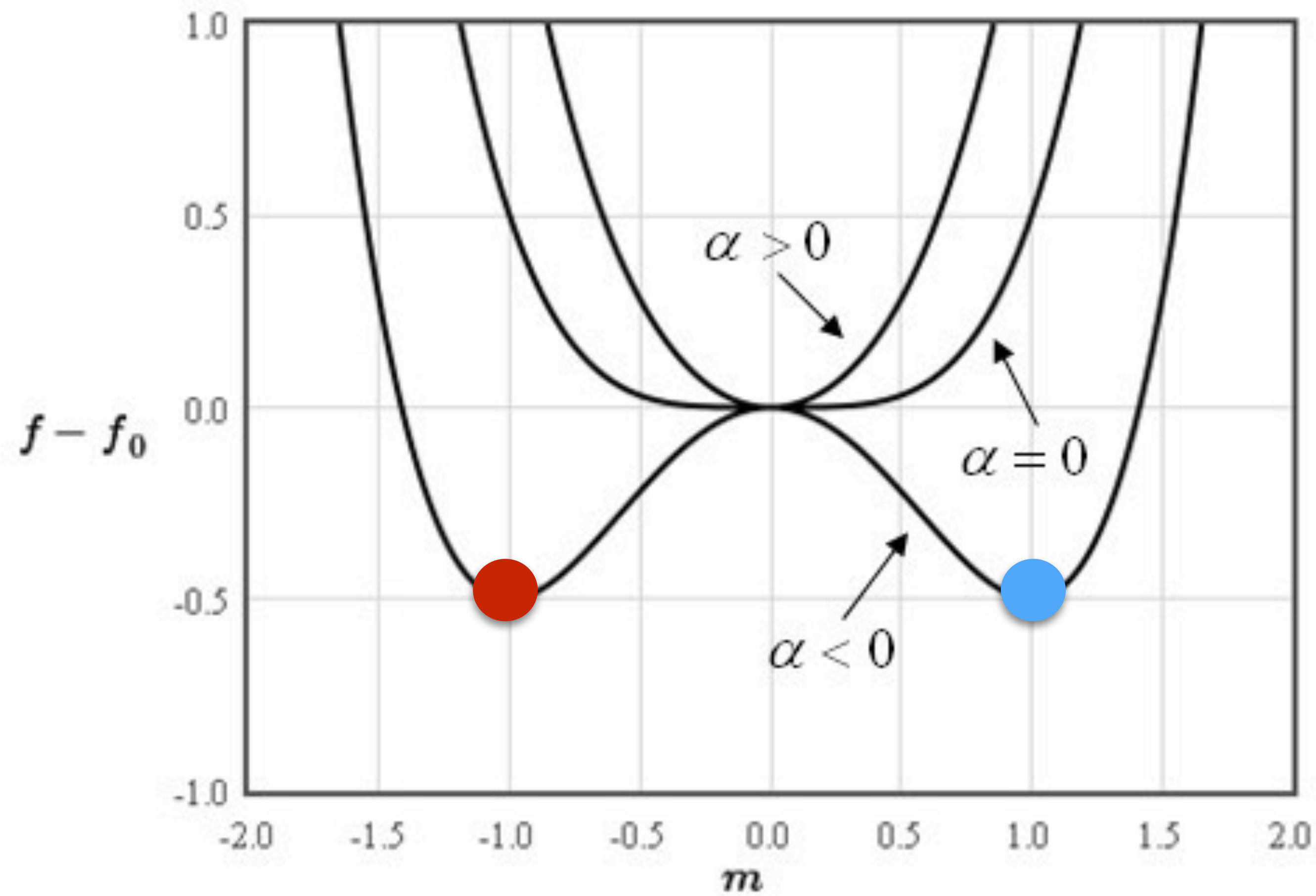
Project 4: Implement the energy flow between the spin and electron systems. Estimate the effect on the dynamics of the electron and lattice temperature.

Project 5: Implement the effect of a constant high magnetic field on the magnetization dynamics. Extend the implementation to an exponentially decaying magnetic field.

Transient ferromagnetic-like state

$$\frac{dm^{\mathrm{Fe}}}{dt} = \frac{\lambda}{\chi}(m_e^{\mathrm{Fe}} - m^{\mathrm{Fe}}) + \lambda_{ex}\left(\frac{1}{\chi^{\mathrm{Fe}}}(m_e^{\mathrm{Fe}} - m^{\mathrm{Fe}}) - \frac{\lambda}{\chi^{\mathrm{Gd}}}(m_e^{\mathrm{Gd}} - m^{\mathrm{Gd}})\right)$$





Transient ferromagnetic-like state

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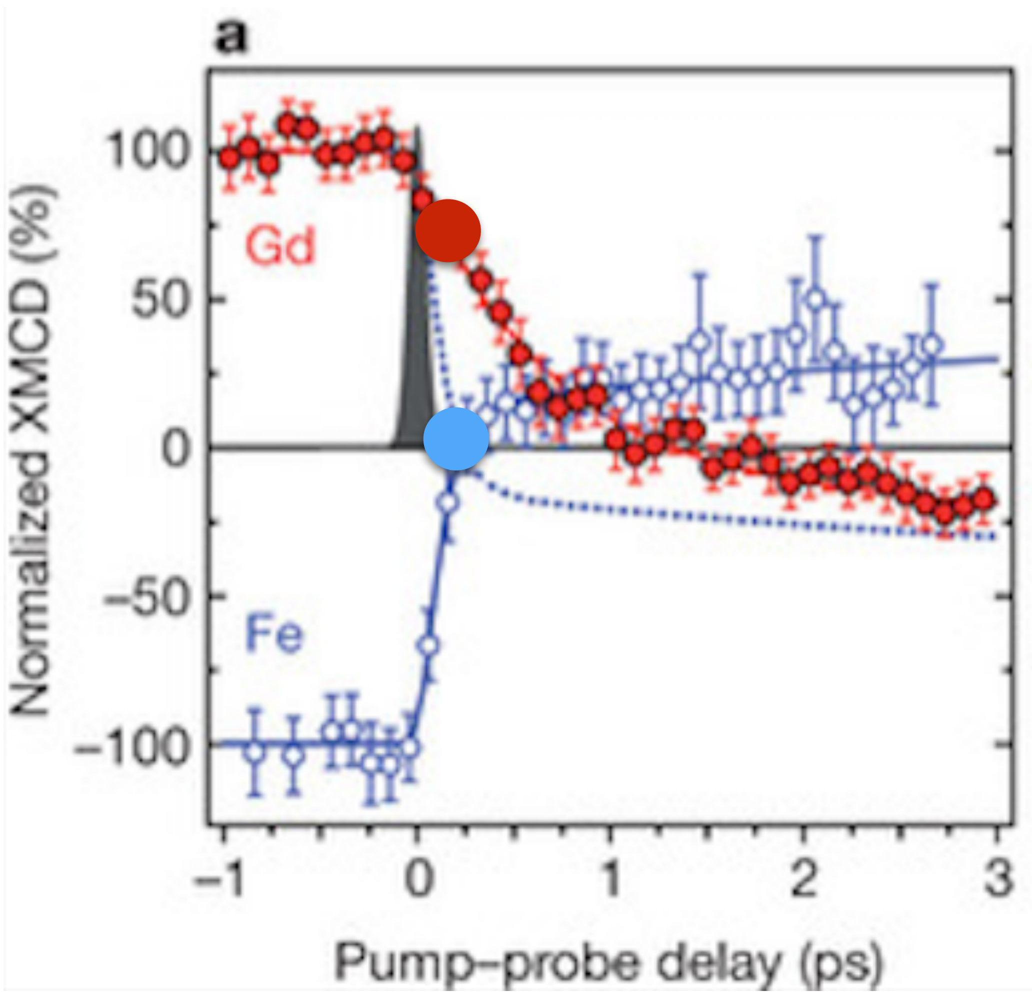

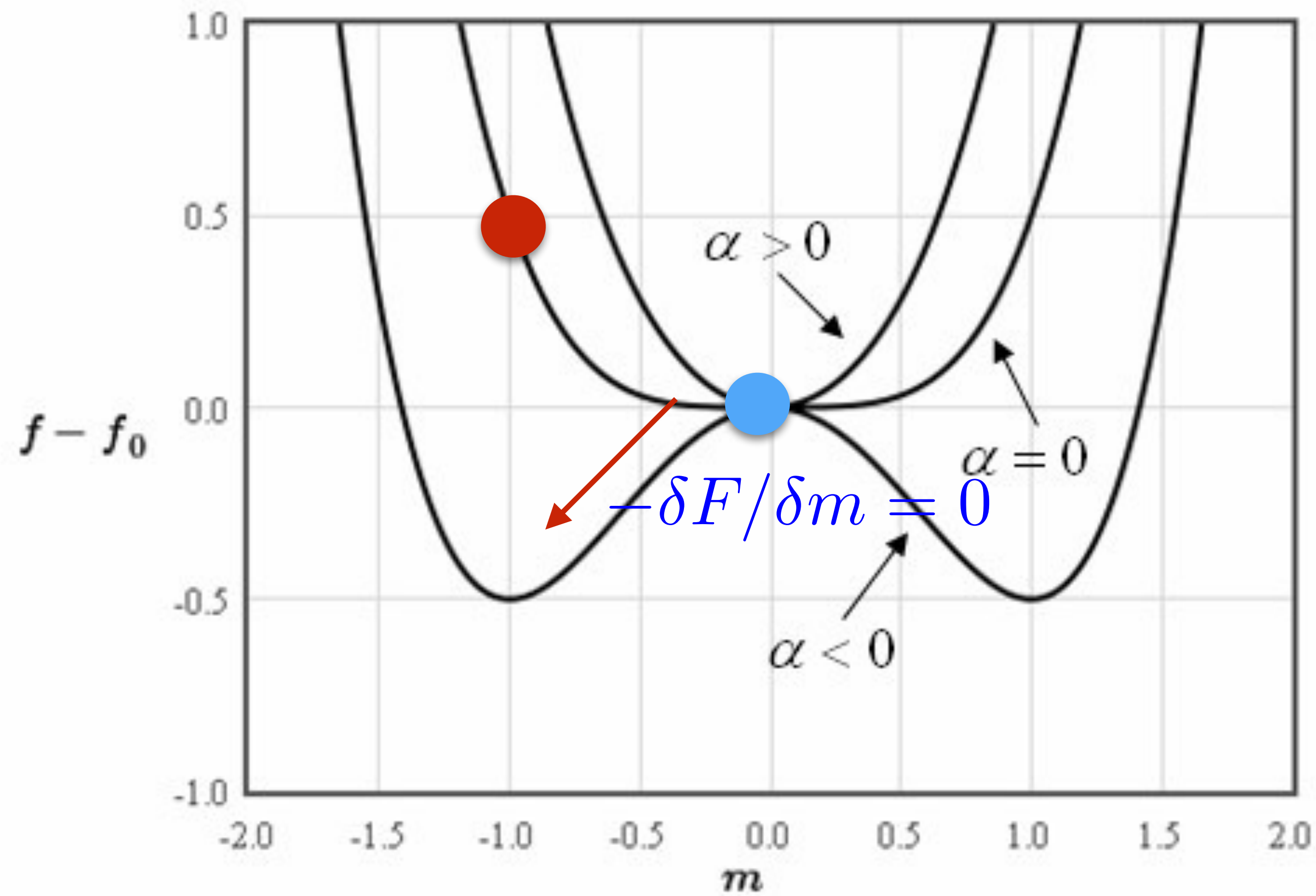

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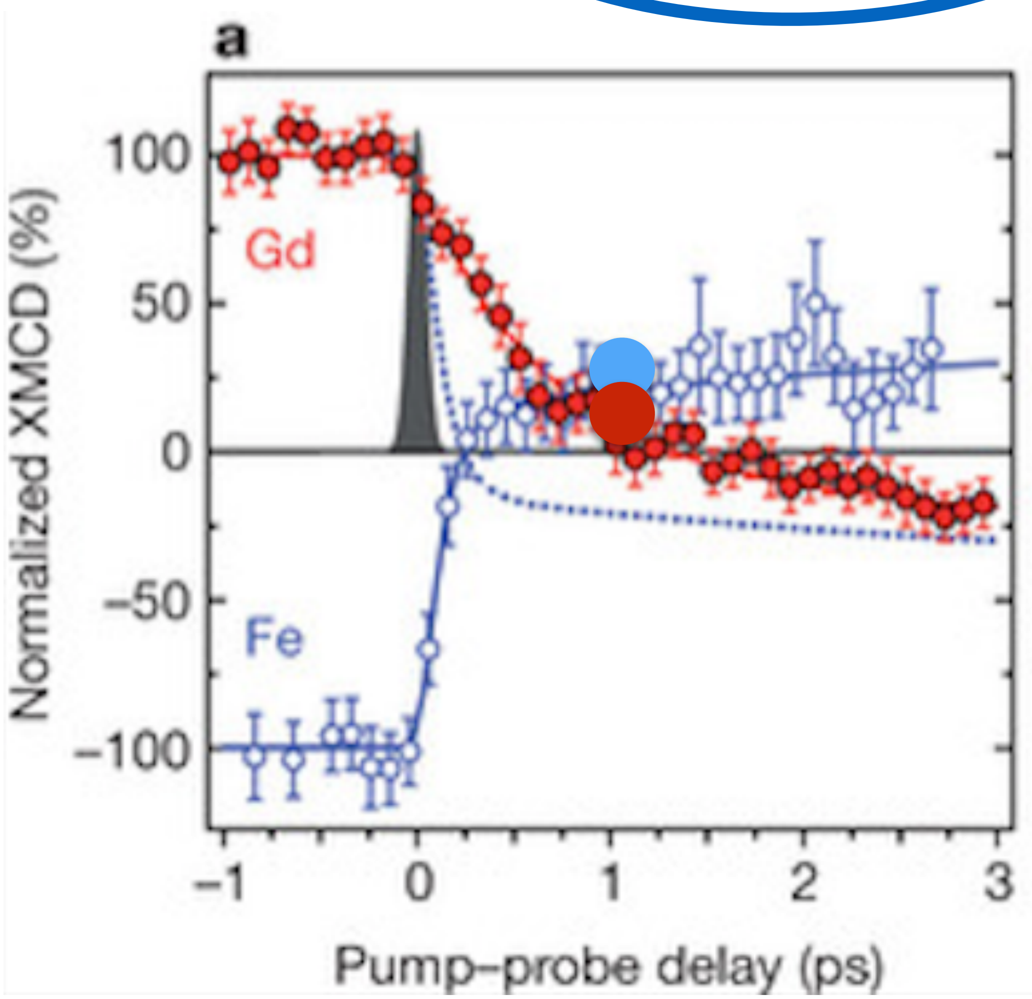

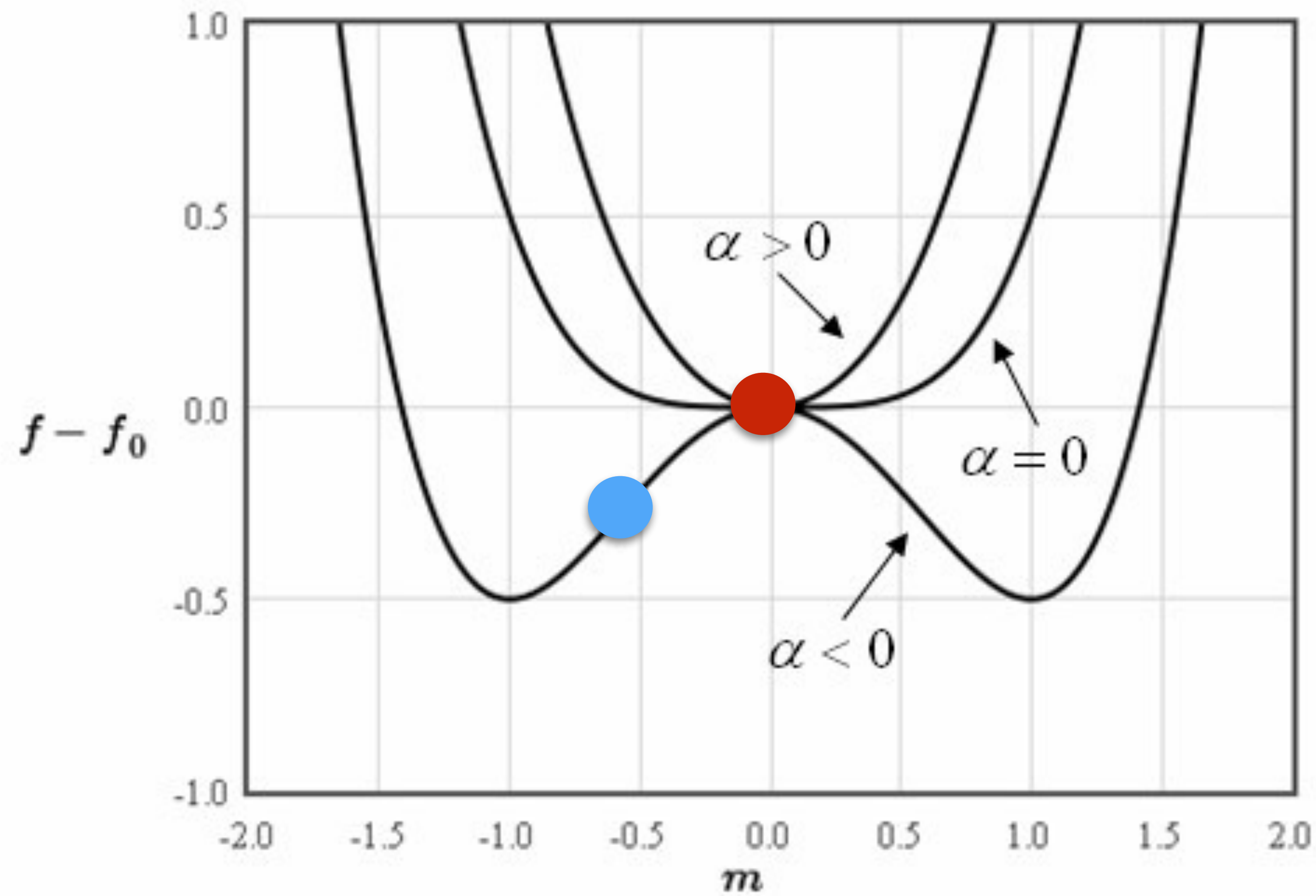

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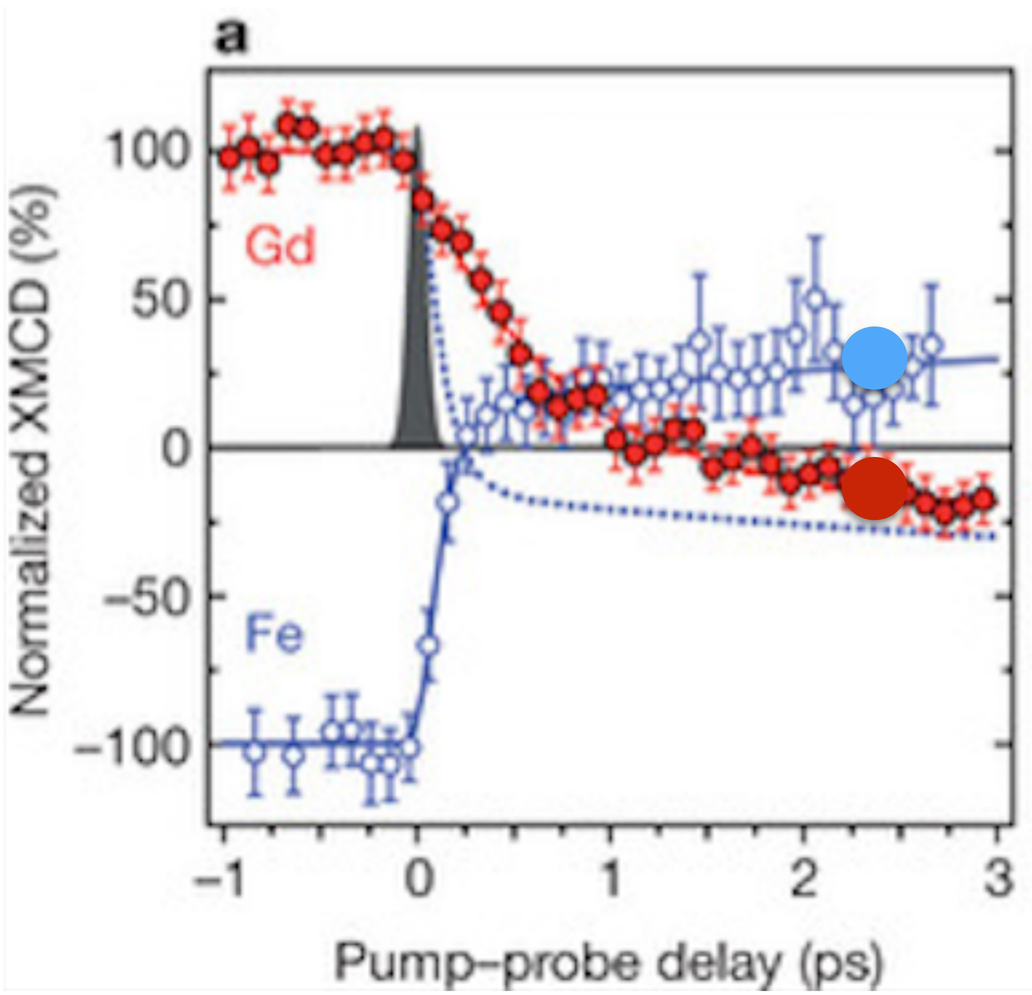
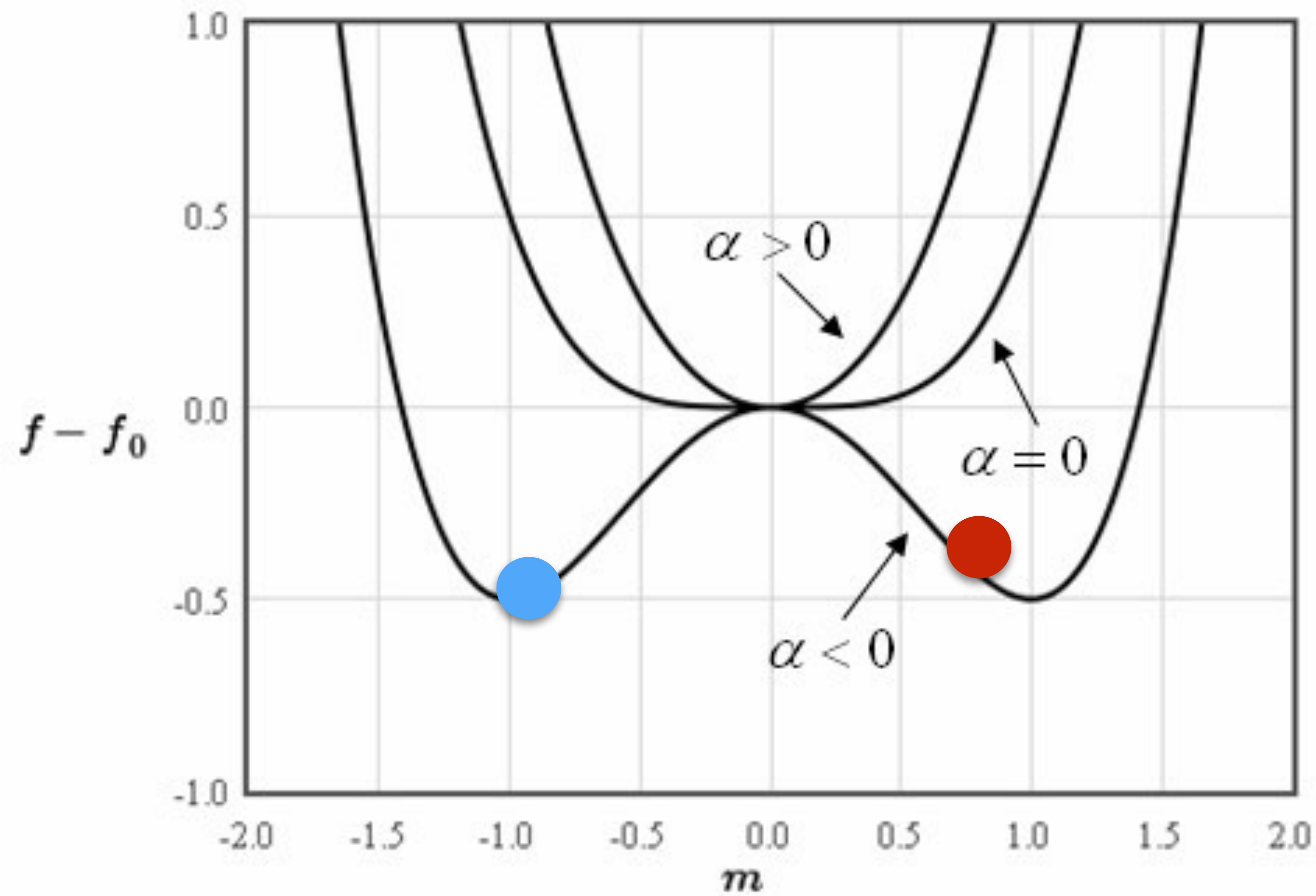
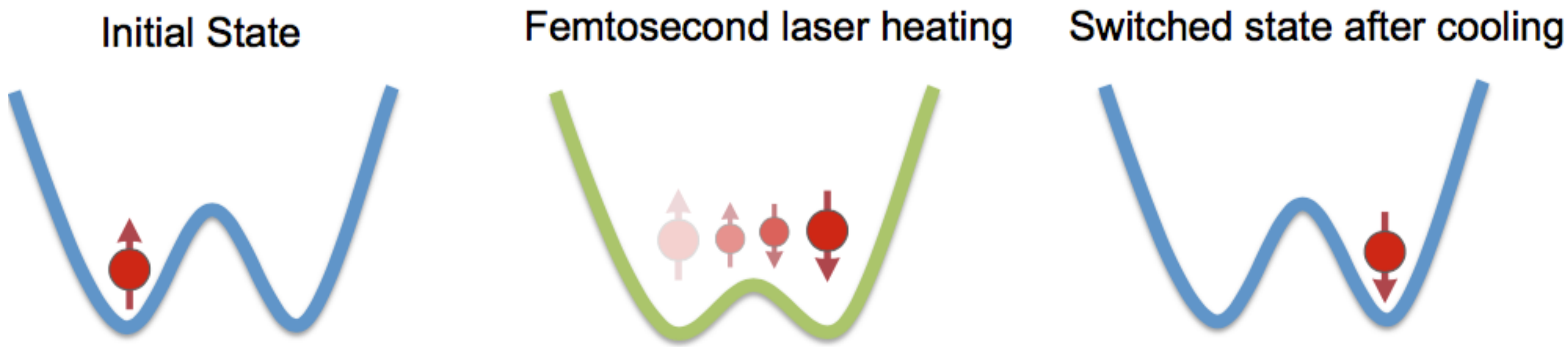




Thermal switching of magnetic domains



$$\frac{dm}{dt} = \lambda \frac{1}{\chi_{\parallel}} \left(\frac{m^2 - m_e^2}{m_e^2} \right) m - \frac{1}{\chi_{\perp}} (m_x^2 + m_y^2) + B_{\text{th}}$$