Spin dynamics in antiferromagnets: from THz to ultrafast switching

Uli Nowak
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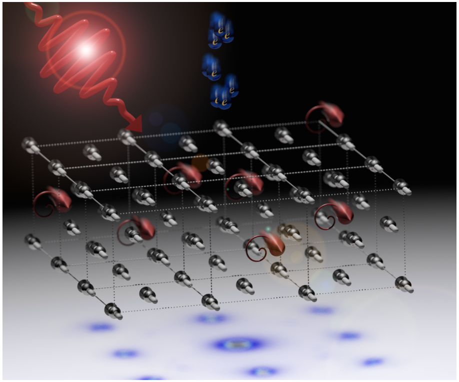
Topics:

- ultrafast spin dynamics in AFMs
- ultrafast transfer of spin angular momentum into the lattice
- spin lattice dynamics



Spin dynamics in antiferromagnets: from THz to ultrafast switching

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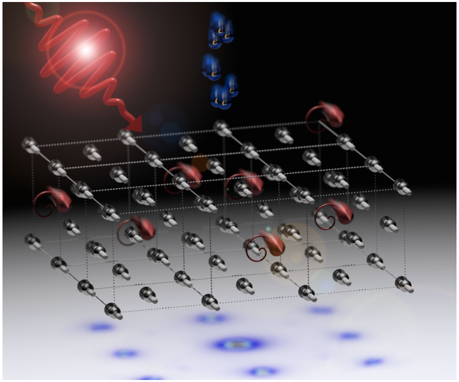
In collaboration with:
M. Berritta, P. Oppeneer, University of Uppsala
C. Karva, Charles University, Prague



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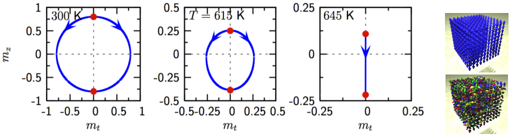
Switching with inverse Faraday effect

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

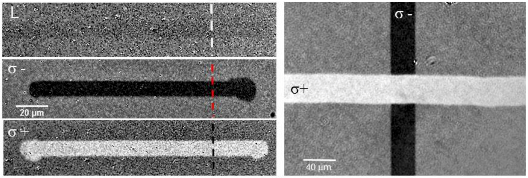


(Kazantseva et al., Europhys. Lett. **86**, *27006 (2009))*

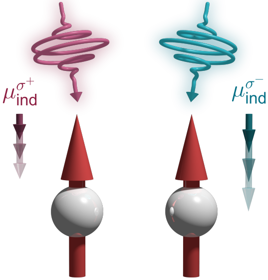
- due to **inverse Faraday effect:** circularly polarized light induces magnetization in a sample *(Battiato et al. PRB* **89**, *0144413 (2014))*

- **what about antiferromagnets?**

all-optical writing of an FePt nanoparticle recording medium

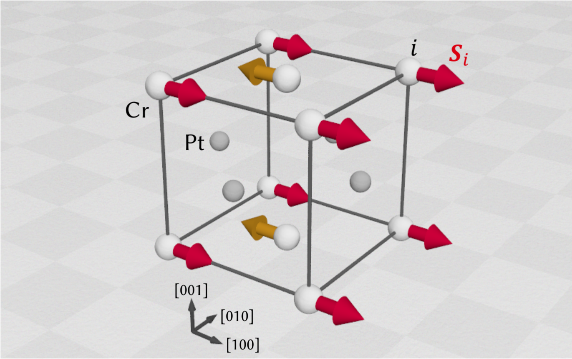


John et al., Scientific Reports **7**, *4114 (2017)*

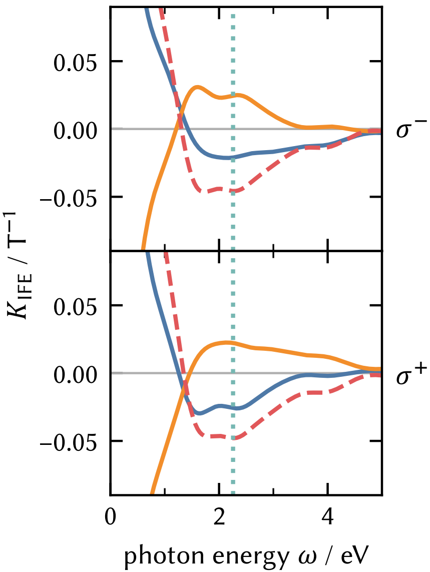


Inverse Faraday effect in CrPt

- 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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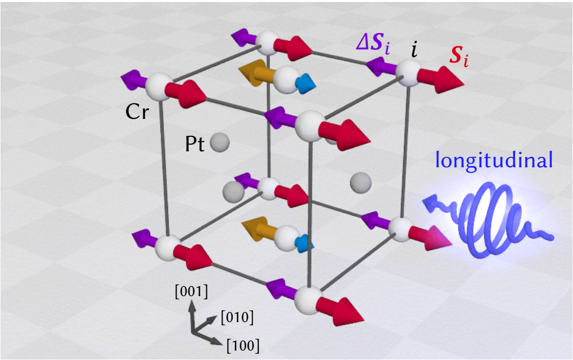
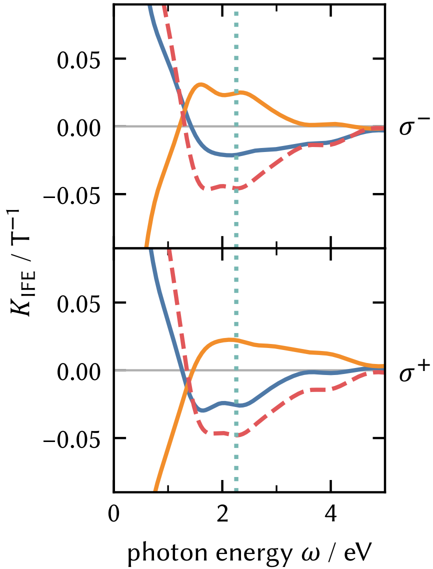


- IFE leads to **staggered induced moments**



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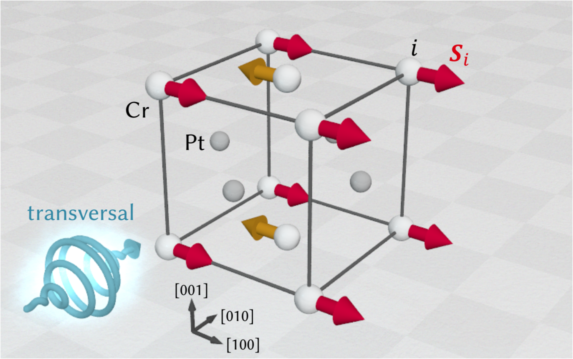
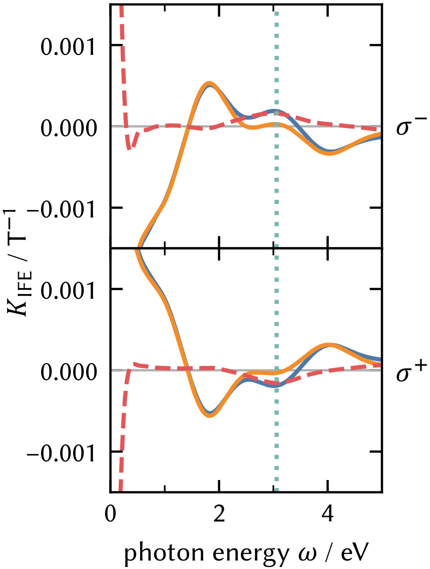




IFE constants Cr moment direction parallel and antiparallel to *k*-vector of the light. Red line indicates difference.

- perpendicular IFE very small

Modelling ultrafast spin dynamics in CrPt

- **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:

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

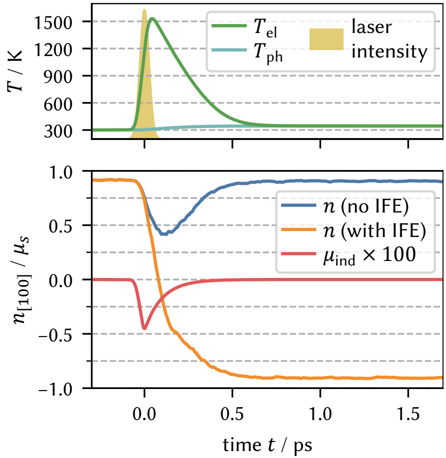
spin model

$$\mathcal{H} = -\sum_{\langle ij\rangle} J_{ij}(\boldsymbol{S}_i + \Delta\boldsymbol{S}_i)\cdot(\boldsymbol{S}_j + \Delta\boldsymbol{S}_j) - \sum_i d_z S_{iz}^2 - 4 d_{ip} S_{ix}^2 S_{iy}^2$$

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

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

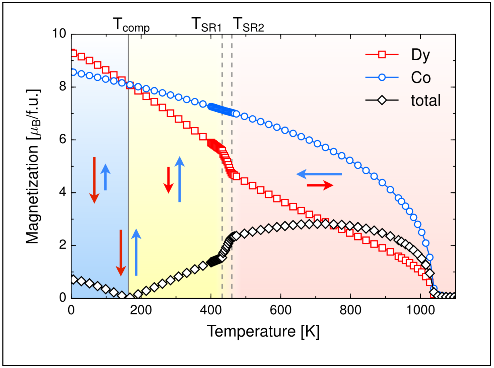
α: quantifies coupling to the electronic/phononic heat bath

spin dynamics

- **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**

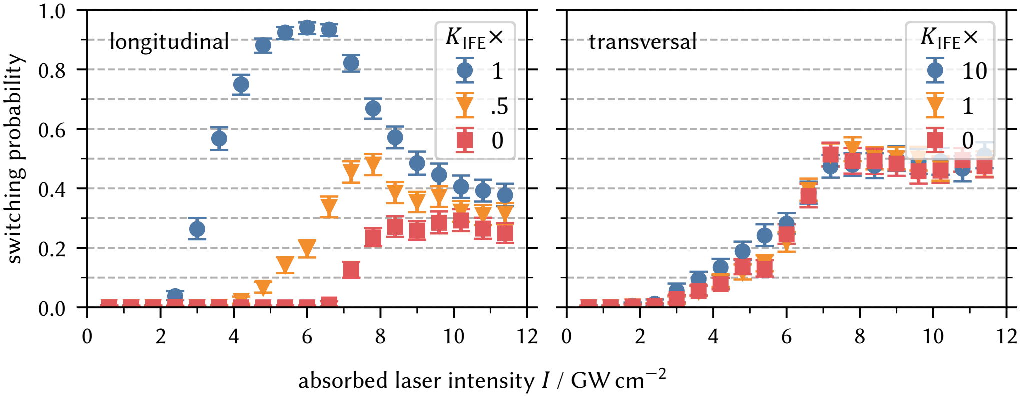
☺ realistic dispersion relations; non-linear prozesses; critical behavior methods

☹ classical approximation; large numerical effort



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

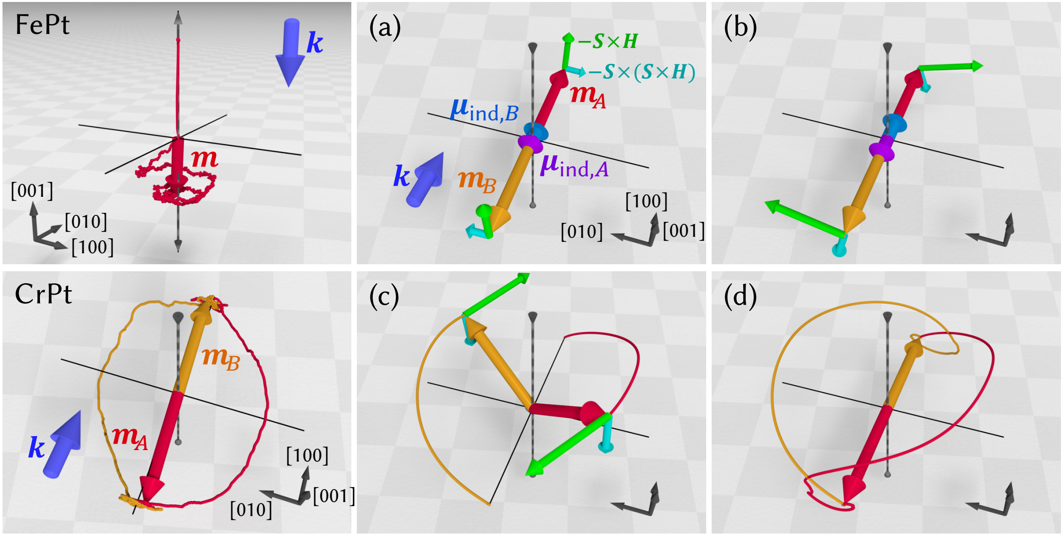
Statistics of the switching process



Dannegger et al., PRB **104**, *L060413 (2021)*

- 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



Dannegger et al., PRB ***104****, L060413 (2021)*

- 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

Spin dynamics in antiferromagnets: from THz to ultrafast switching

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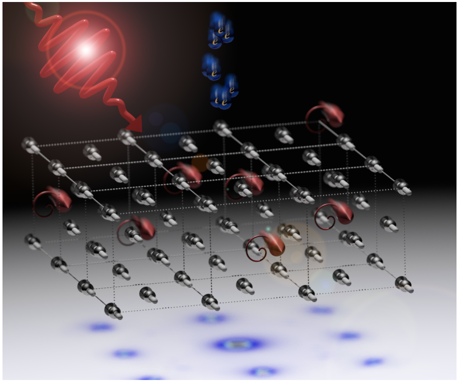

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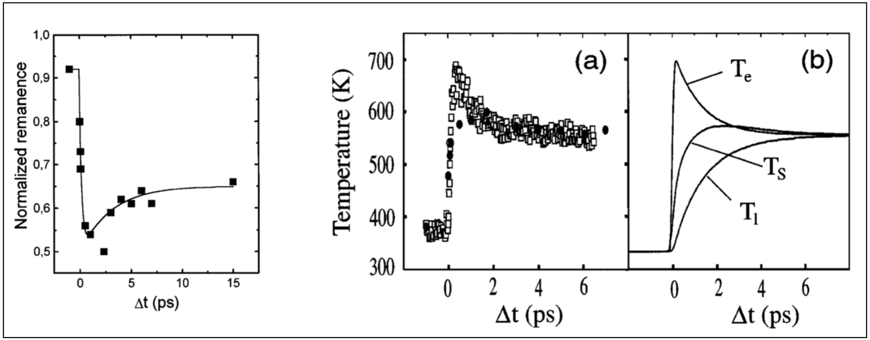
Laser-induced ultrafast spin dynamics

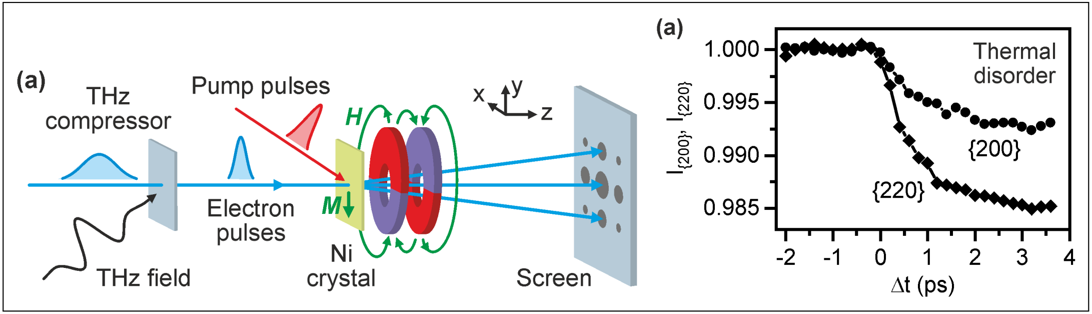


- following a fs laser pulse **magnetisation can break down on a time scale of some hundred femtoseconds and recover on a ps time scale**

  (*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

Ultrafast generation of phonons

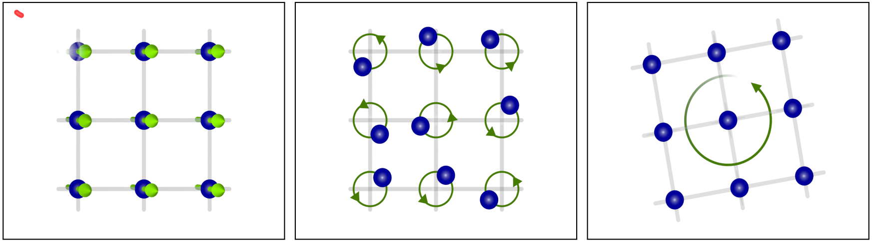


S. Tauchert et al., Nature 602, *73 (2022)*

- **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 polarized phonons: idea and 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 ⇒ **anisotropy indicates chiral character of phonons**

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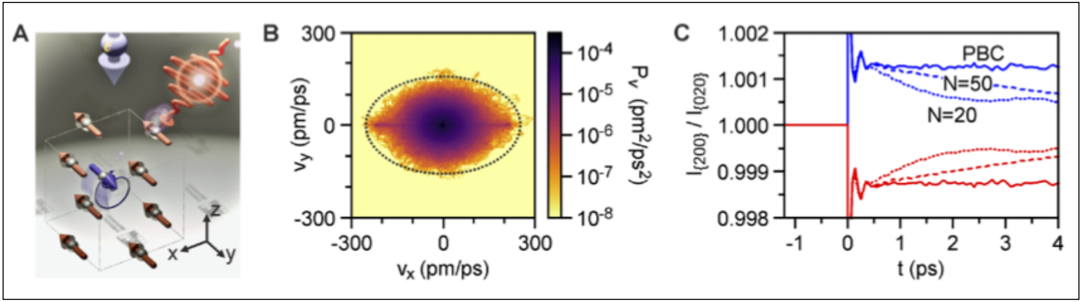

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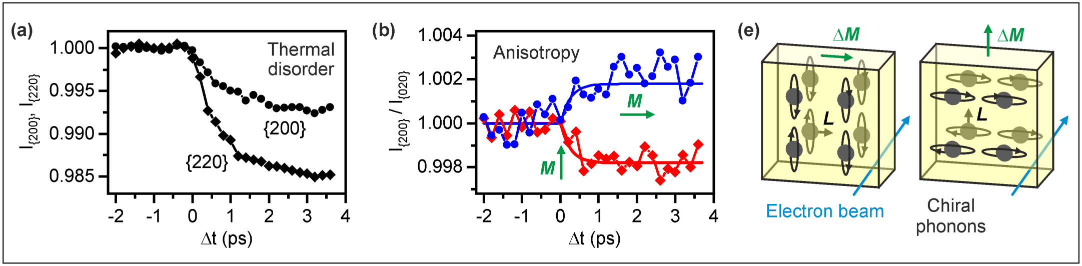
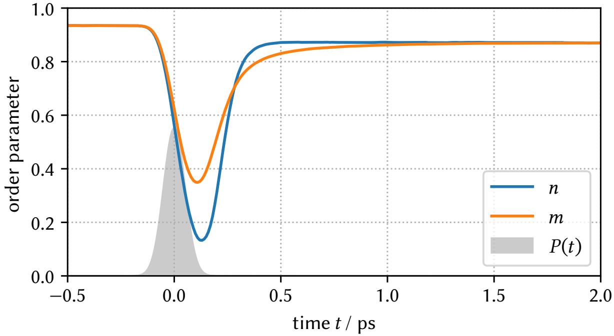
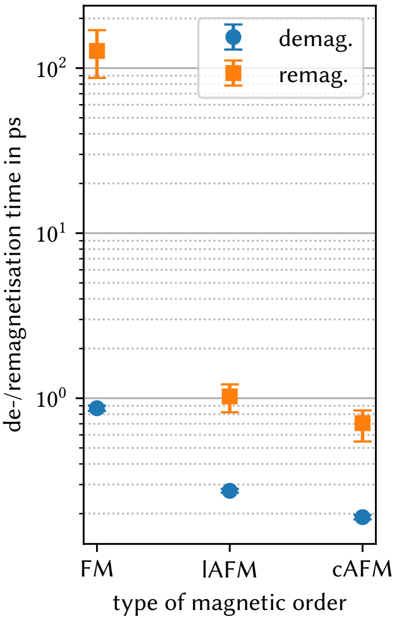


S. Tauchert et al., Nature **602**, *73 (2022)*

- 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))*

De- and remagnetization times in FMs and AFMs





- toy models for a FM, a layered AFM and a cubic AFM are compared
- all model parameters identical except sign of exchange constants
- simulation of sLLG equation
- even then **thermal dynamics is quicker in AFMs**

Spin dynamics in antiferromagnets: from THz to ultrafast switching

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In collaboration with:
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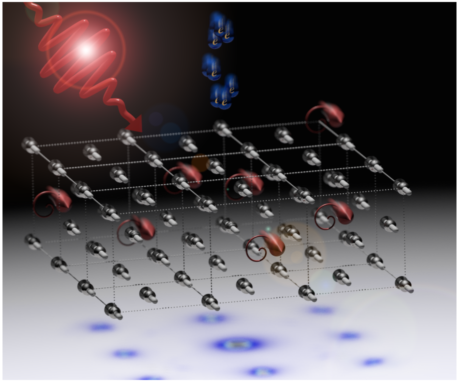

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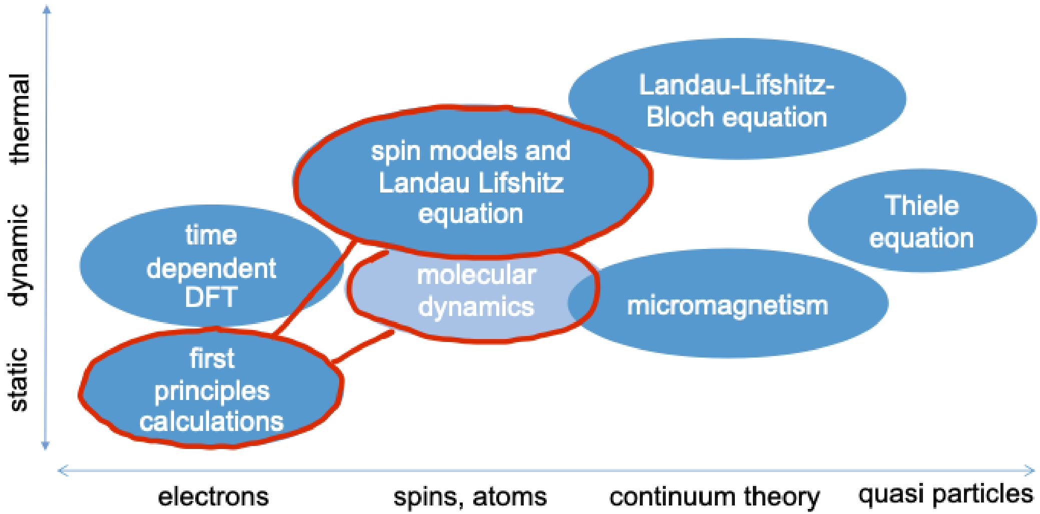
Multi-scale modelling in magnetism

- **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



Spin-molecular dynamics

Hamiltonian for spin and lattice degrees of freedom

$$\mathcal{H}(\boldsymbol{S},\boldsymbol{r},\boldsymbol{p}) = \underbrace{-\sum_{i,j} J(\boldsymbol{S}_i\cdot\boldsymbol{S}_j)}_{\text{spin}} + \underbrace{\mathcal{H}_{sl}(\boldsymbol{S},\boldsymbol{r})}_{\text{spin-lattice}} + \underbrace{\sum_i \frac{\boldsymbol{p}_i^2}{2m_i} + \sum_{i,j} V(\boldsymbol{r}_{ij})}_{\text{lattice}}$$

equations of motion

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

- **no heat bath** $\rightarrow$ microcanonic ensemble
- open problems:
  - expression for spin-lattice coupling
  - determination of the parameters
- see also:
  Strungaru et al., PRB **103**, *024429 (2021)*
  Hellsvik et al., PRB **99**, *104302 (2019)*

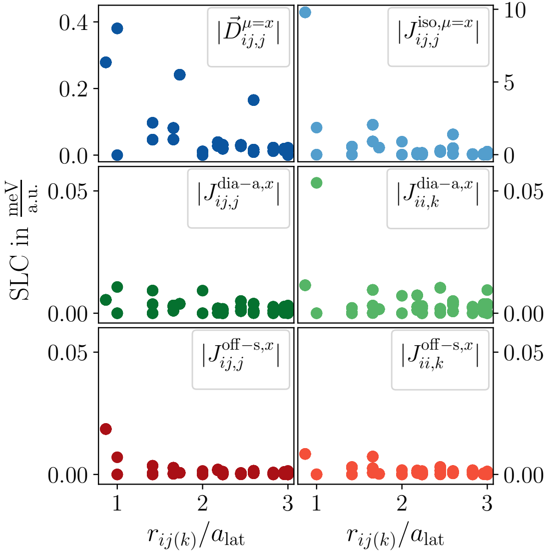
Aßmann et al, JMMM **469**, *217 (2019)*

Spin lattice interactions from first principles

spin-lattice Hamiltonian

$$\mathcal{H}_{sl} = -\sum_{i,j,\alpha,\beta} J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta - \sum_{i,j,\alpha,\beta}\sum_{k,\mu} J_{ij,k}^{\alpha\beta,\mu} S_i^\alpha S_j^\beta u_k^\mu - \sum_{i,j,\alpha,\beta}\sum_{k,l,\mu,\nu} J_{ij,kl}^{\alpha\beta,\mu\nu} S_i^\alpha S_j^\beta u_k^\mu u_l^\nu$$

- **fully-relativistic scheme** treats changes spin configuration plus atomic positions eually
- closed expressions for the **spin-lattice coupling parameters** up to any order
- even in bcc Fe leading term for angular momentum transfer is a **DMI-type interaction** emerging due to the symmetry breaking distortion of the lattice.
- *Mankovsky, Polesya, Lange, Weißenhofer, Nowak, Ebert, submitted*



Thanks to . . .

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- *R. Mondal*, Charles University, Prague
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