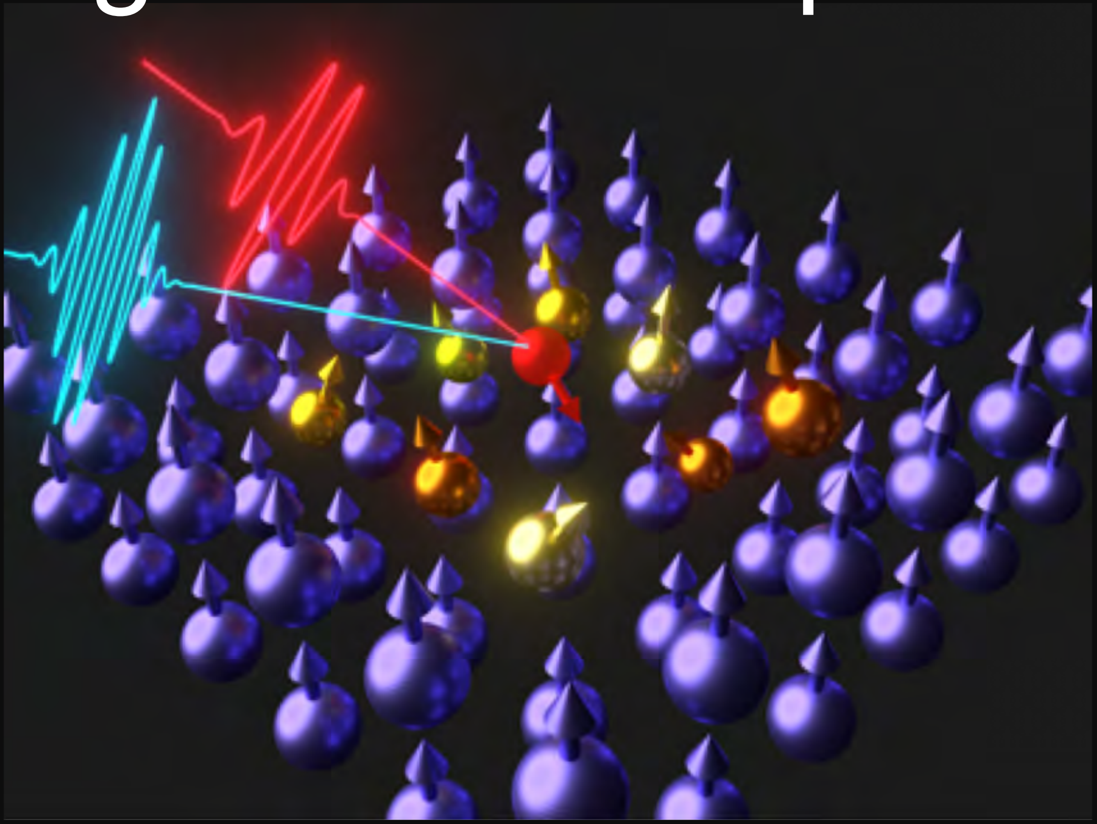
Femtosecond measurement of direct light-induced spin transfer



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Current Affiliation: Ecole Polytechnique Fédérale de Lausanne

Outline for my talk

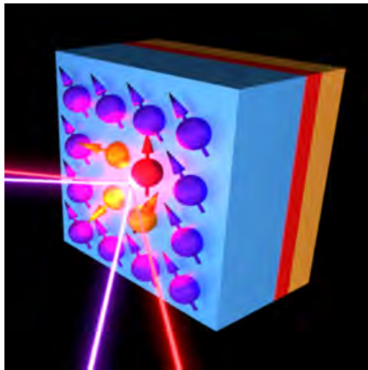
Ultrafast Ferromagnetism

- Previous understanding:
    - Ultrafast demagnetization takes place in 100-200 fs

- Introduction to extreme ultraviolet Magneto-optical Kerr Spectroscopy
    - Element Specific Information
    - 10fs bursts, very high time resolution

- New Understanding: Spin excitations within few femtoseconds!
    - Ultrafast magnetic phase transition in Ferromagnetic Nickel
    - Direct optical spin transfer between different elements in Co2MnGe

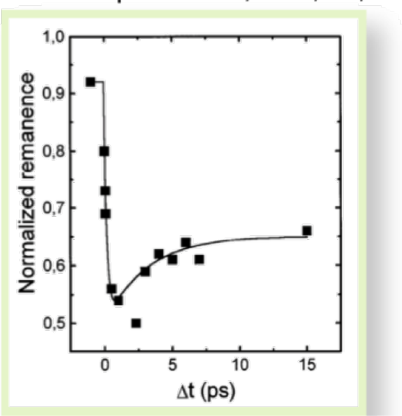
Zusin, D., Tengdin, P., et. al. PRB 97, 024433 (2018)
Tengdin, P. et. al. Sci. Adv. 4 : eaap9744 (2018)
You, W., Tengdin, P., et. al. PRL 121, 077204 (2018)
Tengdin, P. et al., Sci. Adv. in press (2019)
Hofherr, M., et al., Sci. Adv. in press (2019)

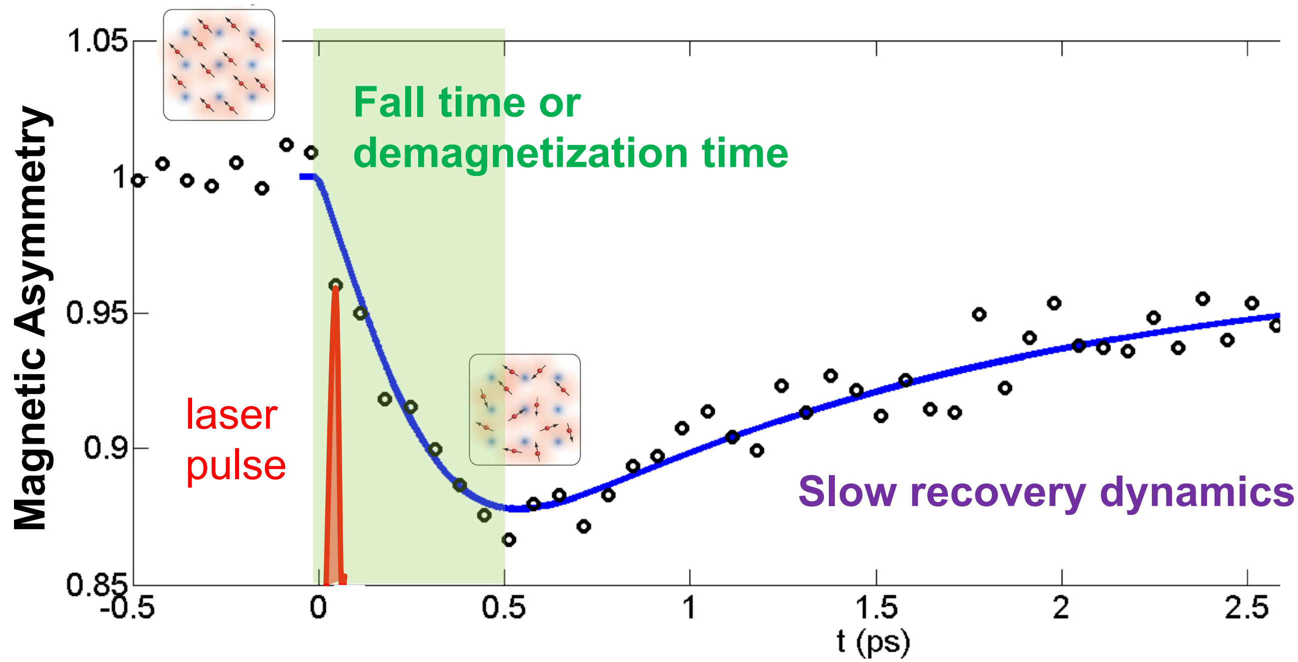


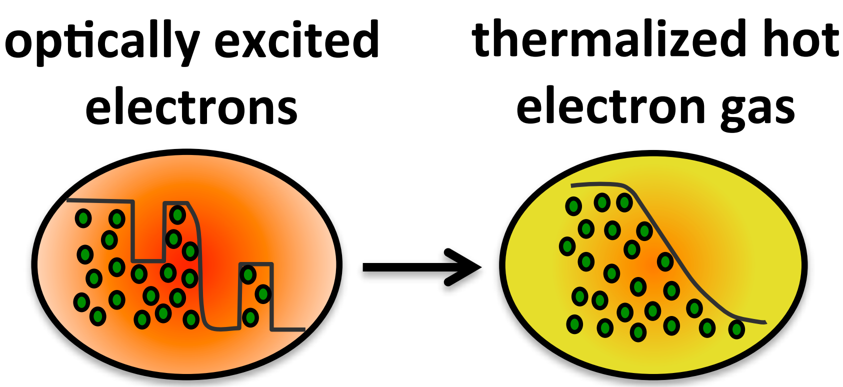
The puzzle of light-induced ultrafast demagnetization

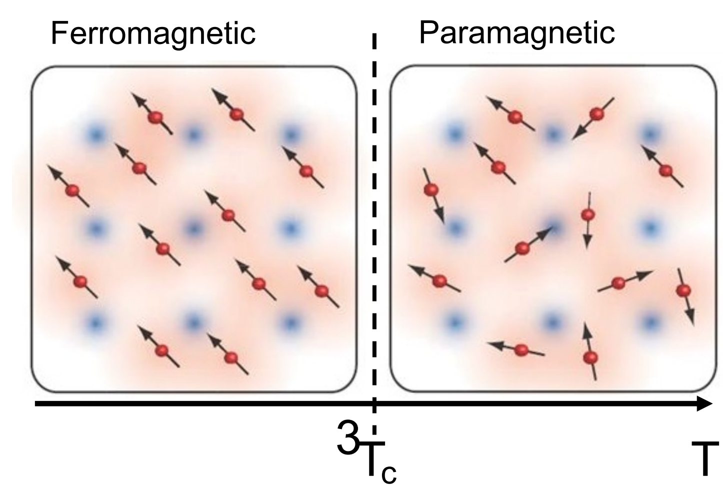
Demagnetization by fs Laser

E Beaurepaire et. al, PRL, 76, 4250 (1996









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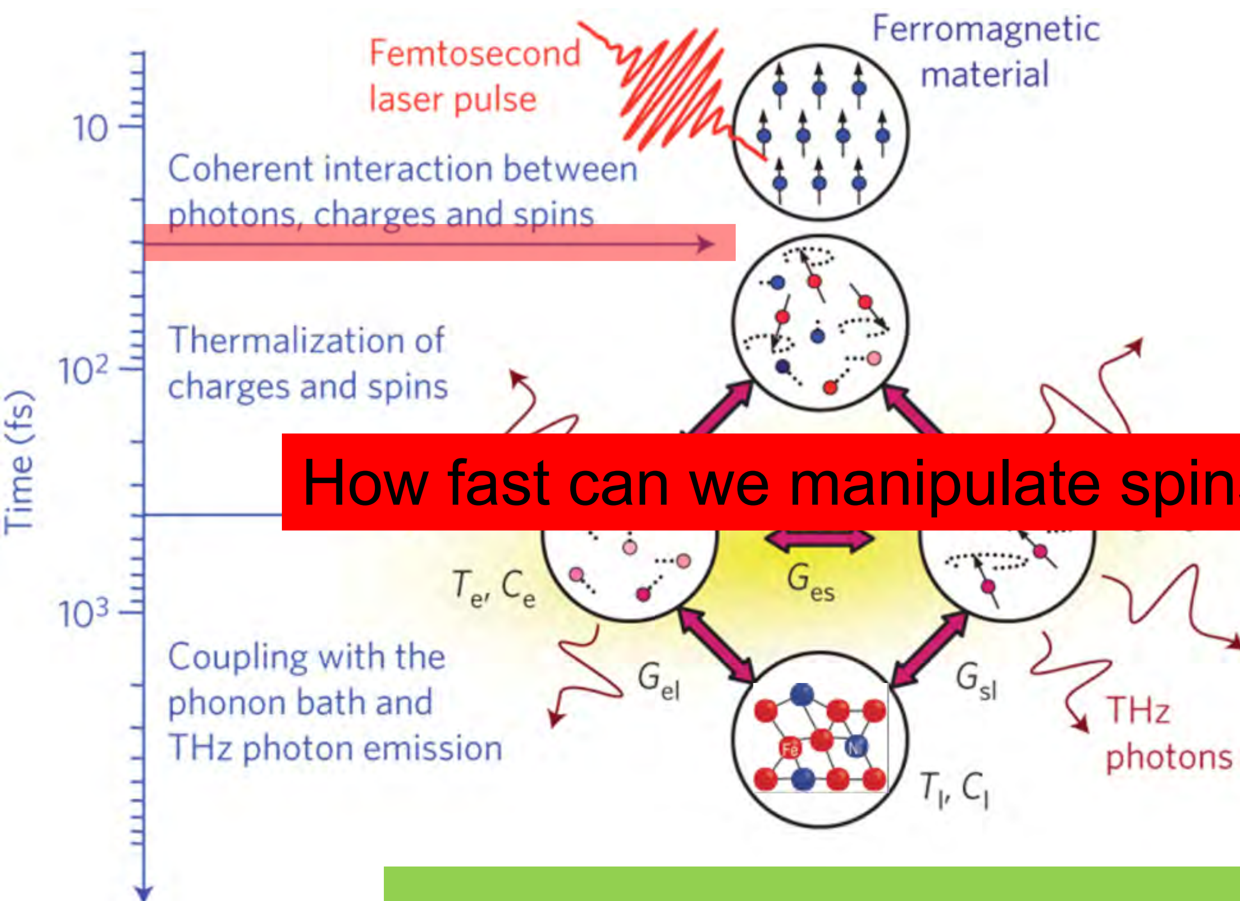
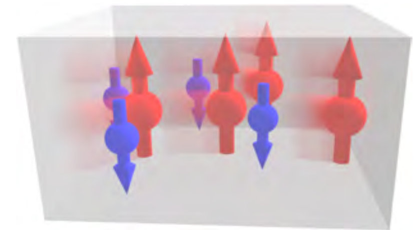
Coupled dynamics of spins/charge/photons: past work assumed that energy flowed from charges, to spins, to lattice



How fast can we manipulate spins with light?

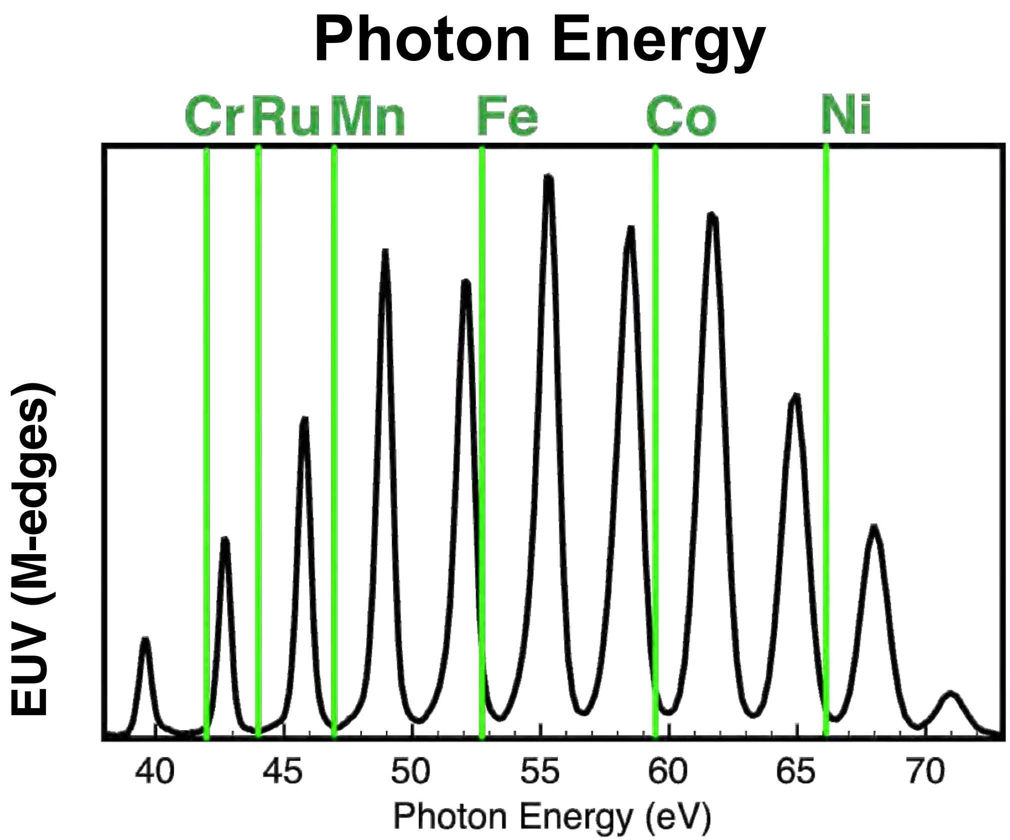


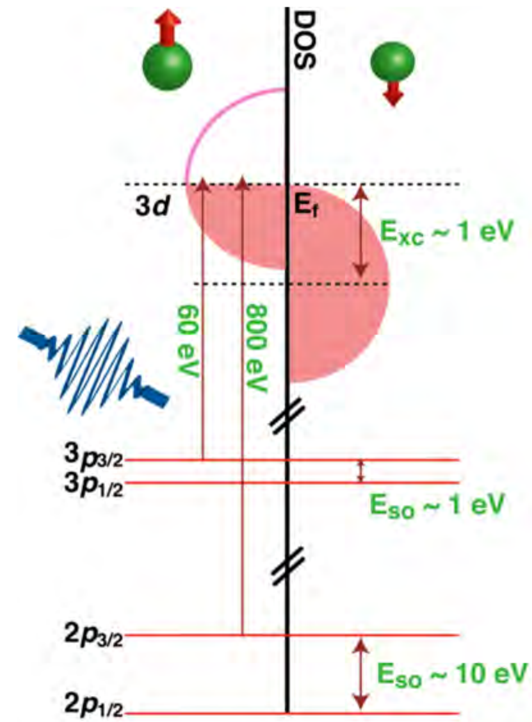
Superdiffusive spin transport : Battiato *et al.*, *PRL* **105**, 27203 (2010)

Bigot et al, Nature Physics ***5****, 515 (2009)*
Koopmans et al., Nature Mater. ***9****, 259 (2010)*

- Spin-flip scattering: Koopmans *et al.*, *Nature Mater.* **9**, 259 (2010)
- Spin-orbit coupling: Töws *et al.*, *PRL* **115**, 217204 (2015)
- Superdiffusive spin transport : Battiato *et al.*, *PRL* **105**, 27203 (2010)
- *Rev. Mod. Phys.* **82**, 2731 (2010)

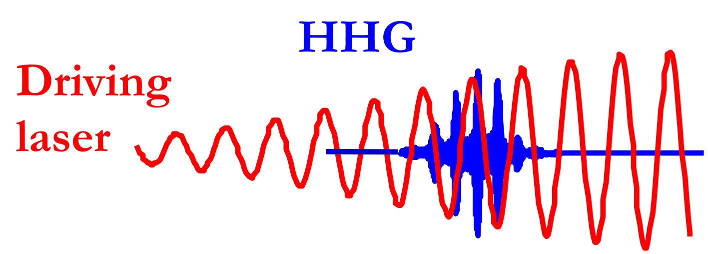
High harmonic generation: ideal for probing ultrafast spin dynamics





UNIQUE ADVANTAGES

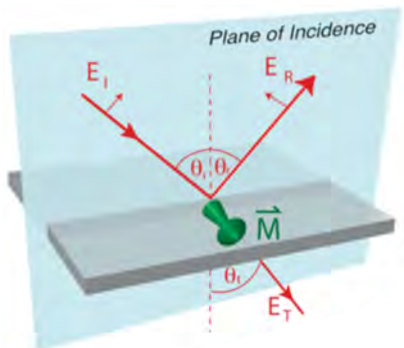
- Element selective
- Multiple elements simultaneously
- <<10fs time resolution, no jitter
- Span entire M edges, L edges



attosecond pulses

Rundquist et al, *Science* ***280****, 1412 (1998)*
Chen et al., PRL **105**, 173901 (2010)
Popmintchev et al, *Science* ***336****, 1287 (2012)*

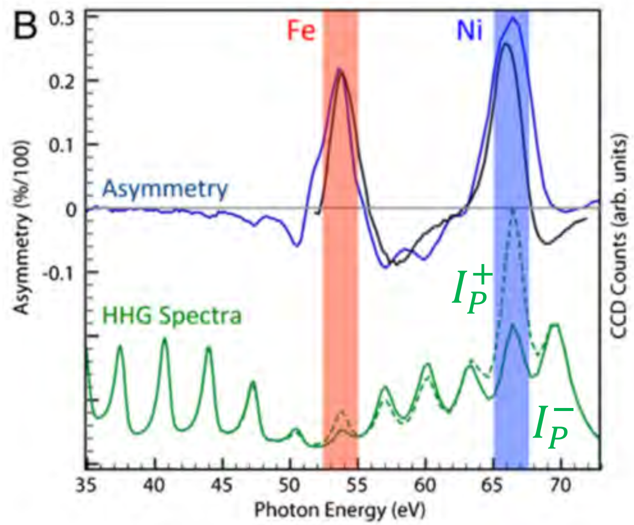
Experimental setup

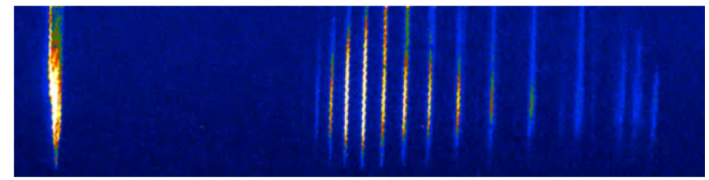


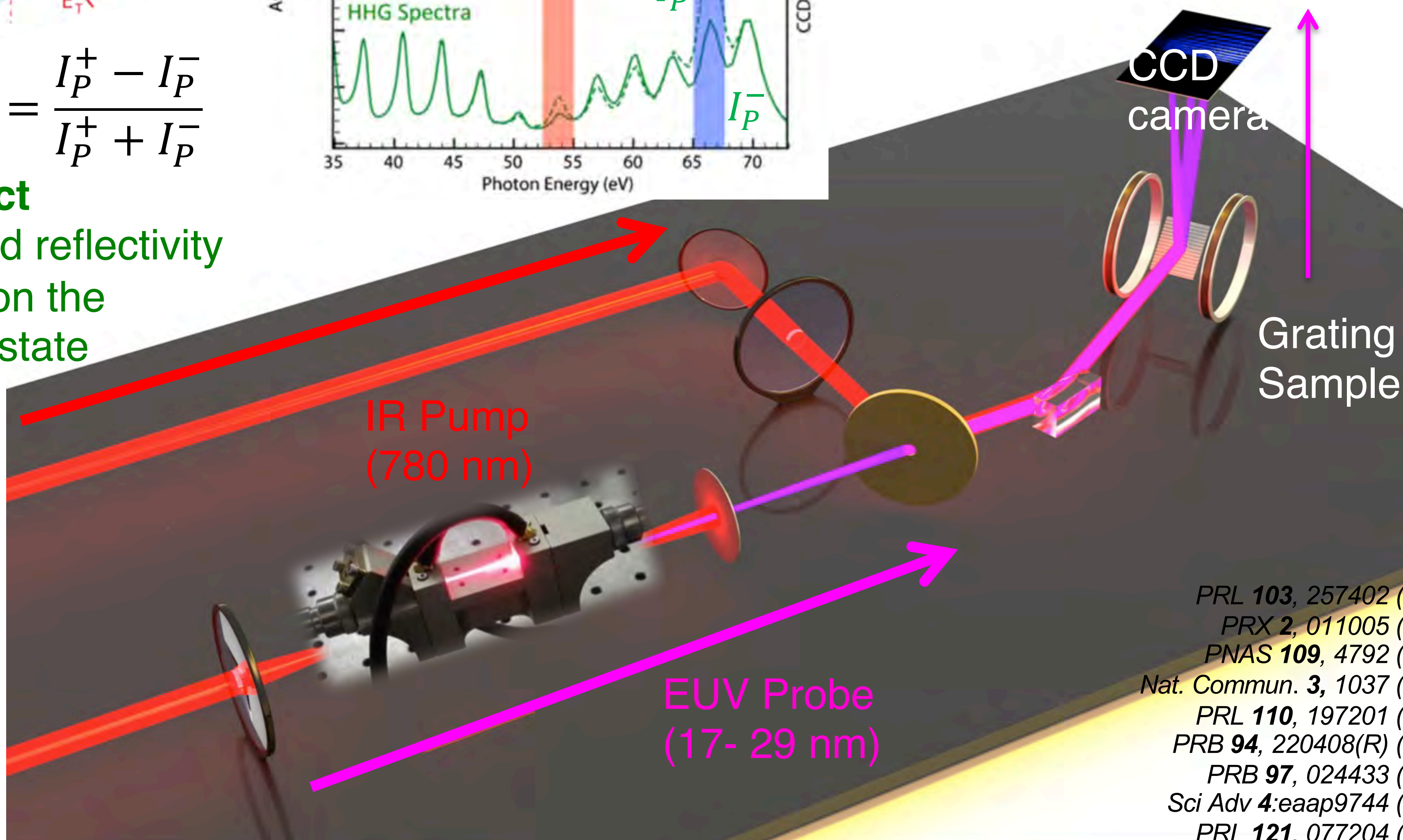
$$\vec{M} \;\alpha\; A = \frac{I_P^+ - I_P^-}{I_P^+ + I_P^-}$$

Kerr Effect

p-polarized reflectivity depends on the magnetic state

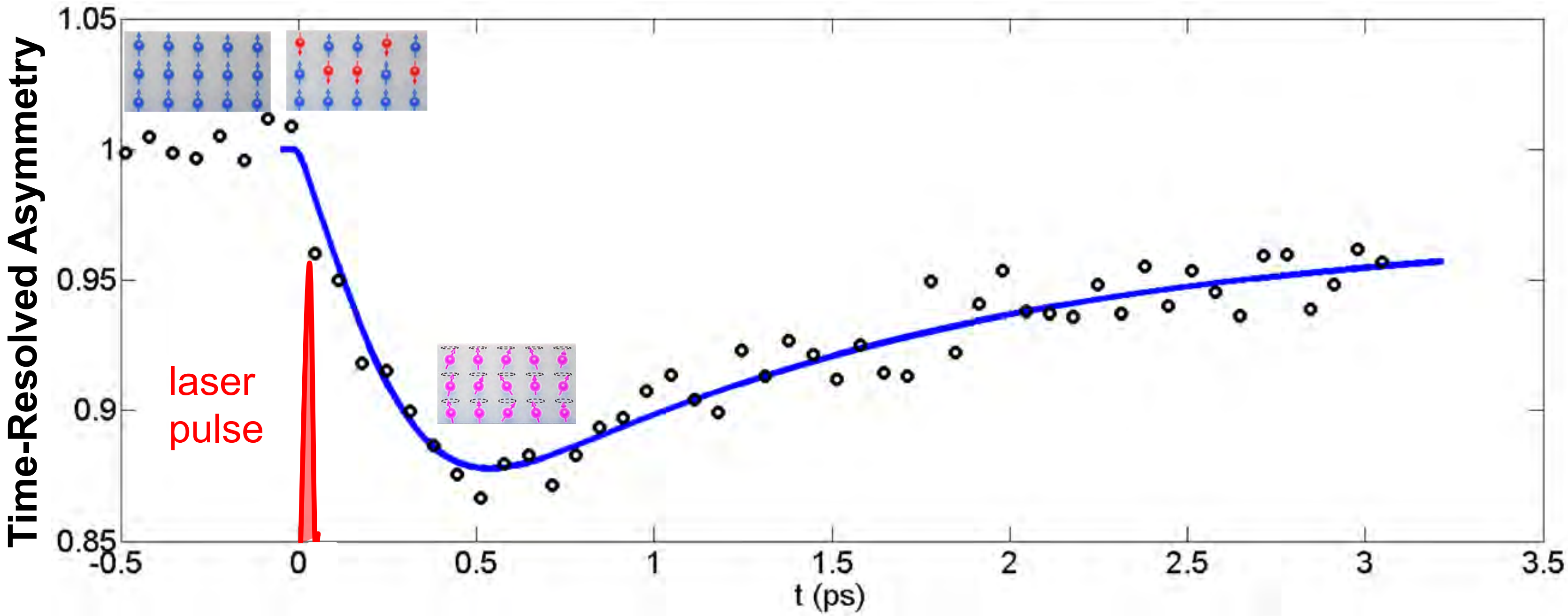


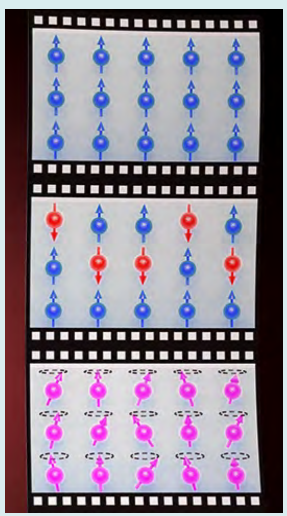




PRL ***103****, 257402 (2009)*
PRX ***2****, 011005 (2012)*
PNAS ***109****, 4792 (2012)*
Nat. Commun. ***3****, 1037 (2012)*
PRL ***110****, 197201 (2013)*
PRB ***94****, 220408(R) (2016)*
PRB ***97****, 024433 (2018)*
Sci Adv ***4****:eaap9744 (2018)*
PRL ***121****, 077204 (2018)*

Direct manipulation of spins:
ultrafast spin excitation in two different materials





20 fs

200 fs

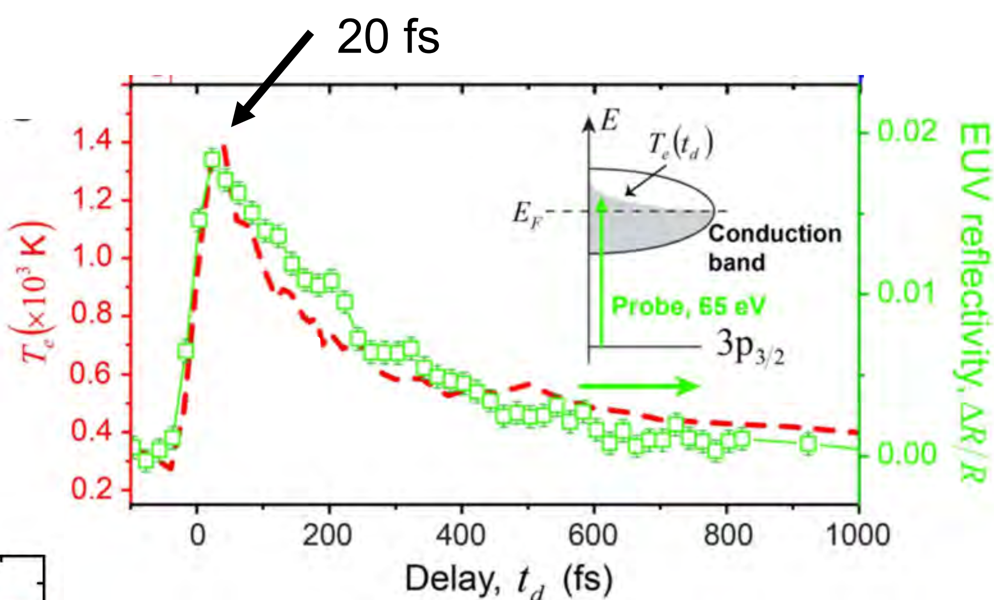
Ferromagnetic Nickel

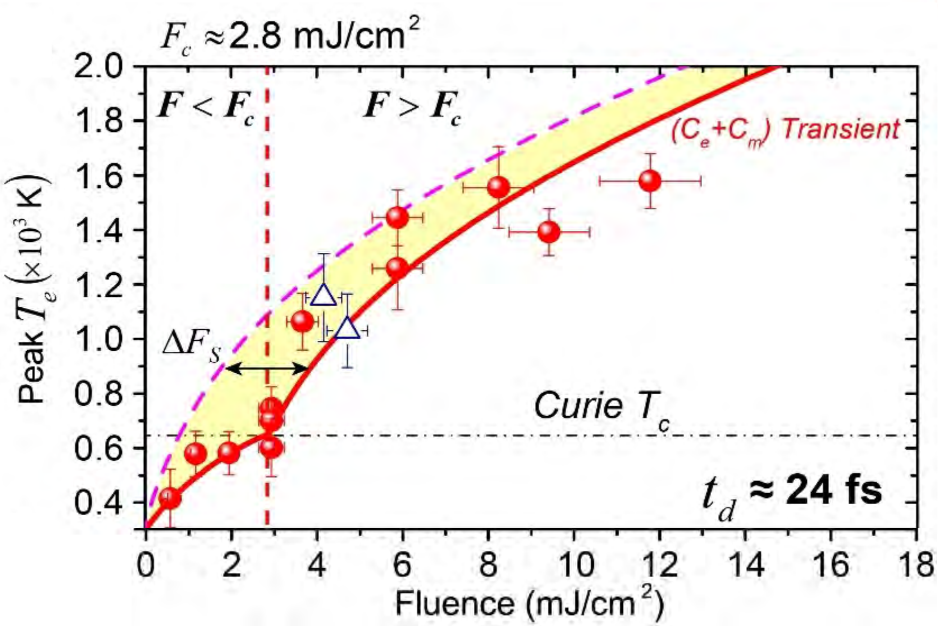
Half-metal Heusler Alloy: Co_2MnGe

Previous Work: Critical Behavior in Nickel

Probed spin dynamics in Ni using tabletop EUV high harmonics

- (ARPES) to probe exchange splitting, electron temperature
- (MOKE) to probe magnetic order, reflectivity



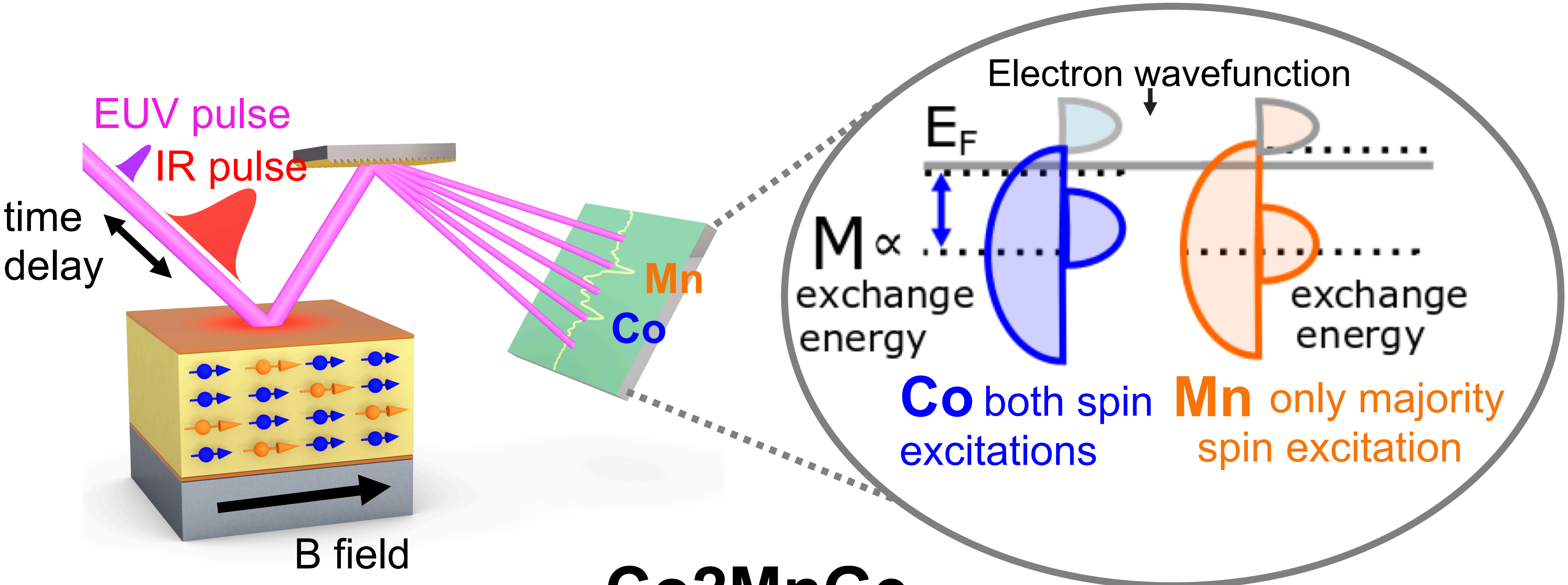


Observed very surprising spin dynamics

- Light excitation couples to spin system within 20fs after the pump pulse (new timescale for field) (ARPES)
- Initial super-excited state dictates future dynamics of the material for the full range of pump fluences (MOKE)

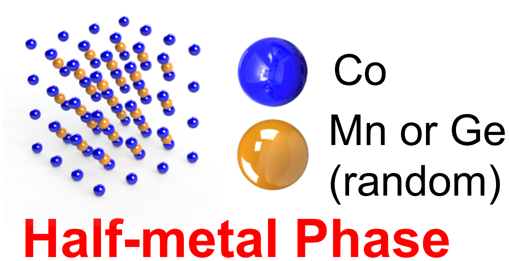
Tengdin, et. al. Sci. Adv. 2018;4 : eaap9744

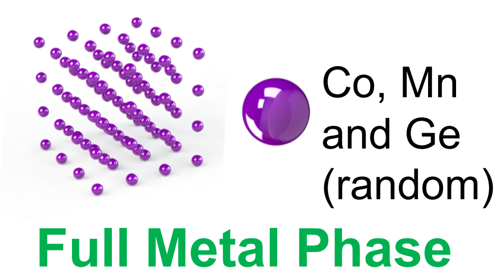
Light induced spin transfer: Half metallic System



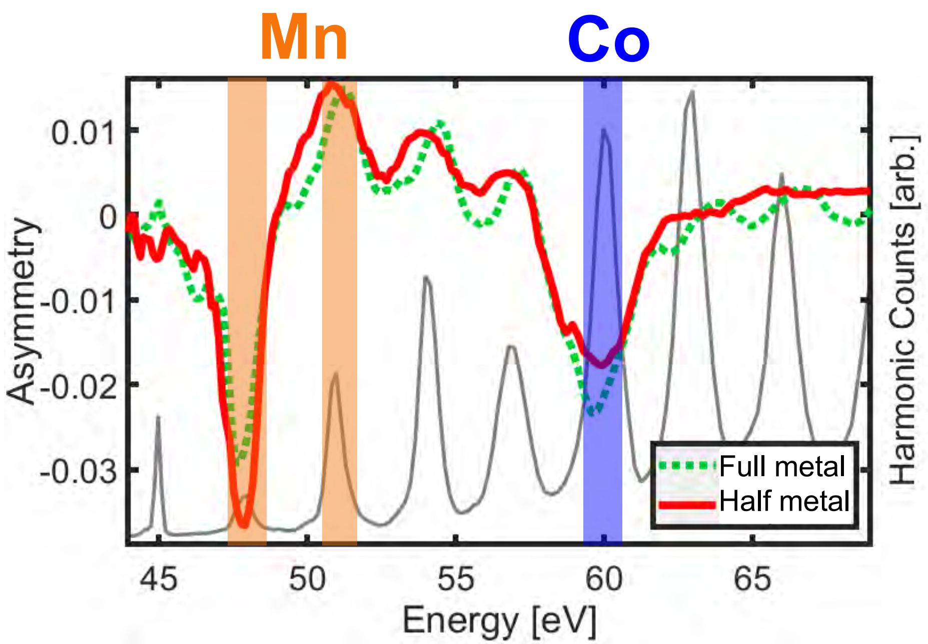
Co2MnGe

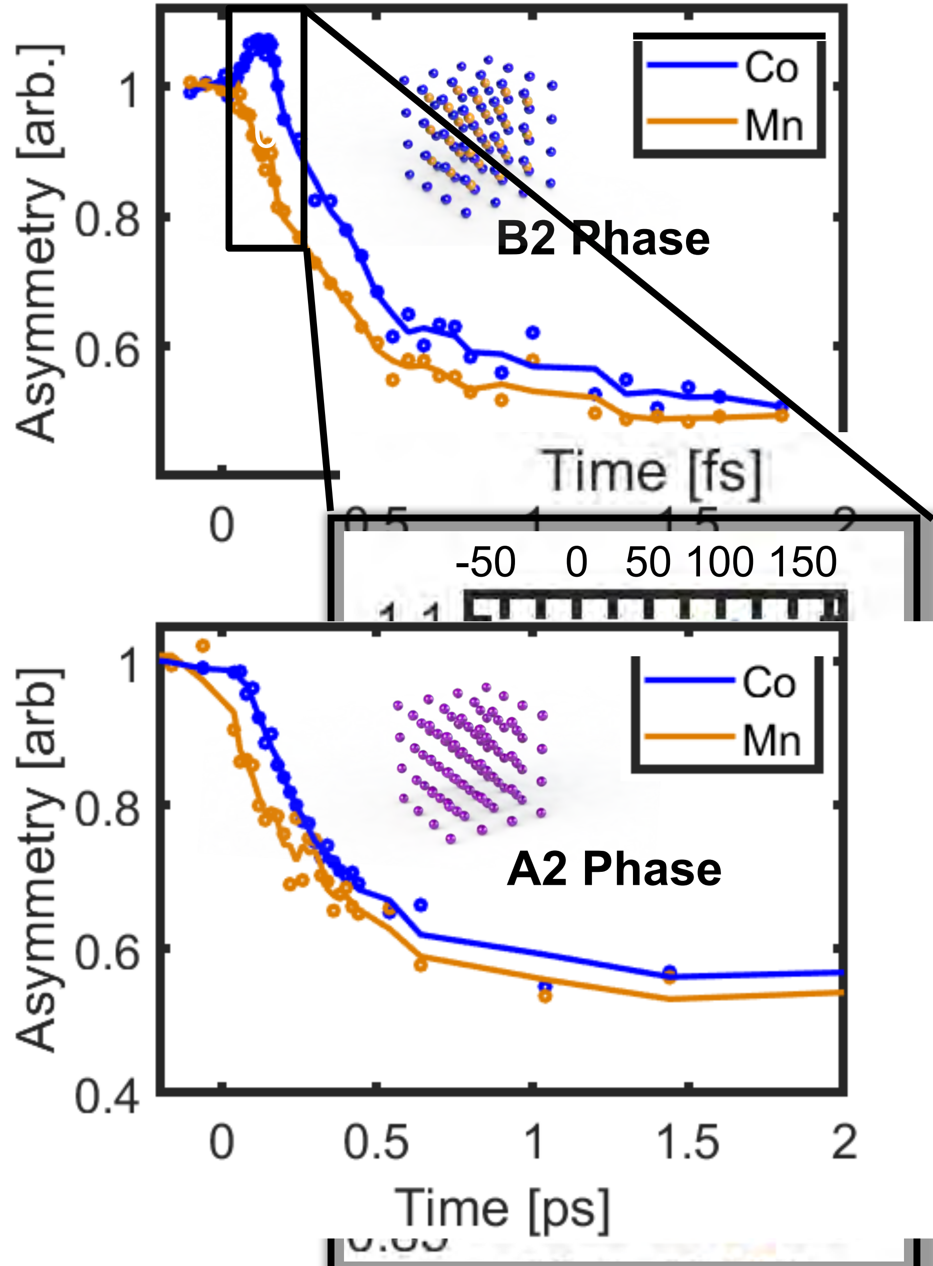
- Half metallic: one spin state conducting, the other insulating
- Half-metal gap in Mn is bigger than in Co
- Expect to see slower dynamics due to reduced scattering processes





Ultrafast transfer of magnetization between elemental sublattices

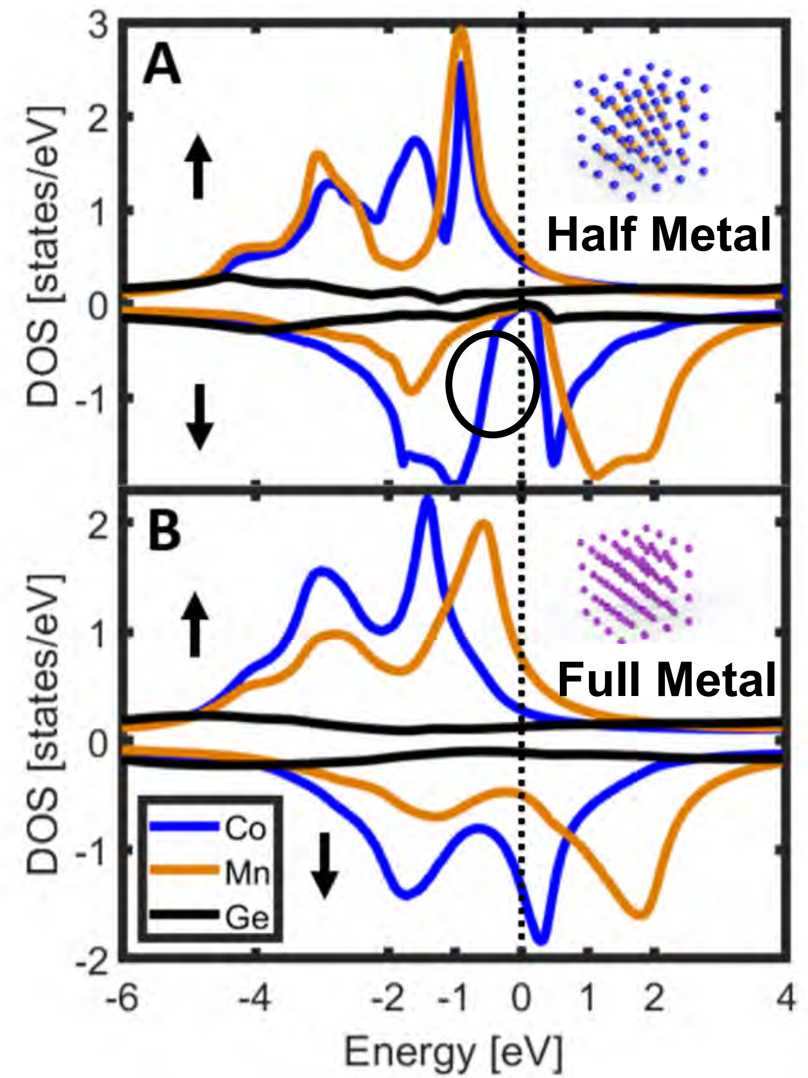




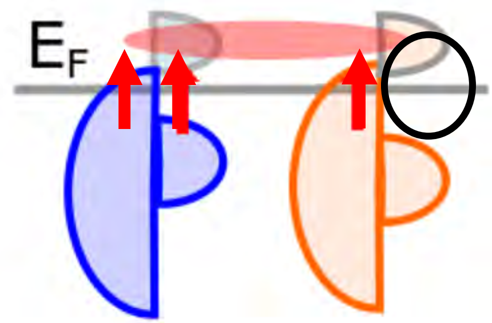
- Element specific measurements reveal transfer of magnetization between elemental sublattices ***within duration of pump pulse***
- Enhancement only present in half metal
- **Purely optically induced**

Tengdin, et. al.Sci. Adv. In press. (2019)

Element resolved density of states predicts mechanism for all-optical spin transfer



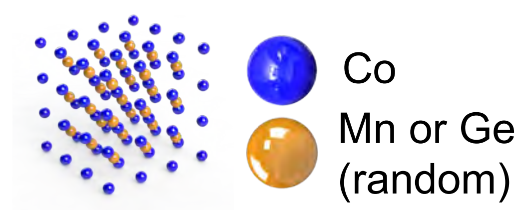
- **Full metal phase:** both spin states of both elements excited
- **Half metal phase:** <u>Only minority carriers excited in Mn</u> while both carriers are excited in Co



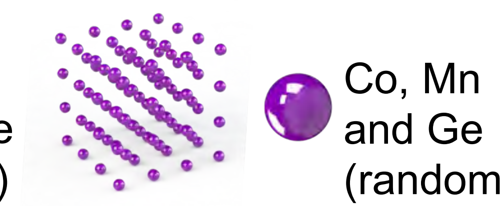
Co – minority spin excitation

Mn – majority spin excitation

Half-metal Phase

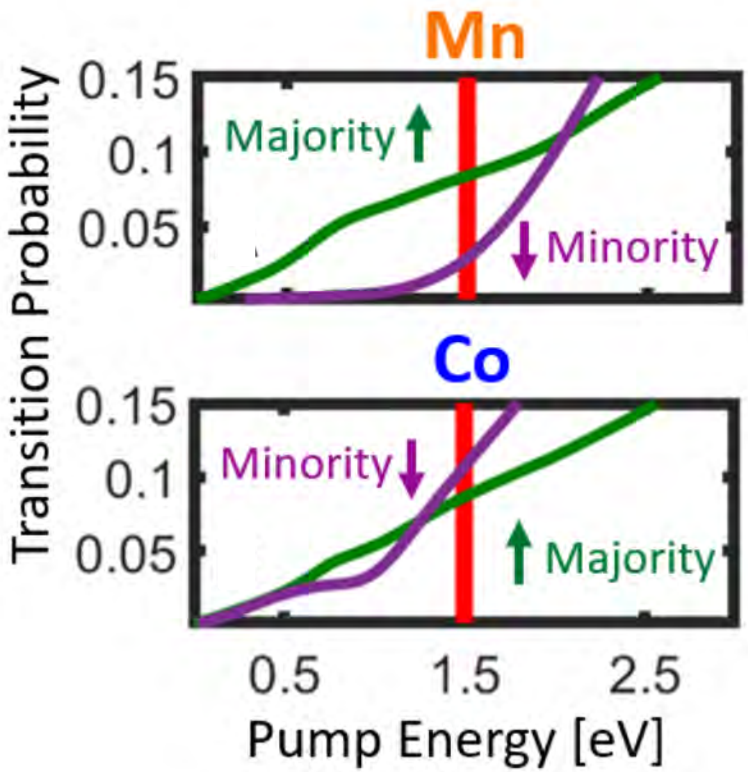


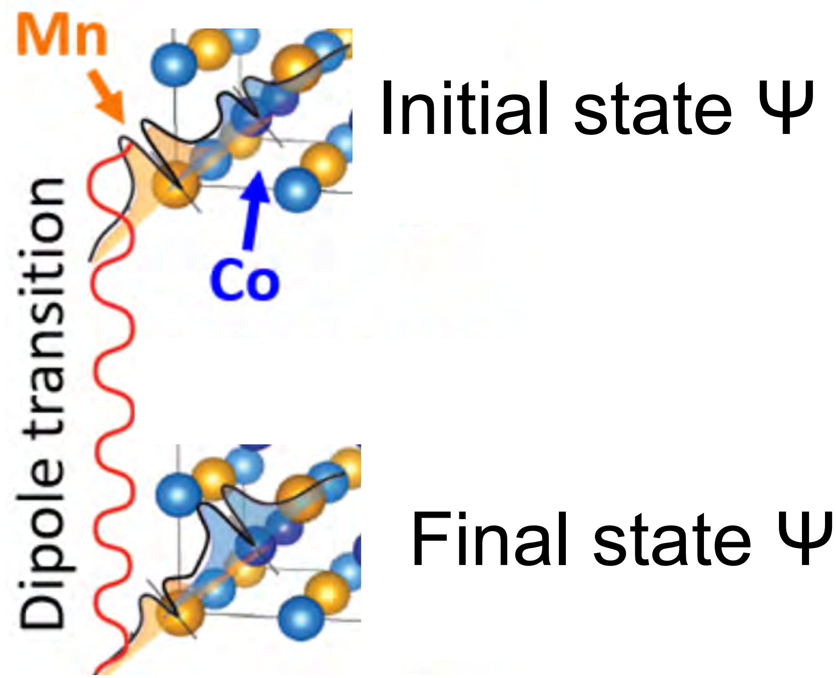
Full Metal Phase



Tengdin, P. et. al.Sci. Adv. In press. (2019)

Microscopic process for direct optical transfer of spin



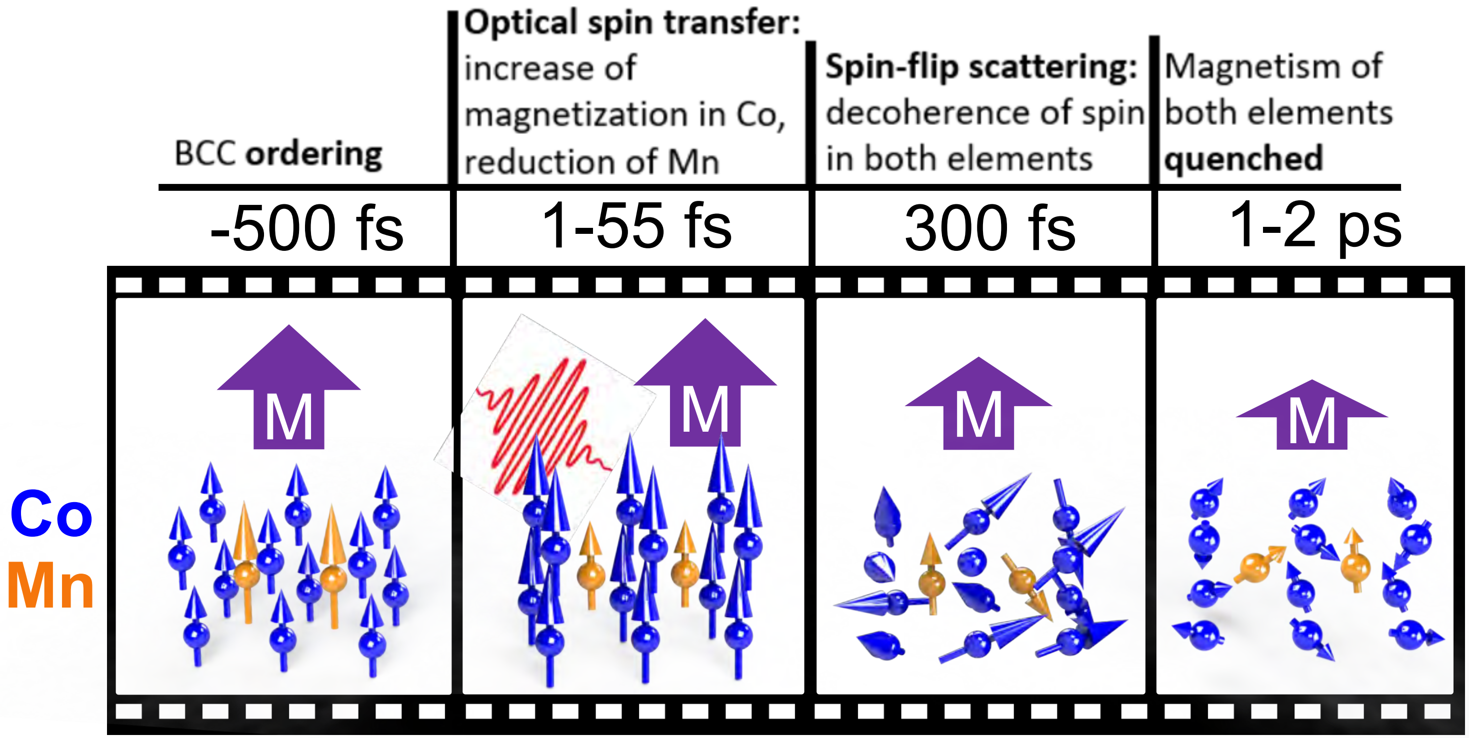


<u>Microscopic Process</u>

1. Preferential absorption of a single spin state in the valence band of Mn (wavefunction has primarily Mn character, electrons more localized)
2. Direct optical transfer via the conduction band (wavefunction has primarily Co character, electrons more itinerant)

Tengdin, P. et. al. Sci. Adv. In press. (2019)

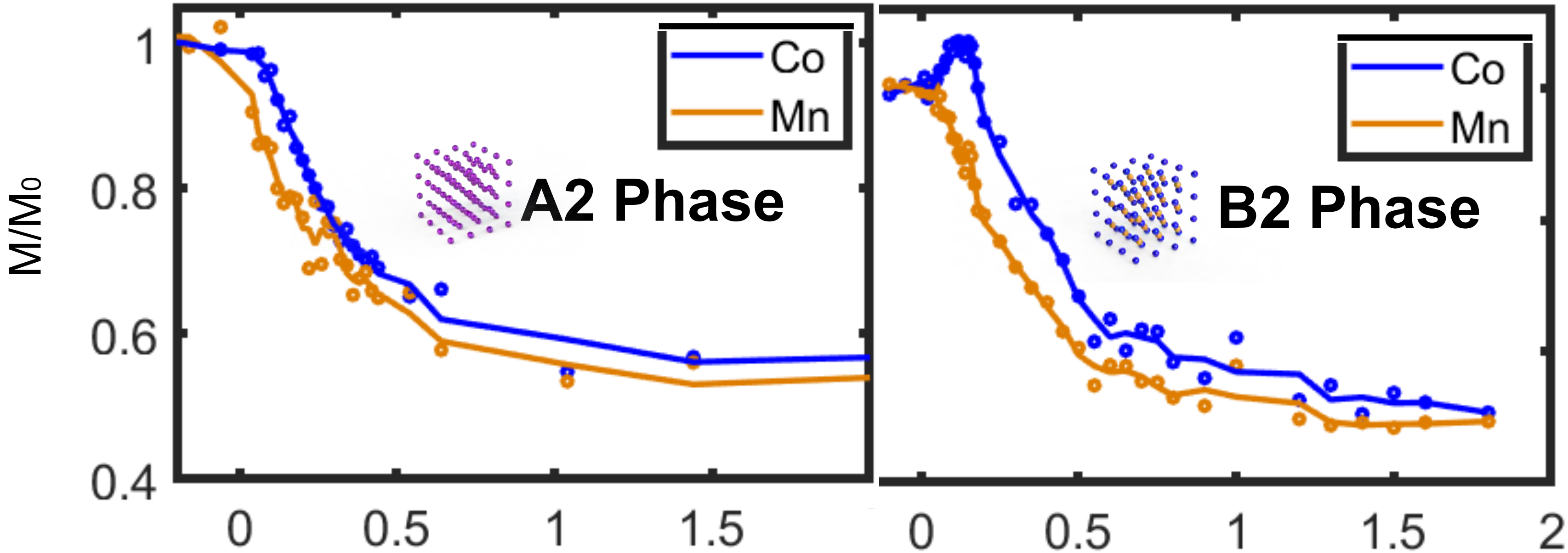
Optical spin transfer between elemental sublattices in half-metallic Co2MnGe on ultrafast 1-55 fs timescales

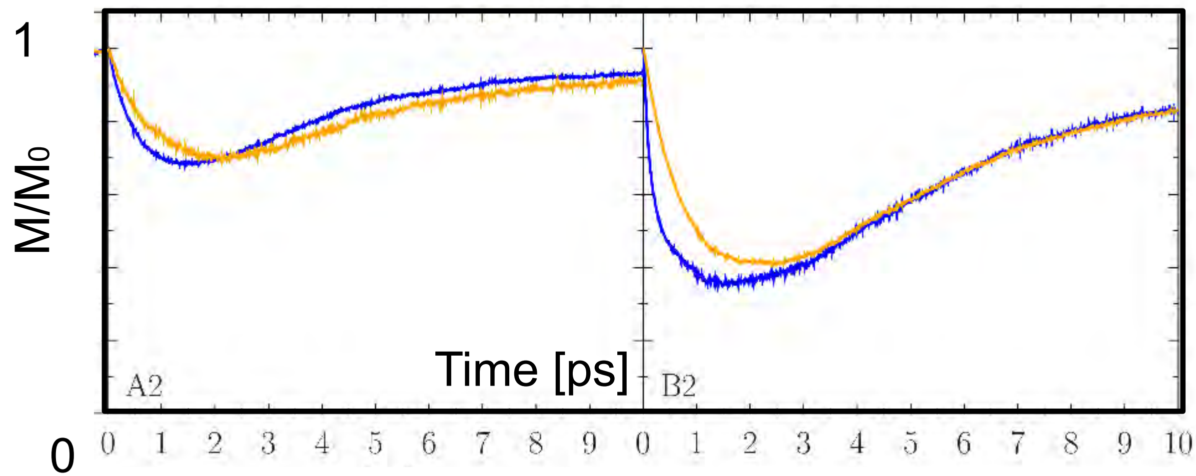


- **Transient enhancement of Co magnetization *within the duration of the pump pulse***
  - Multiple measurements confirm existence of the enhancement occurs only during pump pulse duration
  - **Enhancement possible *only for the half-metallic phase***
  - Direct measurement of nearly-instantaneous spin manipulation

Unlike previous ultrafast measurements: behavior is not captured by LLG modelling!

Experiments

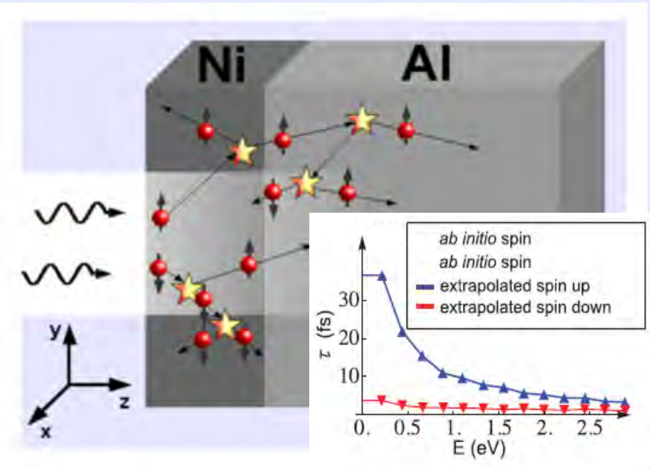




Atomic LLG

Informing and changing theory: *much faster spin manipulation, now we see what is happening*

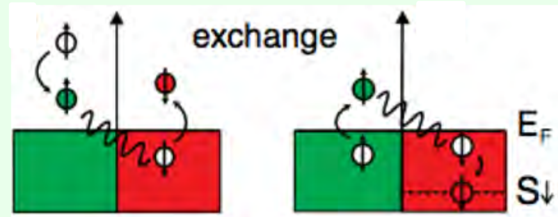
≈5-40 fs



Superdiffusive spin transport

Theory: PRL **105**, 027203 (2010)
Our work: PRL **110**, 197201 (2013)

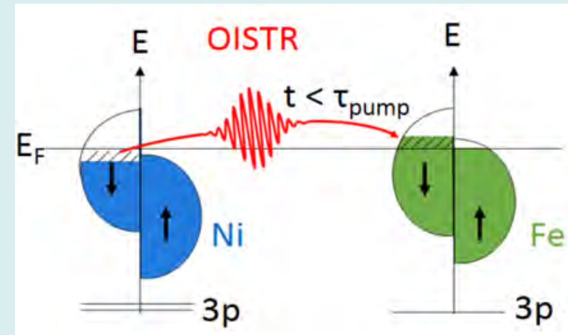
<20 fs



Spin flip scattering

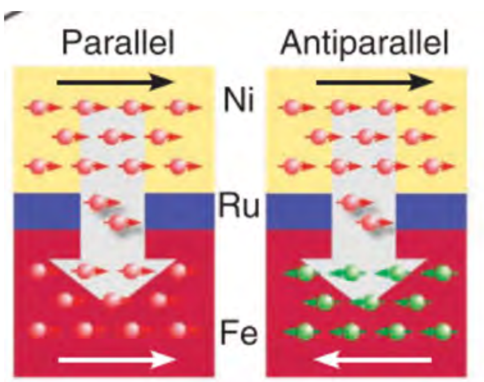
Theory: Nature Materials **9**, 259 (2010)
Our work: PRL **121**, 077204 (2018)
Tengdin, P. et al. Sci. Adv. **4**, *9744 (2018)*

<few-fs

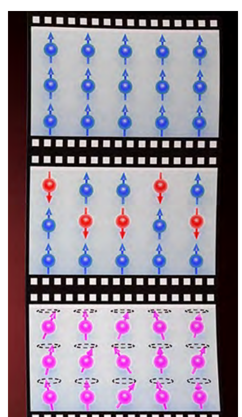


Ultrafast light-induced spin transfer

Theory: Nano Lett. **18**, 1842 (2018)
Our work: Tengdin et al., in press (2019)
Hofherr et al., in press (2019)



FM/NM layers



ARPES/MOKE
Measures:
FM Nickel

Co2MnGe Alloy

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DOE X-ray Scattering Program

LLG Formalism

We consider classical atomic magnetic moments $\{m_i\} = m_i\{e_i\}$ at site i. The dynamics of$\{m_i\}$ is governed by the atomistic Landau-Lifshitz-Gilbert (aLLG) equation (Antropov)

$$\frac{\partial m_i}{\partial t} = m\times\left(-\gamma(B_i + b_i\frac{\alpha}{m}\frac{\partial m_i}{\partial t}\right)$$

Where γ is the gyromagnetic ratio and $B_i = \frac{\partial H}{\partial m_i}$ is the effective precession field related to the spin-Hamiltonian, employing a Heisenberg model where -

$$H = -\sum_{i,j} J_{i,j} m_i \cdot m_j - \mu_B B \sum_i m_i$$

Here, the magnetic moments at site *i* and *j* are coupled by the exchange parameter J_{ij}. The values of the Heisenberg exchange parameters were obtained from first principles electronic structure theory. In the expression above, B is the external magnetic field. From the fluctuation-dissipation theorem, thermal fluctuations enter by a stochastic field, b_i, that fulfills white noise properties, such as $\langle b_i \rangle = 0$ and $\langle b_i^\mu(t) b_k^\nu(t') \rangle = D\delta_{ij}\delta_{\mu\nu}\delta(t-t')$, with the fluctuation amplitude $D = 2\alpha k_b T/\gamma m$. The dissipation part enters the equation of motion via a viscous damping part scaled by the Gilbert damping constant α.

V. Antropov et al. Phys. Rev. B **54**, 1019 (1996).