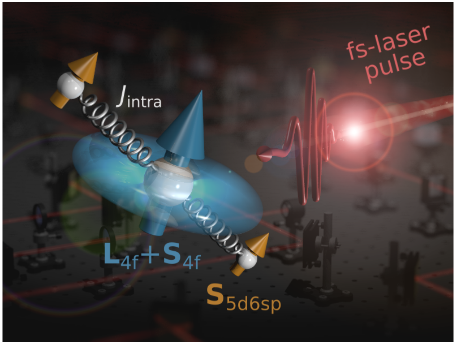
Spin dynamics: the Landau-Lifshitz equation and beyond

Uli Nowak
University of Konstanz, Germany

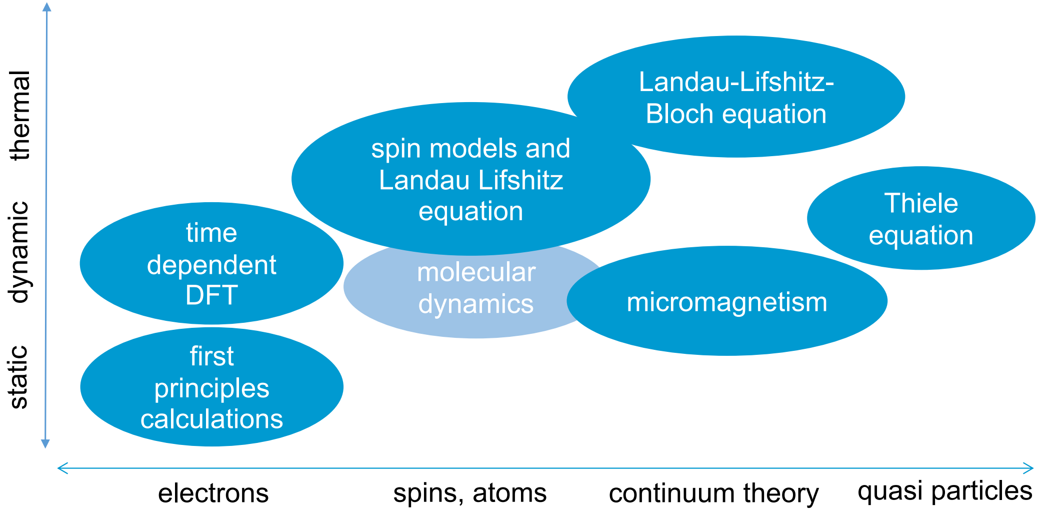
Topics:

- introduction: modelling spin dynamics
- orbital-resolved spin models and inverse Faraday effect in AFMs
- beyond LLG equation:
  field-derivative torques and nutation
- ultrafast transfer of spin angular momentum into the lattice.



Multi-scale modelling in magnetism

- **goals:** understanding spin structures and dynamics, involving
  - time scales from femtoseconds to years
  - length scales from electronic to sample size
  - temperatures from zero up to the Curie temperature
  - opto-magnetic effects, charge currents, rapid heating

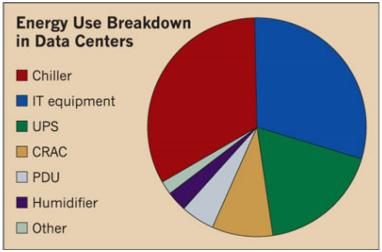


Energy consumption of data centers

- In 2013, data centers in the U.S. consumed an estimated **91 billion kWh of electricity**. *http://ecmweb.com/energy-efficiency/data-center-efficiency-trends*

- per the U.S. Energy Information Administration, that is about **7% of total commercial electric energy consumption**

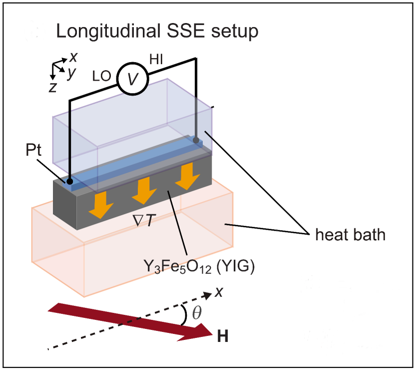
- this number is only going to go up: by 2020, it is estimated consumption will increase to 140 billion kWh, costing about 13 billion $ in power bills.



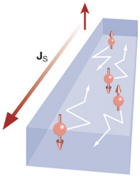
New transport properties: spin caloritronics etc.

Combined transport of spin (with/without charge) and heat

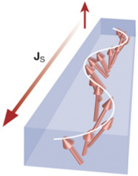
- **Spin Seebeck effect:** generation a spin voltage through a temperature gradient in a conducting ferromagnet *(K. Uchida et al., Nature* **455***, 778 (2008))* or even an insulator *(K. Uchida et al., Nature* **9***, 894 (2010)); Xiao et al. Phys. Rev. B* **81***, 214418 (2010))*



- **Spin current:** two possible types of carriers *(Y. Kajiwara et al., Nature* **464***, 262 (2010))*



- spin current via polarized electrons



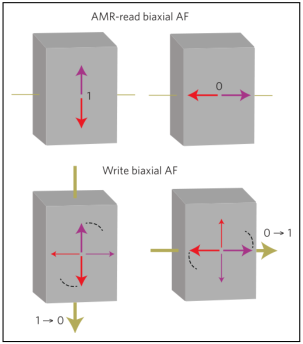
- chargeless spin current via magnons

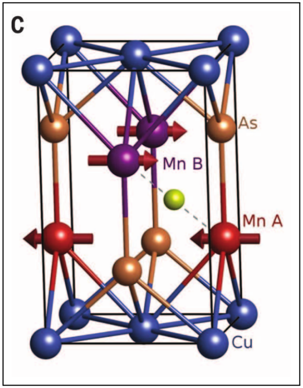
Spintronics with antiferromagnets

- **read antiferromagnetic order parameter** in biaxial AFMs via anisotropic magneto-resistance *(Jungwirth et al. Nat. Nanotech.* **11**, *91 (2017))*

- **write with current-induced staggered fields** *(Wadley et al., Science* **351**, *6273 (2016))*

- **advantages of AFM**:
  - no coupling via stray field
  - not susceptible to external fields
  - faster dynamics
  - new class of materials (low damping, insulators)





Ultrafast: all-optical magnetization switching

- uses **circularly polarised light** pulses of $\approx$ 100 fs duration
- writing bits **without applying an external magnetic field**
- magnetisation direction **defined by helicity of light**
- time scale some hundred femtoseconds
- many open questions regarding fundamental **interactions between laser, electron spins, lattice**

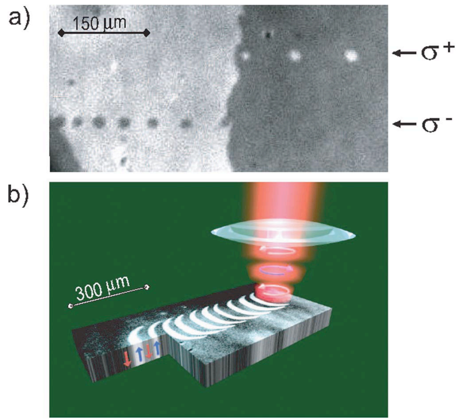


FIG. 4 (color). All-optical magnetic recording by femtosecond laser pulses. (a) The effect of single 40-fs circular polarized laser pulses on the magnetic domains in $Gd_{22}Fe_{74.6}Co_{3.4}$. The domain pattern was obtained by sweeping at high-speed (~50 mm/s) circularly polarized beams across the surface so that every single laser pulse landed at a different spot. The laser fluence was about 2.9 mJ/cm^2. The small size variation of the written domains is caused by the pulse-to-pulse fluctuation of the laser intensity.

Stanciu et al. PRL **99**, *047601 (2007)*

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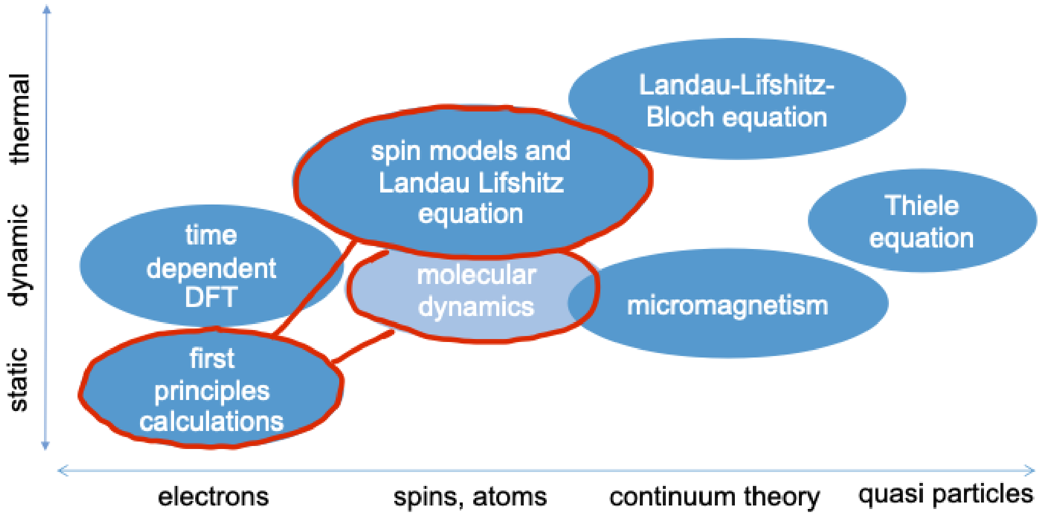


Micromagnetic continuum theory

- on a length scale $\gg$ lattice constant a the **magnetisation** is assumed as field $\boldsymbol{m}(\boldsymbol{r})$ with constant length and **energy**:

$$E = \frac{J}{2a}\int_V (\nabla \boldsymbol{m})^2 \, \mathrm{d}V - \frac{d_z}{a^3}\int_V S_z^2 \, \mathrm{d}V - M_\mathrm{s}\int_V \boldsymbol{m}\cdot\boldsymbol{B} \, \mathrm{d}V - \frac{\mu_0}{2}\int_V \boldsymbol{m}\cdot\boldsymbol{H}_\mathrm{s} \, \mathrm{d}V$$

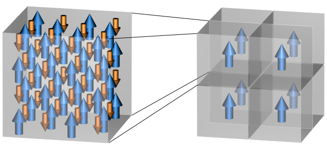
exchange anisotropy external field magneto-static

- equation of motion:

Landau-Lifshitz-Gilbert equation

$$\dot{\boldsymbol{m}} = -\frac{\gamma}{(1+\alpha^2)M_s}\boldsymbol{m}_i \times \boldsymbol{H}_\mathrm{eff} - \frac{\alpha\gamma}{(1+\alpha^2)M_s}\boldsymbol{m} \times (\boldsymbol{m}_i \times \boldsymbol{H}_\mathrm{eff})$$

- with effective field
- describes domain structures and magnetisation dynamics
- **at finite temperatures**:
  - Langevin dynamics: add stochastic field *(Brown, Phys. Rev.* **130**, *1677 (1963))*
  - ☹ at elavated temperatures approach will fail since spin wave spectrum is limited by cell size (*Grinstein and Koch, PRL* **90**, *207201 (2003)*)



Atomistic spin model

- Hamiltonian for spins $\underline{S}_i = \underline{\mu}_i/\mu_s$ on a given lattice:

spin model including relativistic interactions

$$\mathcal{H} = -\frac{1}{2}\sum_{i,j} \boldsymbol{S}_i \mathrm{J}_{ij} \boldsymbol{S}_j - \sum_i d_i^z (S_i^z)^2 - \boldsymbol{B}\cdot\sum_i \mu_i \boldsymbol{S}_i - \sum_{i,j} \frac{\mu_0 \mu_i \mu_j}{8\pi} \frac{3(\boldsymbol{S}_i\cdot\boldsymbol{e}_{ij})(\boldsymbol{e}_{ij}\cdot\boldsymbol{S}_j) - \boldsymbol{S}_i\cdot\boldsymbol{S}_j}{r_{ij}^3}$$

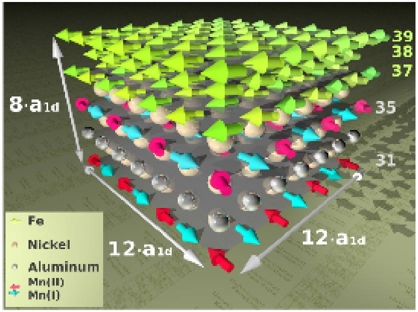
exchange anisotropy external field dipole-dipole

- **tensorial exchange interactions** J_{ij} can be decomposed in:

$$\mathcal{H}_{\mathrm{ex}} = -\tfrac{1}{2}\sum_{i,j} J_{ij}^{\mathrm{iso}} \boldsymbol{S}_i\cdot\boldsymbol{S}_j \quad -\tfrac{1}{2}\sum_{i,j} \boldsymbol{S}_i \mathrm{J}_{\mathrm{ij}}^{\mathrm{S}} \boldsymbol{S}_j \quad -\tfrac{1}{2}\sum_{i,j} \boldsymbol{D}_{ij}\cdot(\boldsymbol{S}_i\times\boldsymbol{S}_j)$$

isotropic exchange two-site anisotropy Dzyaloshynskii-Moriya

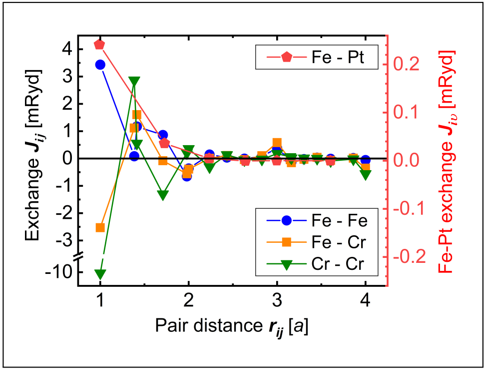
- different types of **anisotropies** ...
- **dipole-dipole interaction** leads to:
  - shape anisotropy
  - domain structures
  - large numerical effort



Yanes et al., PRB **96**, *064435 (2017)*

Spin model parameters from first principles

- exchange integrals, isotropic but **orbital resolved** *(Liechtenstein et al., JMMM* **67***, 65 (1987))*
- fully **relativistic** screened Korringa-Kohn-Rostoker (SKKR) method plus **spin cluster expansion** for layered systems and clusters *(Szunyogh et al., PRB* **83***, 024401(2011))*
- calculations of **opto-magnetic effects** such as IFE *(Berritta et al., PRL* **117***, 137203 (2016); John et al., Scientific Reports* **7***, 4114 (2017))*



Schmidt et al., PRB **102***, 214436 (2020)*

Gd	orbital-resolved dynamics	*Frietsch et al., Nature Com.* **6** *8262 (2015)*
Tb	orbital-resolved dynamics	*Frietsch et al., Science Advances,* **6***, eabb1601 (2020)*
CrPt	switching with IFE in an AFM	*Dannegger et al., submitted (2021)*

Equation of motion

stochastic Landau-Lifshitz-Gilbert equation

$$\dot{\boldsymbol{S}}_i = -\frac{\gamma}{(1+\alpha^2)\mu_s}\boldsymbol{S}_i \times \boldsymbol{H}_i(t) \qquad \text{precession}$$

$$-\frac{\alpha\gamma}{(1+\alpha^2)\mu_s}\boldsymbol{S}_i \times \left(\boldsymbol{S}_i \times \boldsymbol{H}_i(t)\right) \qquad \text{dissipation}$$

with $\boldsymbol{H}_i(t) = -\frac{\partial \mathcal{H}}{\partial \boldsymbol{S}_i} + \boldsymbol{\zeta}_i(t)$ fluctuations

and $\langle \zeta_i(t) \rangle = 0, \quad \langle \zeta_{i\eta}(0)\zeta_{j\vartheta}(t)\rangle = \delta_{ij}\delta_{\eta\vartheta}\delta(t) 2\alpha k_{\mathrm{B}} T \mu_s/\gamma.$

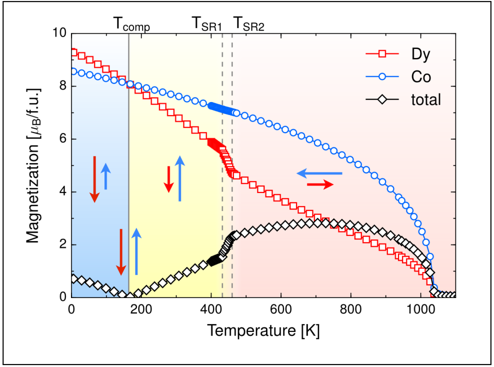
spin dynamics

α: quantifies coupling to the electronic/phononic heat bath

- **numerical integration** of the stochastic LLG equation *(Lyberatos et al., J. Phys. C **5**, 8911 (1993))*
- simulation of 10^8 spins possible
- statistical average in the **canonical ensemble**

☺ realistic dispersion relations; non-linear prozesses; critical behavior methods

☹ classical approximation; large numerical effort



*Donges et al., PRB **96**, 024412 (2017)*

Spin dynamics: the Landau-Lifshitz equation and beyond

Tobias Dannegger, Andreas Donges, Severin Selzer, Uli Nowak
University of Konstanz, Germany

In collaboration with:
M. Berritta, P. Oppeneer, University of Uppsala
C. Karva, Charles University, Prague
B. Frietsch, U. Ritzmann, J. Bowlan, R. Carley, M. Teichmann, M. Weinelt, FU Berlin

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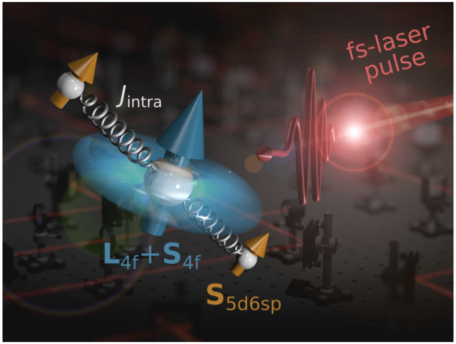
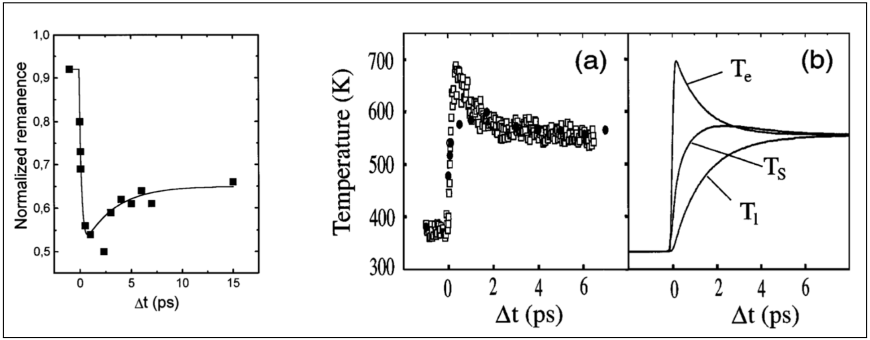
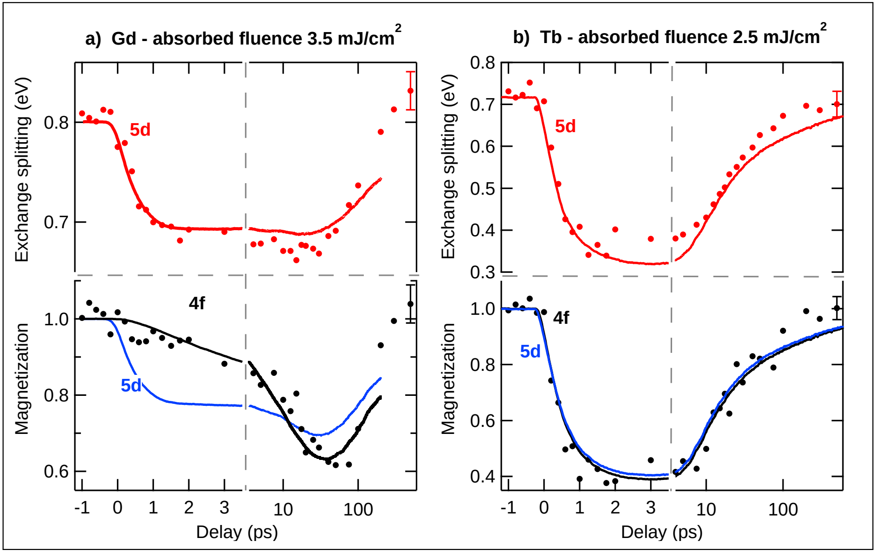


Laser-induced ultrafast spin dynamics



- following a fs laser pulse **magnetisation can break down on a time scale of some hundred femtoseconds and recover on a ps time scale**
  (*Beaurepaire et al., PRL* **76**, *4250 (1996)*)
- the phenomenological **three-temperature model** gives an idea of the energy flow
- flow of **angular momentum** remains an open question
- coupling between **microscopic degrees of freedom** (spin, electrons, lattice) hardly understood

Demagnetization dynamics of Gd and Tb



(Frietsch et al., Science Adv. **6**, *eabb1601 (2020))*

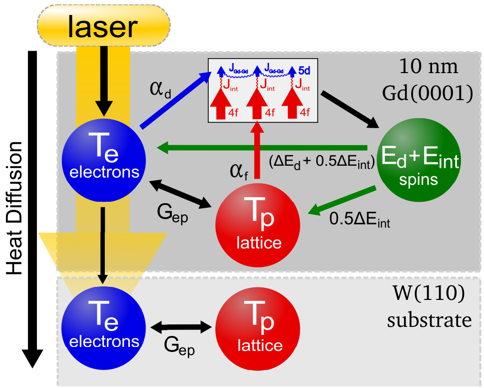
- photoemission data: concurrent measurement of 5d-6s-exchange splitting and 4f magnetic linear dichroism *(B. Frietsch et al., Nature Com.* **6**, *8262 (2015))*
- Gd shows **distinct demagnetization dynamics** of d and f electrons
- Tb does not
- excellent agreement between theory (lines) and measurement (points)

Modelling the magnetic response to the laser pulse: Gd and Tb

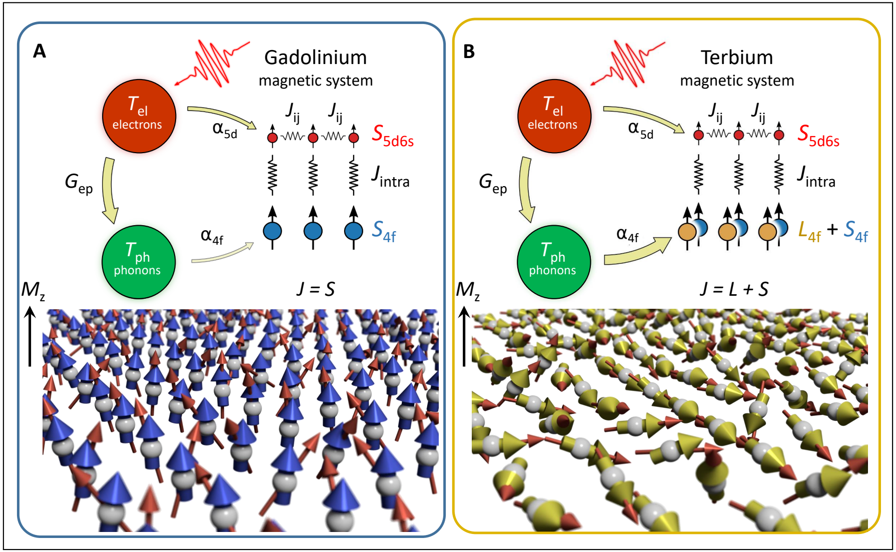
- **two temperature model** for electron and phonon temperature *(Kaganov et al. JETP* **4**, *173 (1957))*

$$C_e \frac{dT_e}{dt} = -G_{el}(T_e - T_p) + P(t)$$
$$C_p \frac{dT_p}{dt} = G_{el}(T_e - T_p)$$

- add heat diffusion and substrate
- **orbital-resolved spin model** for spin dynamics simulations with parameters form ab-initio calculations *(Carva, Oppeneer)*
- cooling effect of spin system taken into account



Demagnetization dynamics of Gd and Tb

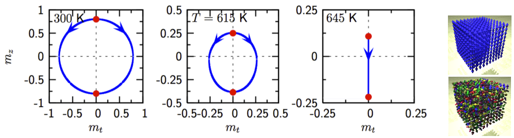


(Frietsch et al., Science Adv. **6***, eabb1601 (2020))*

- Gd shows distinct demagnetization dynamics of d and f electrons
- Tb does not because of the **larger coupling to the phonons**

Switching with inverse Faraday effect

- **helicity-dependent all-optical switching** demonstrated in many materials
- strong influence of heating, **switching is linear**:

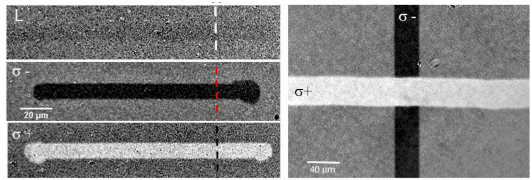


(*Kazantseva et al., Europhys. Lett.* **86**, *27006 (2009)*)

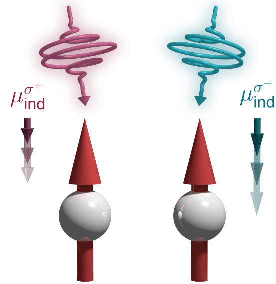
- due to **inverse Faraday effect:** circularly polarized light induces magnetization in a sample (*Battiato et al. PRB* **89**, *0144413 (2014)*)

- **what about antiferromagnets?**

all-optical writing of an FePt nanoparticle recording medium

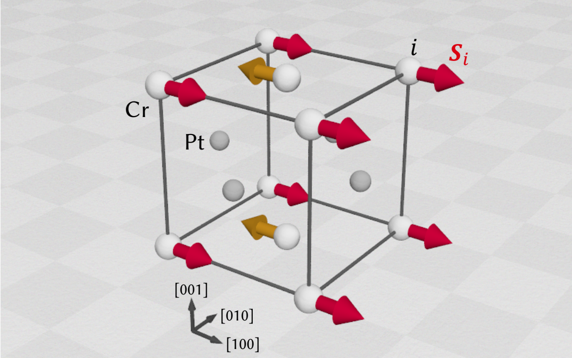


John et al., Scientific Reports **7**, *4114 (2017)*

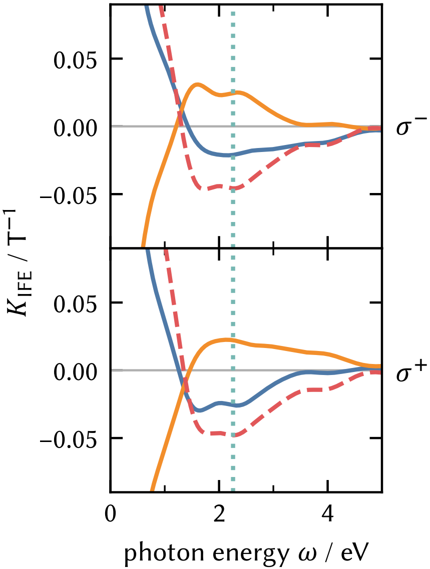


Inverse Faraday effect in CrPt

- CrPt: **antiferromagnet** with two sublattices
- **IFE quantified** by first priciples calculations *(Berritta et al., Phys. Rev. Lett.* ***117****, 137203 (2016))*



- lattice and spin moments



IFE constants Cr moment direction parallel and antiparallel to k-vector of the light. Red line indicates difference.

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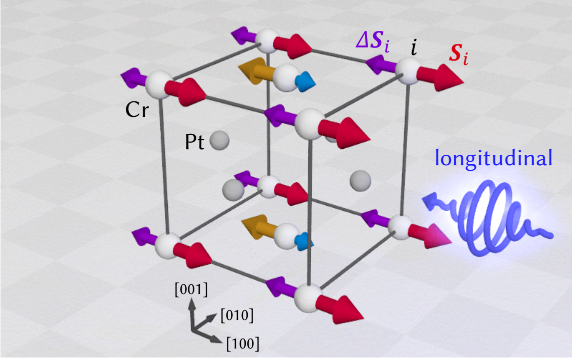
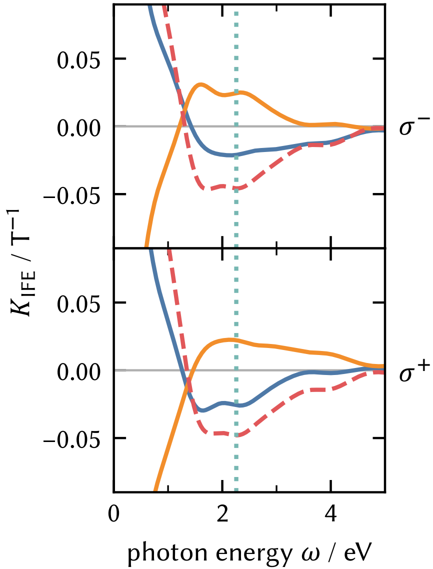


- IFE leads to **staggered induced moments**



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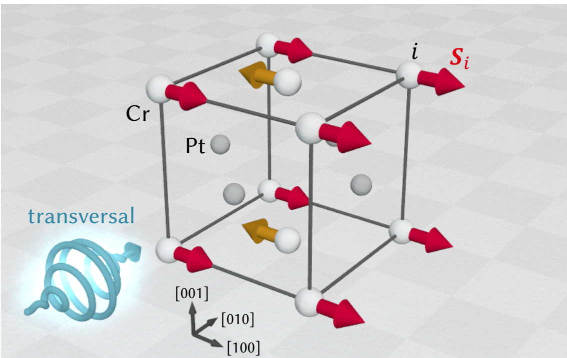
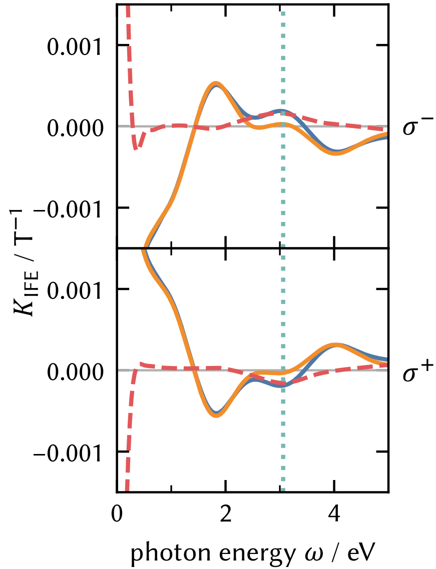




IFE constants Cr moment direction parallel and antiparallel to k-vector of the light. Red line indicates difference.

- perpendicular IFE very small

Atomistic spin dynamics

- **induced magnetic moments** from DFT calculations via:

$$\mu_{\text{ind}} = \frac{K^{\sigma}_{\text{IFE}}(\omega)V}{c} I(t)$$

- add to the existing moments:

$$\boldsymbol{\mu}_i \to \boldsymbol{\mu}_i + \boldsymbol{\mu}_{\text{ind}}, \quad \boldsymbol{S}_i \to \boldsymbol{S}_i + \Delta\boldsymbol{S}_i$$

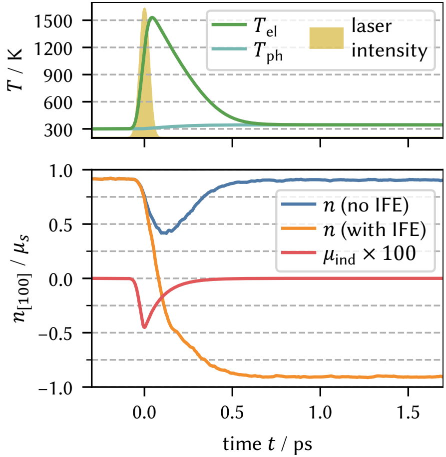
spin model

$$\mathcal{H} = -\sum_{\langle ij\rangle} J_{ij}(\boldsymbol{S}_i + \Delta\boldsymbol{S}_i)\cdot(\boldsymbol{S}_j + \Delta\boldsymbol{S}_j) - d_z \sum_i S_{iz}^2$$

- induced moments **couple via exchange interaction** with neighbouring spins
- dynamics via stochastic LLG equation

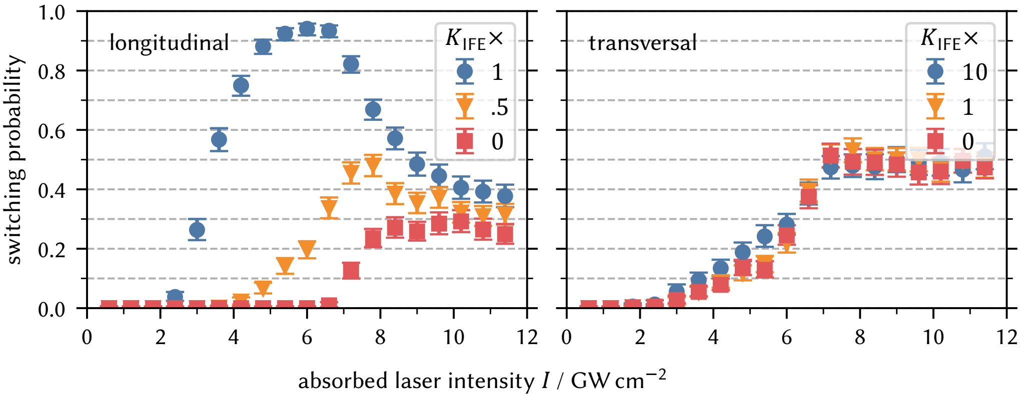
two-temperature model:

$$C_{\text{el}}\dot{T}_{\text{el}} = G(T_{\text{ph}} - T_{\text{el}}) + P(t),$$
$$C_{\text{ph}}\dot{T}_{\text{ph}} = G(T_{\text{el}} - T_{\text{ph}})$$



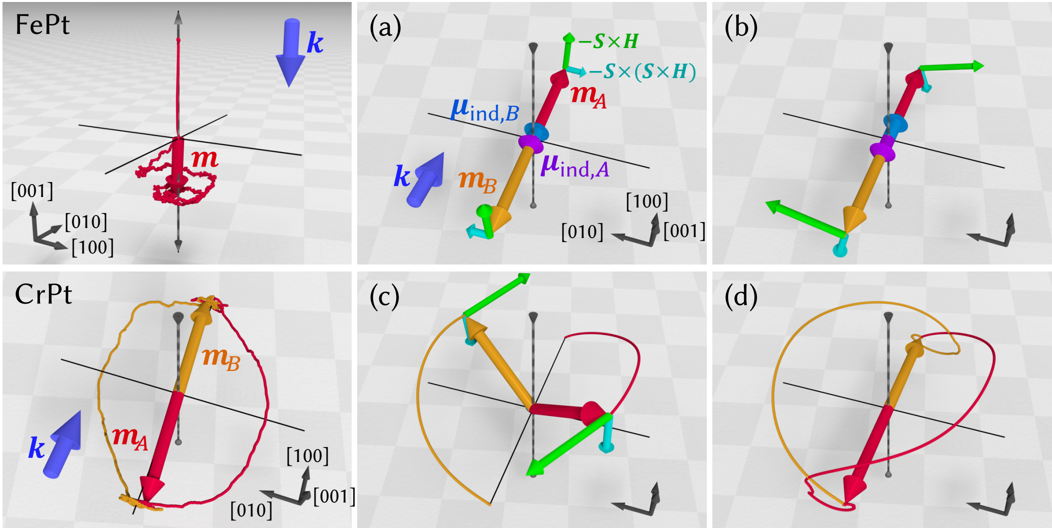
time-dependence: $\mu_{\text{ind}} \sim e^{-t/\tau}$

Statistics of the switching process



- switching process can be stochastic due to heating
- **deterministic switching** observed e.g. for $I \approx 6\mathrm{GW/cm}^2, \tau \gtrsim 200\mathrm{fs}$
- perpendicular switching also possible for lower intensities

Exchange-enhanced switching dynamics



- in the ferromagnet FePt swiching is linear
- in CrPt we find an **elliptical switching path**
- dynamics **exchange-enhanced** and driven by precessional torque
- very fast, time scale < 500fs

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Ritwik Mondal, Sebastian Großenbach, Levente Rózsa, Andreas Donges, and Uli Nowak
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R. Mondal, Charles University, Prague
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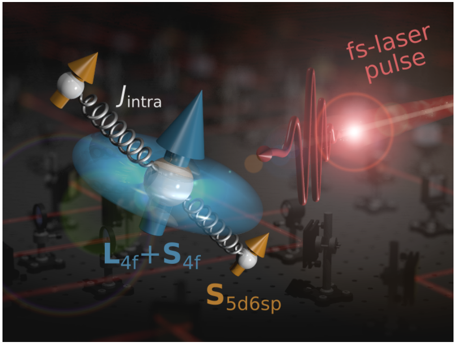
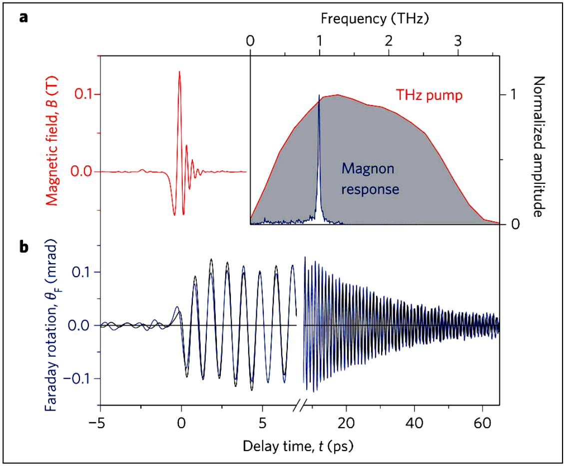
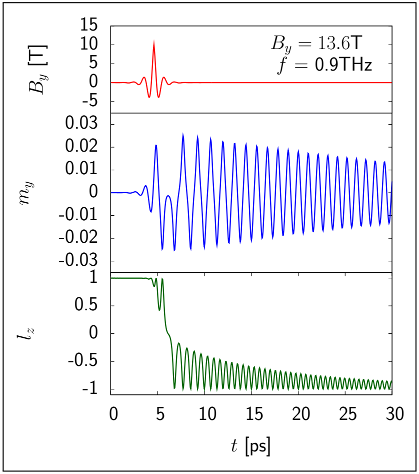


Excitation of NiO with single cycle THz pulses



Kampfrath et al., Nature Photonics **5**, *31(2010)*

- **resonant excitation** with B field component of an intense ultrashort THz laser pulse



Wienholdt et al., PRL **111**, *217202 (2013)*

- simulations show: even **switching** possible at 13.6T with frequency around 0.9THz

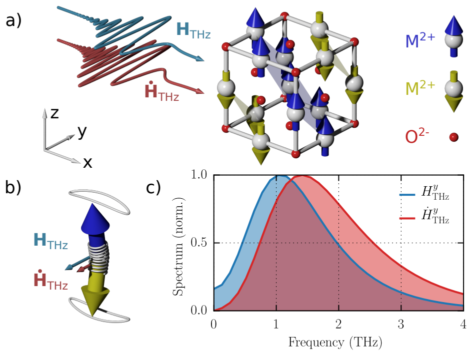
Beyond LLG: field derivative torque

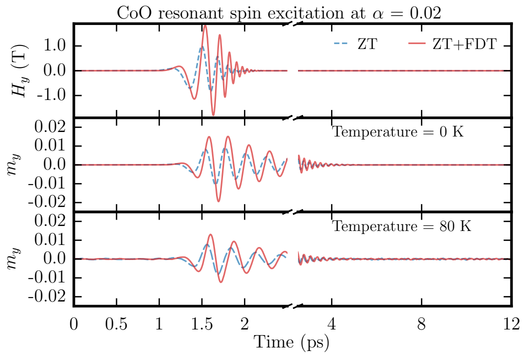
- equation of motion for spins derived starting from relativistic Dirac-Kohn-Sham equation
- several new extensions to LLG equation: one is a **field-derivative torque** *(Mondal et al., PRB* **94***, 144419 (2016))*

extended LLG equation

$$\dot{\boldsymbol{m}}_i = -\frac{\gamma}{(1+\alpha^2)\mu_s}\boldsymbol{m}_i \times \left(\boldsymbol{H}_i^{\text{eff}} - \frac{\alpha a^3}{\gamma}\frac{\partial \boldsymbol{H}}{\partial t}\right) - \frac{\gamma\alpha}{(1+\alpha^2)\mu_s}\boldsymbol{m}_i \times \left(\boldsymbol{m}_i \times \left(\boldsymbol{H}_i^{\text{eff}} - \frac{\alpha a^3}{\gamma}\frac{\partial \boldsymbol{H}}{\partial t}\right)\right)$$

- simulations of the stochastic LLG equation with field-derivative torque for CoO show:
- field-derivative torque leads to **phase shift and higher amplitude**





Mondal et al., PRB **100***, 060409R (2019)*

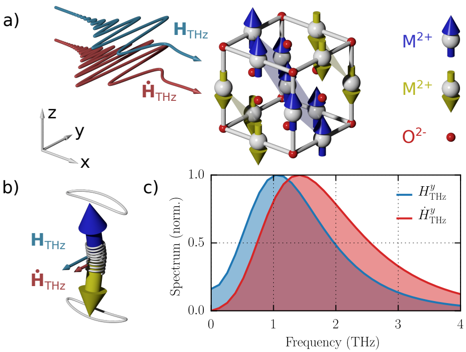
Beyond LLG: field derivative torque

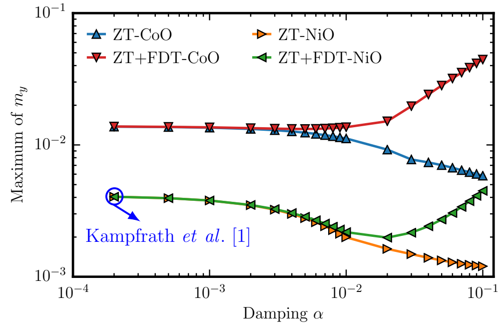
- equation of motion for spins derived starting from relativistic Dirac-Kohn-Sham equation
- several new extensions to LLG equation: one is a **field-derivative torque** *(Mondal et al., PRB* ***94****, 144419 (2016))*

extended LLG equation

$$\dot{\boldsymbol{m}}_i = -\frac{\gamma}{(1+\alpha^2)\mu_s}\boldsymbol{m}_i \times \left(\boldsymbol{H}_i^{\mathrm{eff}} - \frac{\alpha a^3}{\gamma}\frac{\partial \boldsymbol{H}}{\partial t}\right) - \frac{\gamma\alpha}{(1+\alpha^2)\mu_s}\boldsymbol{m}_i \times \left(\boldsymbol{m}_i \times \left(\boldsymbol{H}_i^{\mathrm{eff}} - \frac{\alpha a^3}{\gamma}\frac{\partial \boldsymbol{H}}{\partial t}\right)\right)$$

- simulations of the stochastic LLG equation with field-derivative torque for CoO show:
- effect **enhanced for high resonance frequency and spin-orbit coupling**





Mondal et al., PRB ***100****, 060409R (2019)*

Beyond LLG: nutation

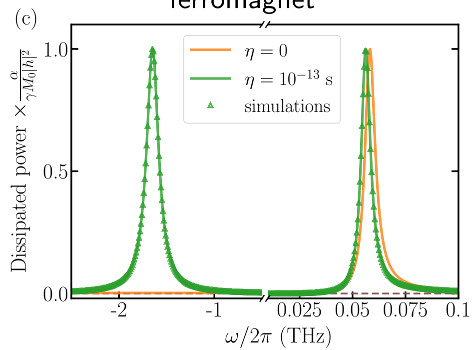
- another extension to LLG equation is a second order time derivative *(Mondal et al., PRB* **94***, 144419 (2016))*
- new effects: **intertia and nutation** *(Ciornei et al. PRB* **83***, 020410R (2011); Thonig et al., Sci. Rep.* **7***, 931 (2017); Neeraj et al., Nat. Phys.* **17***, 245 (2021))*

intertial LLG equation

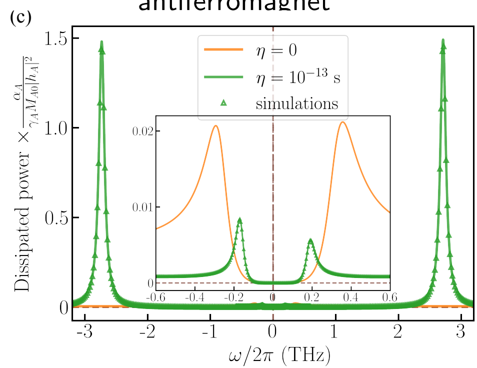
$$\dot{\boldsymbol{M}}_i = -\gamma_i \boldsymbol{M}_i \times \boldsymbol{H}_i + \frac{\alpha_i}{M_{i0}} \boldsymbol{M}_i \times \dot{\boldsymbol{M}}_i + \frac{\eta}{M_{i0}} \boldsymbol{M}_i \times \ddot{\boldsymbol{M}}_i$$

- calculations of the (A)FMR resonance spectra show:
  - precession frequencies decrease
  - inertia reduces the effective damping
  - additional high-frequency nutation peaks
  - effects larger in AFMs

ferromagnet



antiferromagnet



Mondal et al., PRB **103***, 104404 (2021)*

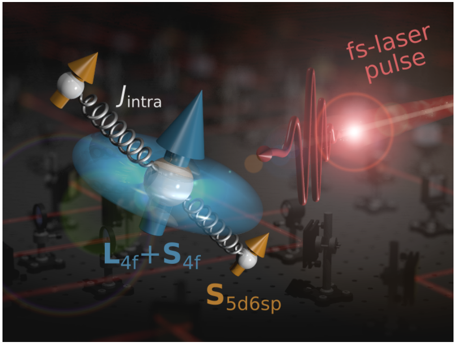
Spin dynamics: the Landau-Lifshitz equation and beyond

Martin Evers, Hannah Lange, Andreas Donges, and Uli Nowak
University of Konstanz, Germany

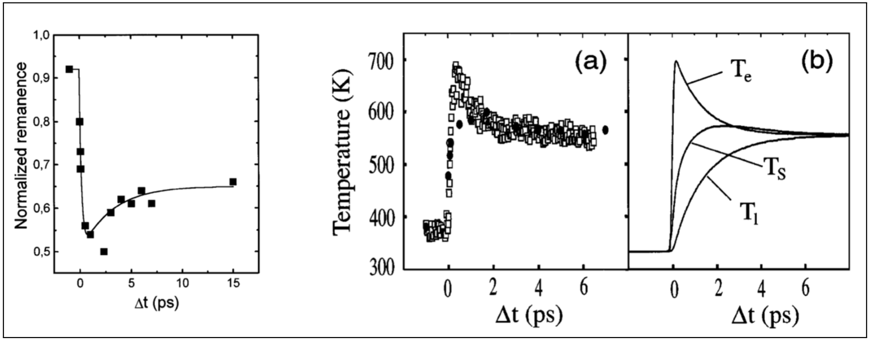
In collaboration with:
S. Tauchert, M. Volkov, D. Ehberger, D. Kazenwadel, P. Baum, University of Konstanz
A. Book, W. Kreuzpaintner, LMU München

Topics:

- introduction: modelling spin dynamics
- orbital-resolved spin models and inverse Faraday effect in AFMs
- beyond LLG equation:
  field-derivative torques and nutation
- **ultrafast transfer of spin angular momentum into the lattice**

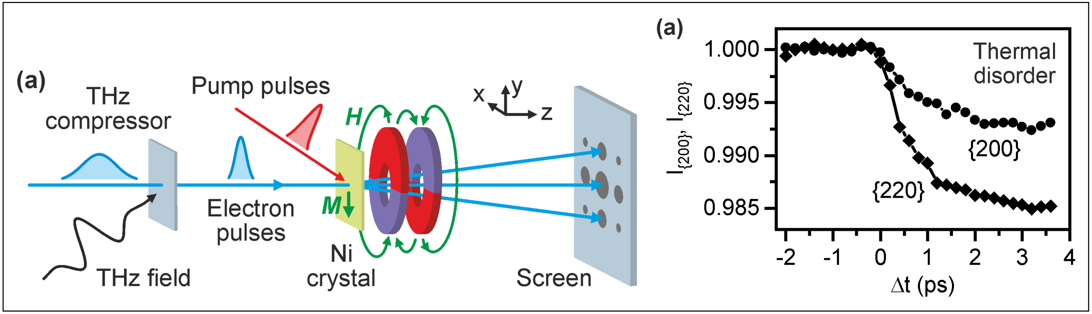


Laser-induced ultrafast spin dynamics



- following a fs laser pulse **magnetisation can break down on a time scale of some hundred femtoseconds and recover on a ps time scale**
  (*Beaurepaire et al., PRL* **76**, *4250 (1996)*)
- the phenomenological **three-temperature model** gives an idea of the energy flow
- flow of **angular momentum** remains an open question
- coupling between **microscopic degrees of freedom** (spin, electrons, lattice) hardly understood

Ultrafast generation of chiral phonons

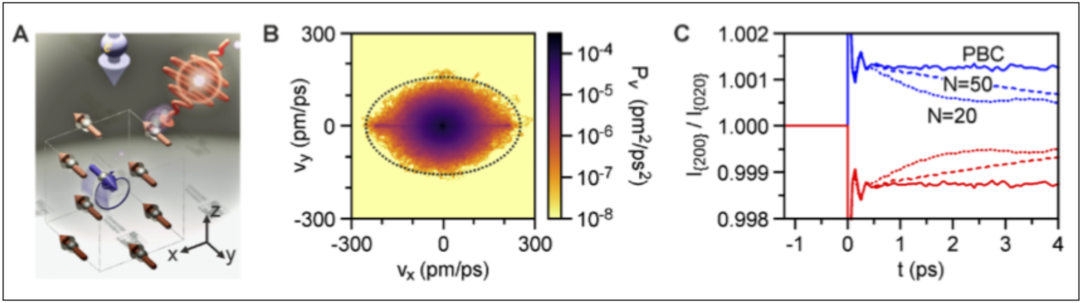


experiment by *S. Tauchert et al.*

- **time-resolved electron diffraction** delivers structural dynamics and atomic motions with picometer and femtosecond resolutions in space and time
- THz fields compress electron pulses to $<$ 100 fs duration
- electron diffraction of a single-crystalline Ni layer on Si membrane reveals the epitaxy and distinct **Bragg spots**
- when laser pulse hits the sample (2 2 0) spot decays by $\approx$ 1.5 % with a $<$ 500 fs time constant indicating the ultrafast population of phonons

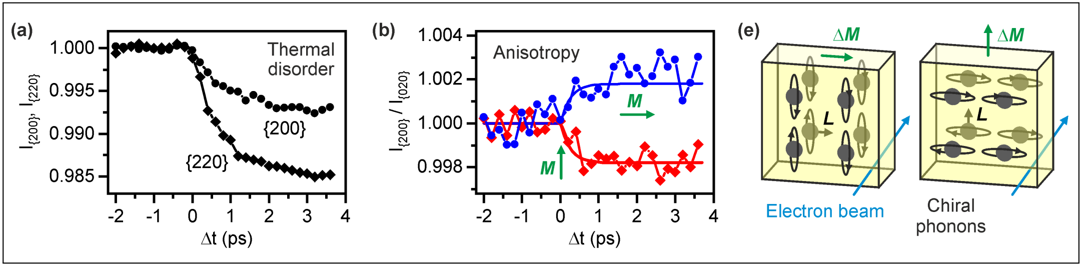
Ultrafast generation of chiral phonons: theory

How can we detect the **phonon angular momentum**?



- idea: develop a model for local phonon excitations of the lattice with finite angular momentum $L = \hbar \boldsymbol{e}_M = m \Delta \boldsymbol{r} \times \boldsymbol{p}$
- number of local excitation corresponding to a demagnetization of 50 %
- **molecular dynamics simulations** using LAMMPS: free Ni crystal with 500000 atoms
- calculate and analyze diffraction patterns
- broadaning of (200) and (020) peaks different $\Rightarrow$ **anisotropy indicates chiral character of phonons**

Ultrafast generation of chiral phonons



experiment by *S. Tauchert et al.*

- after laser excitation time-resolved electron diffraction shows an **anisotropy of the (normally equivalent) Bragg peaks within less then a ps**
- this indicates a **transfer of angular momentum from the spin system into the lattice** within less then a ps
- this is not an ultrafast Einstein-de Haas effect *(Dornes et al., Nature* **565**, *209 (2019))*

Spin lattic coupling

spin-lattice Hamiltonian

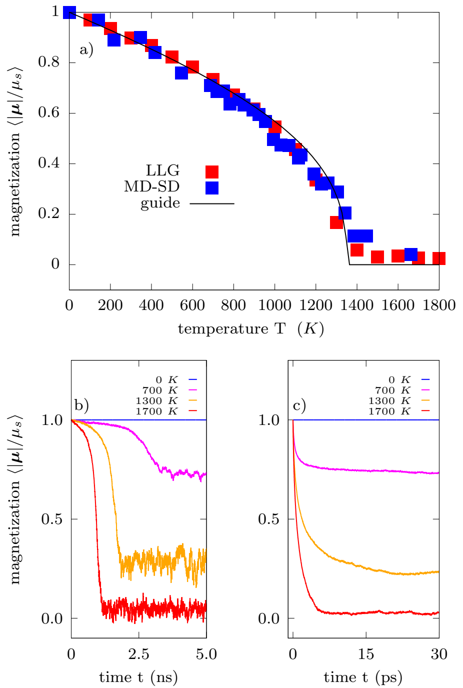
$$\mathcal{H} = -\underbrace{\sum_{i,j} J(r_{ij})(\boldsymbol{S}_i \cdot \boldsymbol{S}_j)}_{\text{exchange}} + \underbrace{\lambda \sum_{i,j} \frac{(\boldsymbol{S}_i \cdot \boldsymbol{r}_{ij})(\boldsymbol{r}_{ij} \cdot \boldsymbol{S}_j)}{r_{ij}{}^6}}_{\text{spin-lattice}}$$

$$\underbrace{-\mu_s \boldsymbol{B} \cdot \sum_i \boldsymbol{S}_i}_{\text{Zeeman}} + \underbrace{\sum_i \frac{p_i^2}{2m_i}}_{\text{kinetic}} + \underbrace{\sum_{i,j} V(\boldsymbol{r}_{ij})}_{\text{lattice}}$$

equations of motion

$$\dot{\boldsymbol{r}}_i = \frac{\partial \mathcal{H}}{\partial \boldsymbol{p}_i} \qquad \dot{\boldsymbol{p}}_i = -\frac{\partial \mathcal{H}}{\partial \boldsymbol{r}_i} \qquad \dot{\boldsymbol{S}}_i = \frac{\gamma}{\mu_s} \boldsymbol{S}_i \times \frac{\partial \mathcal{H}}{\partial \boldsymbol{S}_i}$$

- **no heat bath** neccessary
- same equilibrium magnetisation as with stochhastic LLG equation but different dynamics
- see also: *Strungaru et al., PRB* **103***, 024429 (2021)*



Aßmann et al, JMMM **469***, 217 (2019)*

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Schwerpunktprogramm Skyrmionics

SFB 1214 Anisotropic nanoparticles

SFB 1432 Fluctuations

Summary: Landau-Lifshitz equation and beyond

- **stochastic LLG equation**:
  - ultrafast demagnetization with orbital-resolved spin models for Gd and Tb
  - switching with the inverse Faraday effect in the antiferromagnet CrPt
- **beyond LLG equation**:
  - THz dynamics with field-derivative torques in NiO and CoO
  - (A)FMR with intertial dynamics and nutation
- **no LLG equation** (but should be connected):
  - ultrafast transfer of spin angular momentum into the phonon systems
  - outlook: a framework for spin-lattice dynamics