Demagnetization Dynamics in Ferromagnets and Spin-polarized Transport in Metals

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2012-10-18

Condensed-Matter Physics Seminar

Thanks

Theory

Yao-Hui Zhu

Sven Essert

Michael Krauss

Steffen Kaltenborn

Bärbel Rethfeld & Benedikt Müller

Experiment

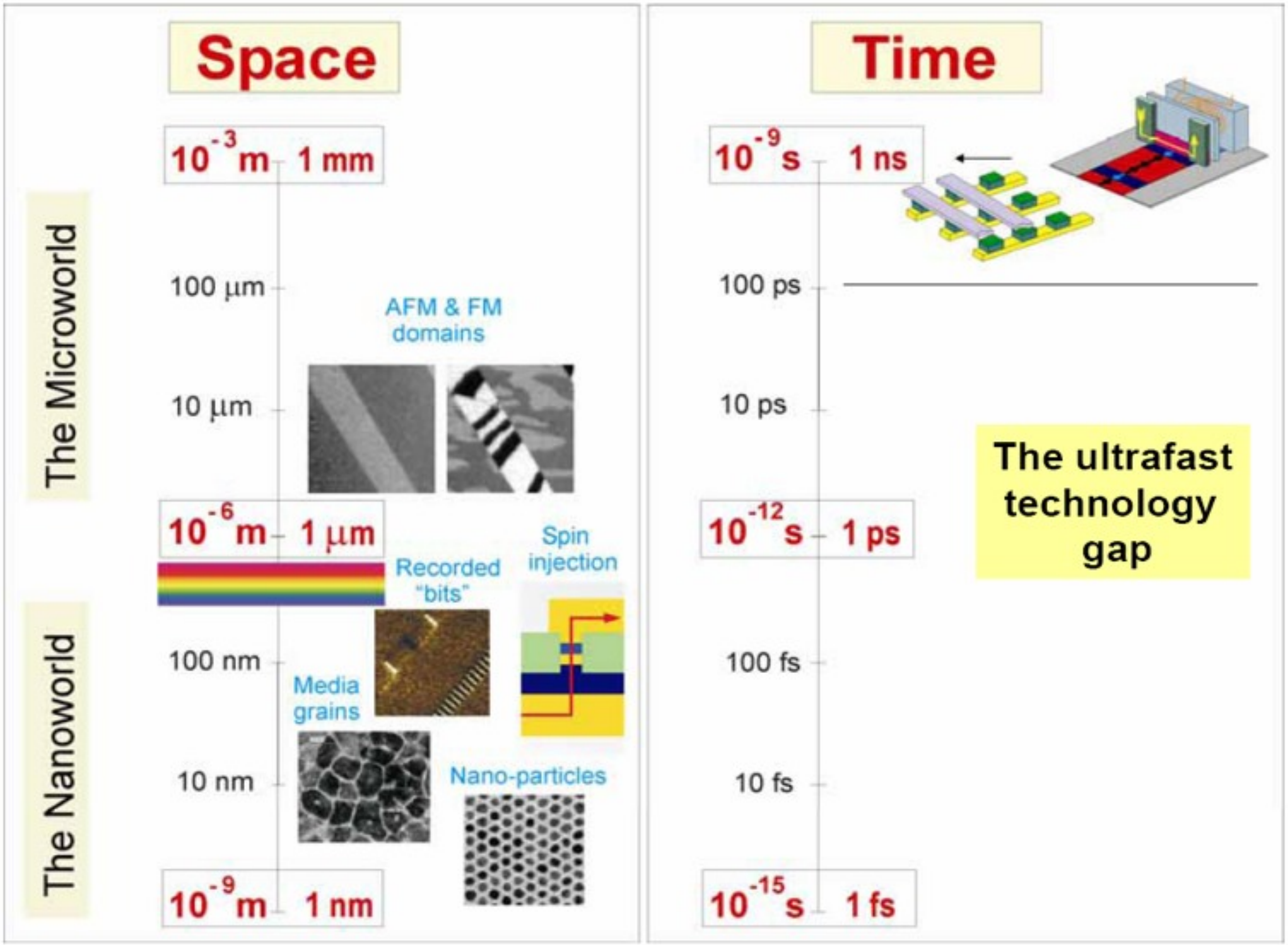
group of Martin Aeschlimann (Daniel Steil, Sabine Alebrand, and Mirko Cinchetti)

group of Burkard Hillebrands (Frederik Fohr, Helmut Schultheiss, and A. Serga)

Outline

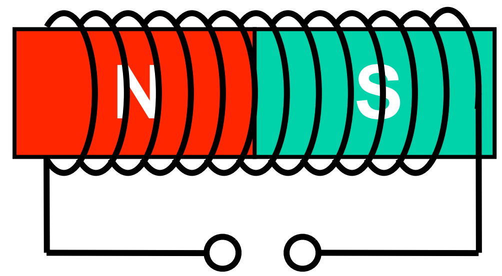
1. Ultrafast demagnetization in ferromagnets
2. Elliott-Yafet demagnetization due to electron-phonon scattering and optical excitation dynamics from ab-initio calculations
3. Limits to scattering in a fixed bandstructure
4. Extensions of the Elliott-Yafet approach
5. Wave-diffusion theory of spin and charge transport in metals
6. Application to noncollinear spin currents
7. Application to optically excited spin-polarized currents

How Small & Fast Can Magnetism/Spintronics Get?



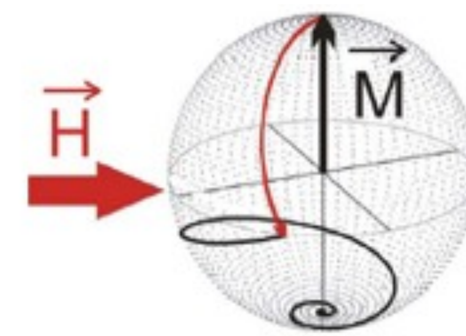
Magnetization Dynamics: Scenarios in Ferromagnets

- Conventional switching: magnetic field (pulses) → Domain-wall propagation (>1ns)

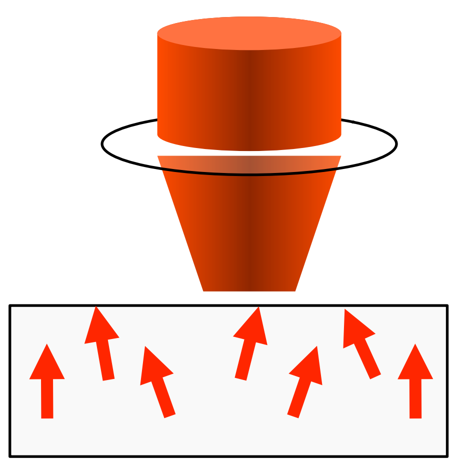


slow!

- Coherent rotation → "precessional switching" (>10ps)



- Optically induced magnetization dynamics



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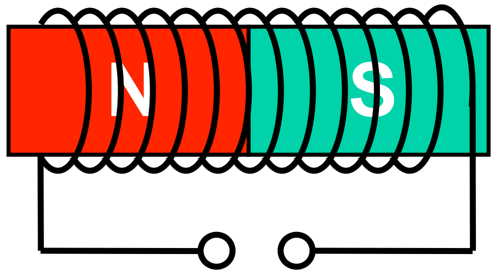

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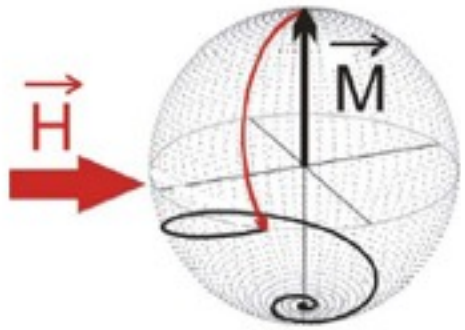

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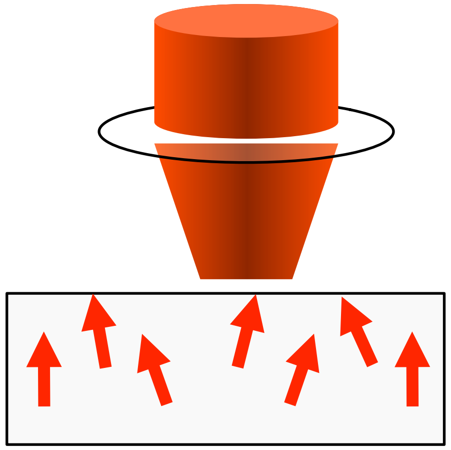

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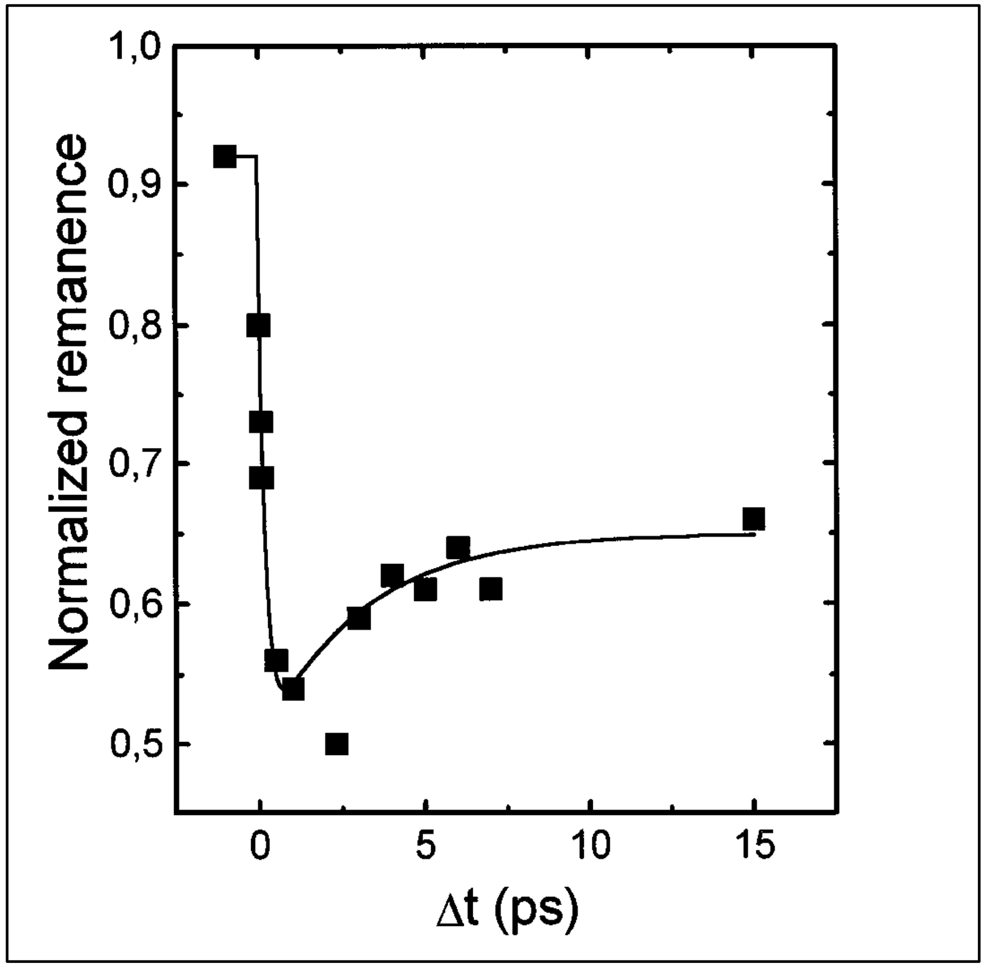
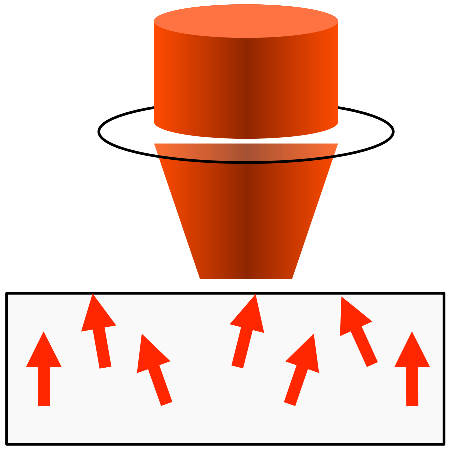
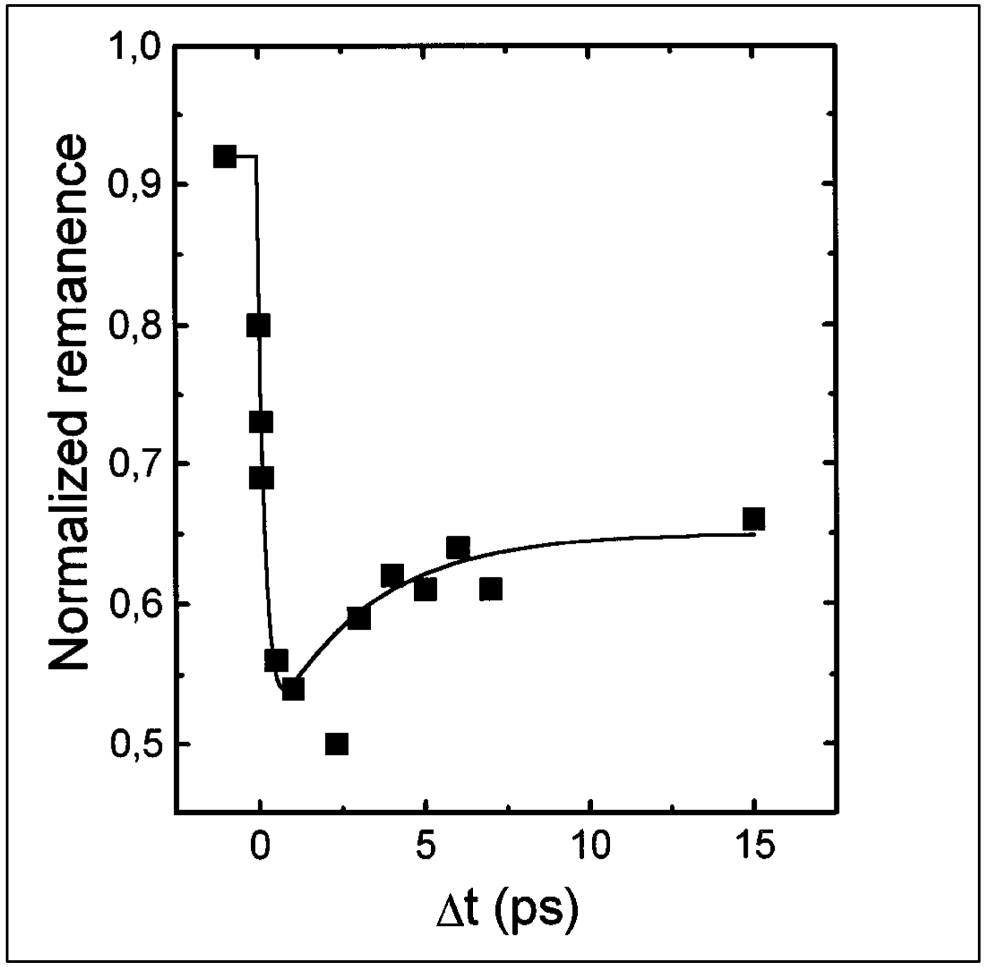
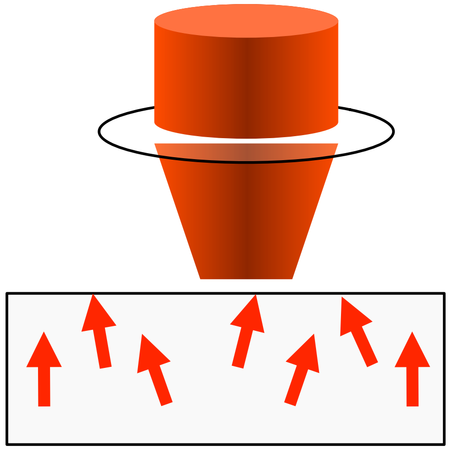




Beaurepaire, Merle, Daunois, Bigot, Phys. Rev. Lett. **76**, 4250 (1996)

Magnetization Dynamics: Scenarios in Ferromagnets





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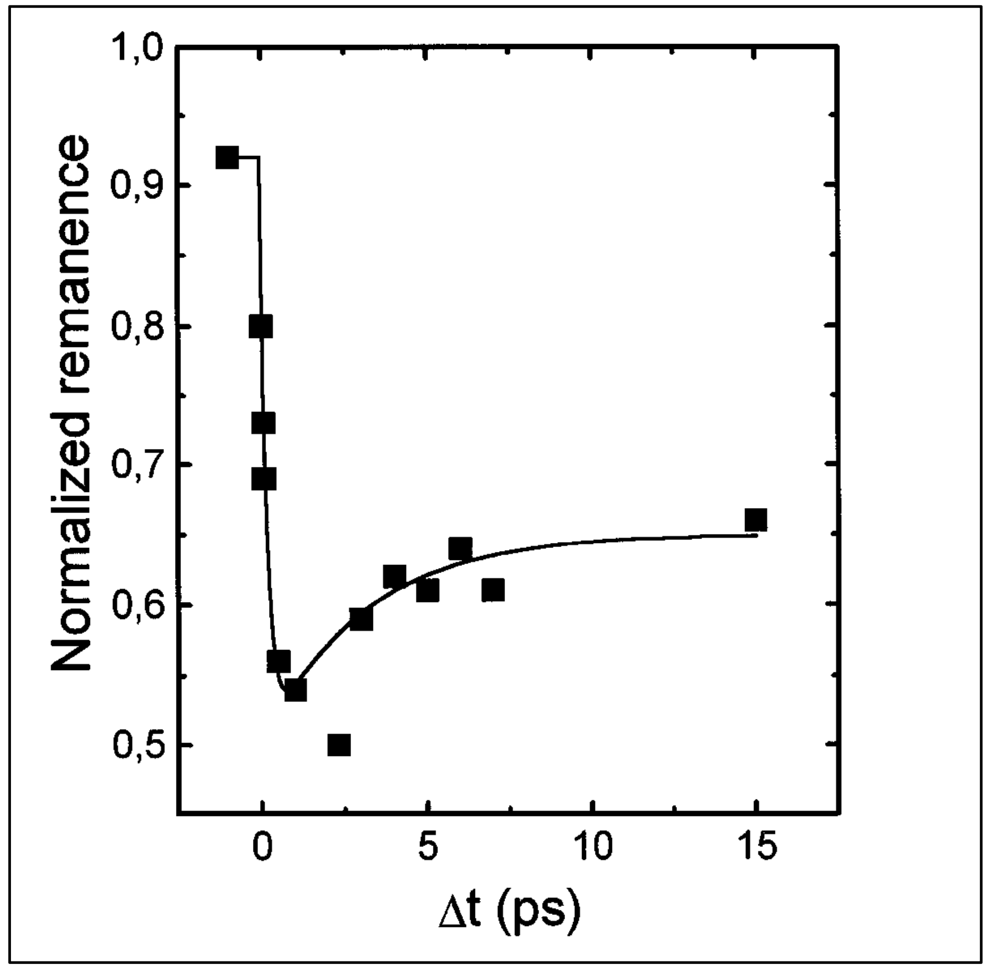
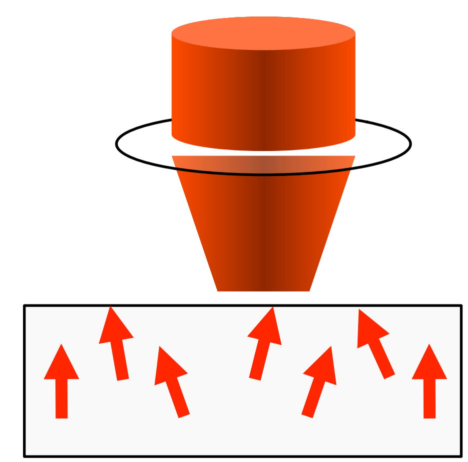




Beaurepaire, Merle, Daunois, Bigot, Phys. Rev. Lett. **76**, 4250 (1996)

Ultrafast Optical Swiching Demonstrated

Questions/Follow-ups:

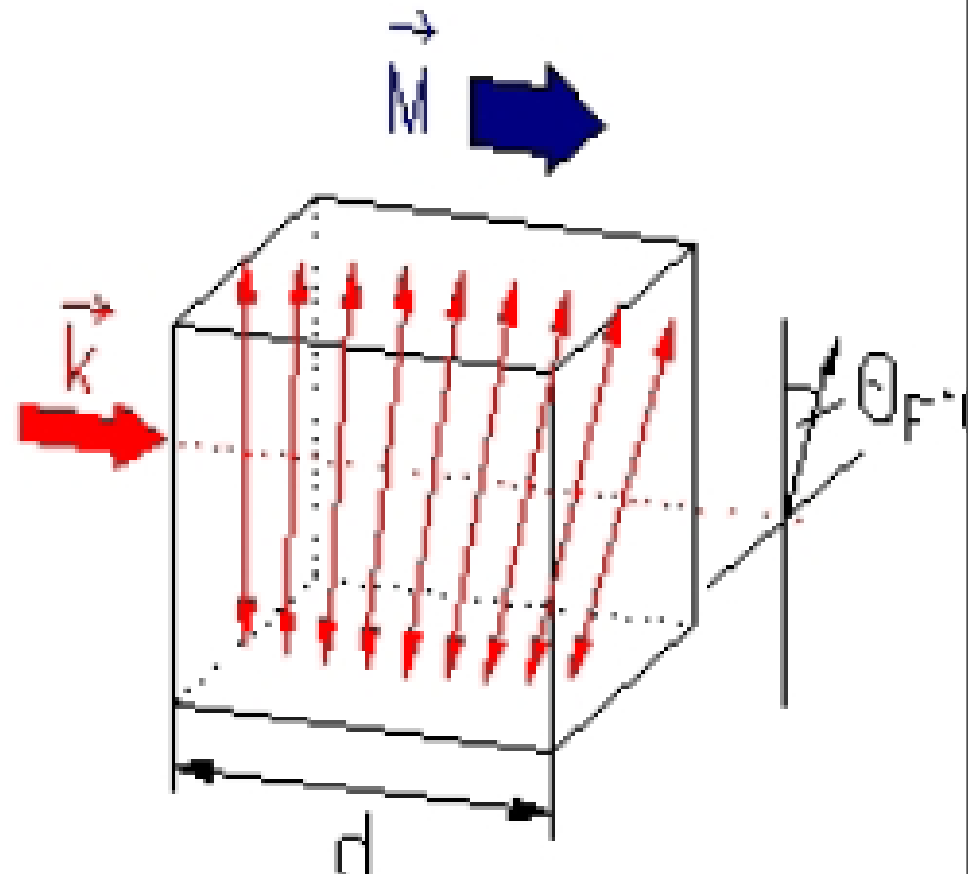
- Is it real?
- Clarify physical processes involved (angular momentum balance?)
- Determine timescales (ultimate switching speed)
- Invent new scenarios (employing optical fields)
- Look at new materials

Magneto-Optical Kerr Effect: MOKE

- Magneto-optical effects: Magnetization *M* influences reflected (*Kerr* effect) and transmitted light (*Faraday* effect) and
- MOKE: Light polarization angle rotated by $\Theta_F(M)$
- Faraday geometry: Intensity changes = magnetic contrast

What is measured on ultrashort time scales? Only reflectivity?

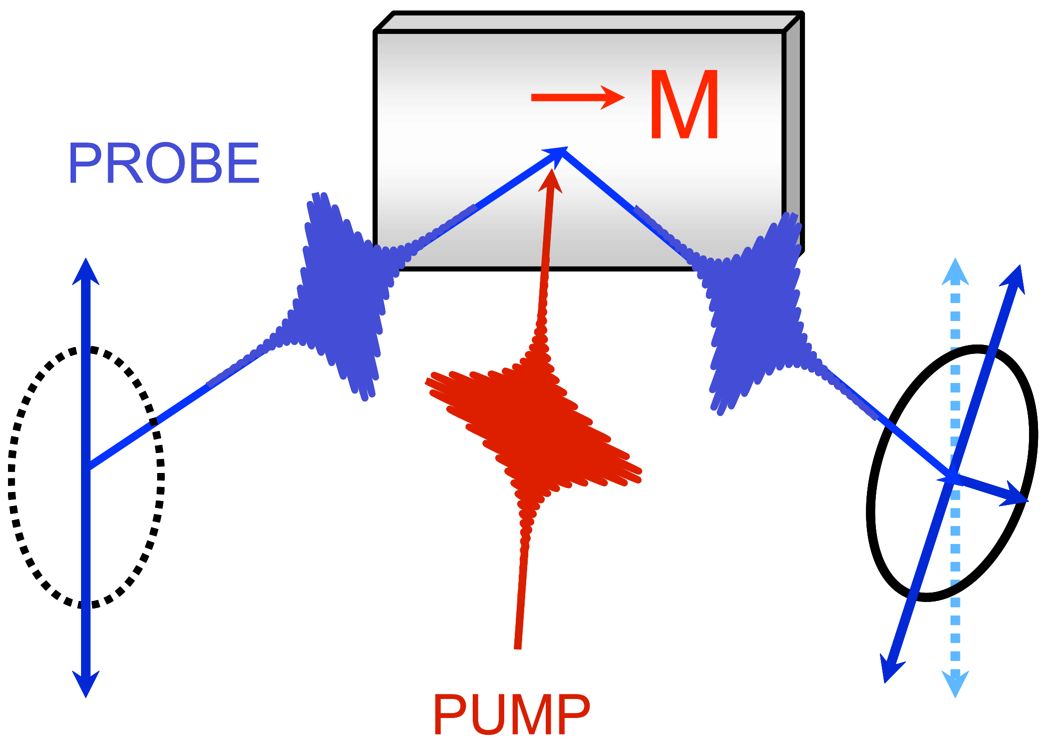
- Light-matter interaction: electric dipole moments
- Magnetization: spin expectation value
- MOKE signal OK at experimentally relevant photon energies and pulse durations

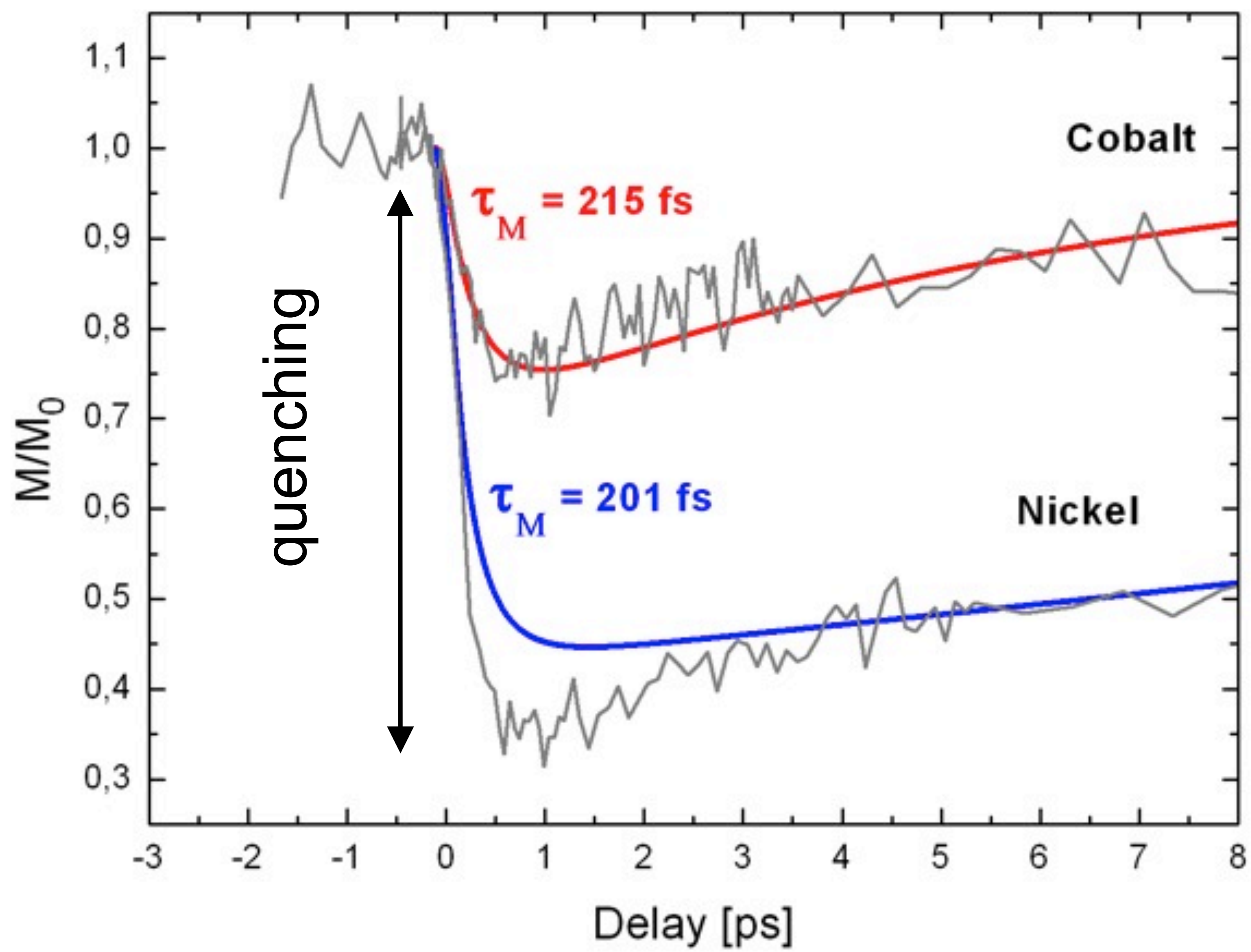


Zhang et al., Nature Physics **5**, 449 (2009) + comment
Carva et al., Nature Physics **7**, 665 (2011)

Ultrafast Demagnetization in Experiment

- Pump-Probe-Measurement of the Magneto-optical Kerr Effect (MOKE)
- Ultrafast magnetization quenching

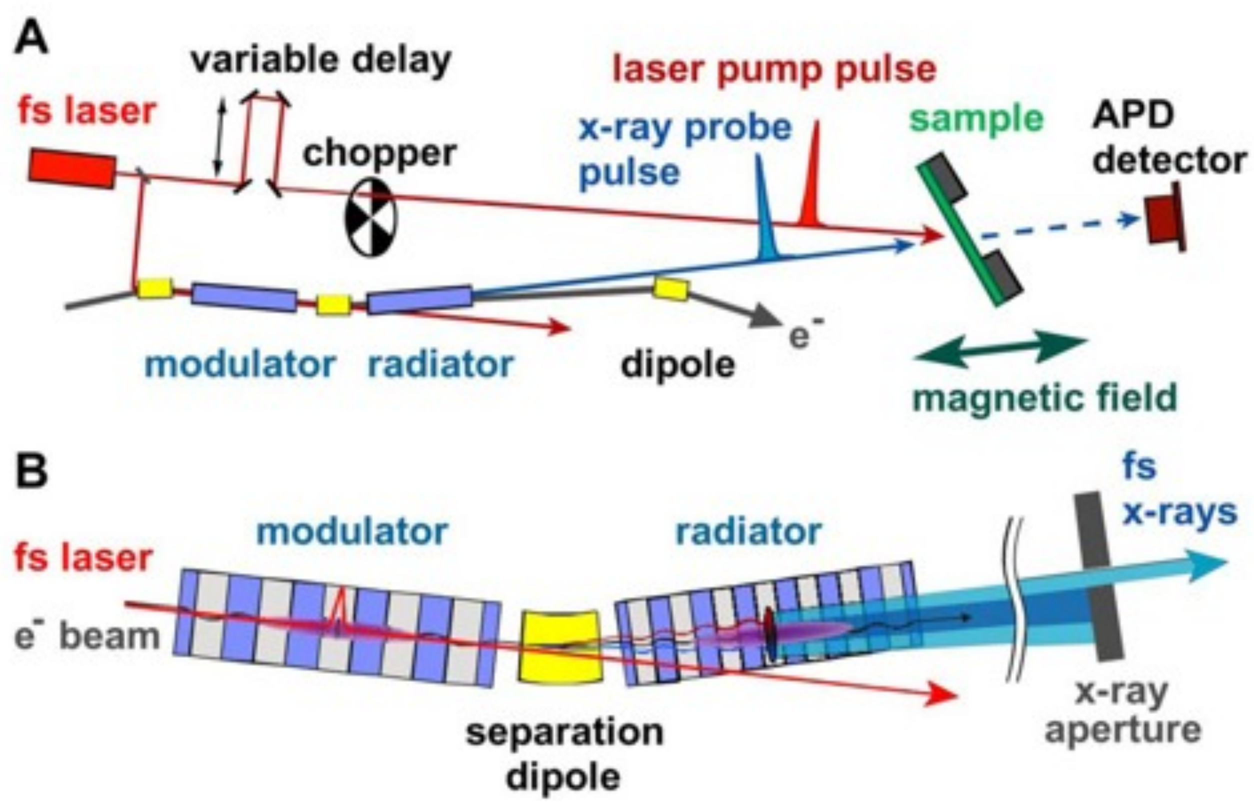


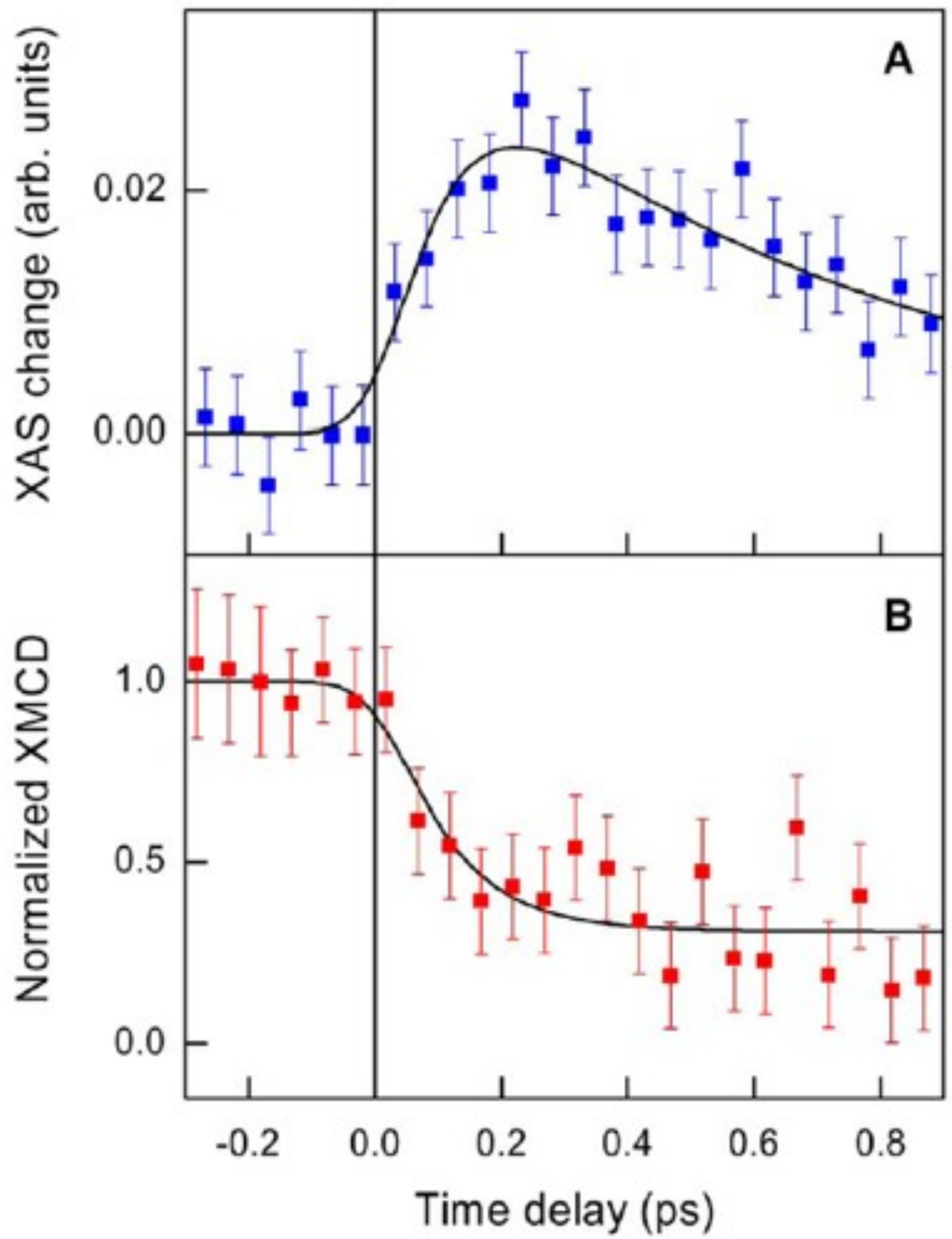


Beaurepaire, Merle, Daunois, Bigot, Phys. Rev. Lett. **76**, 4250 (1996)
M. Krauß et al., Phys. Rev. B **80**, 180407(R) (2009)

Ultrafast Demagnetization in Experiment

- X-ray magnetic circular dichroism (XMCD)

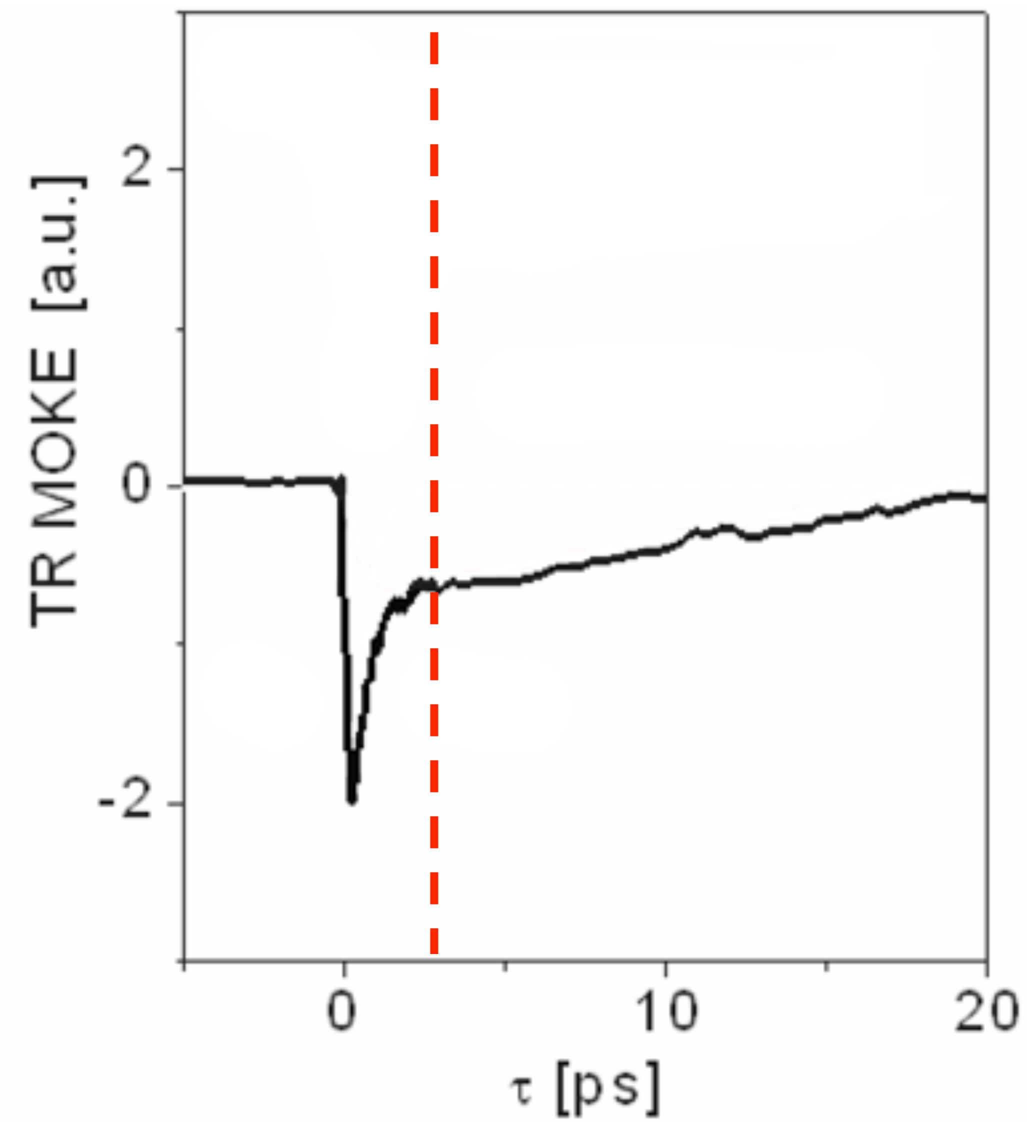


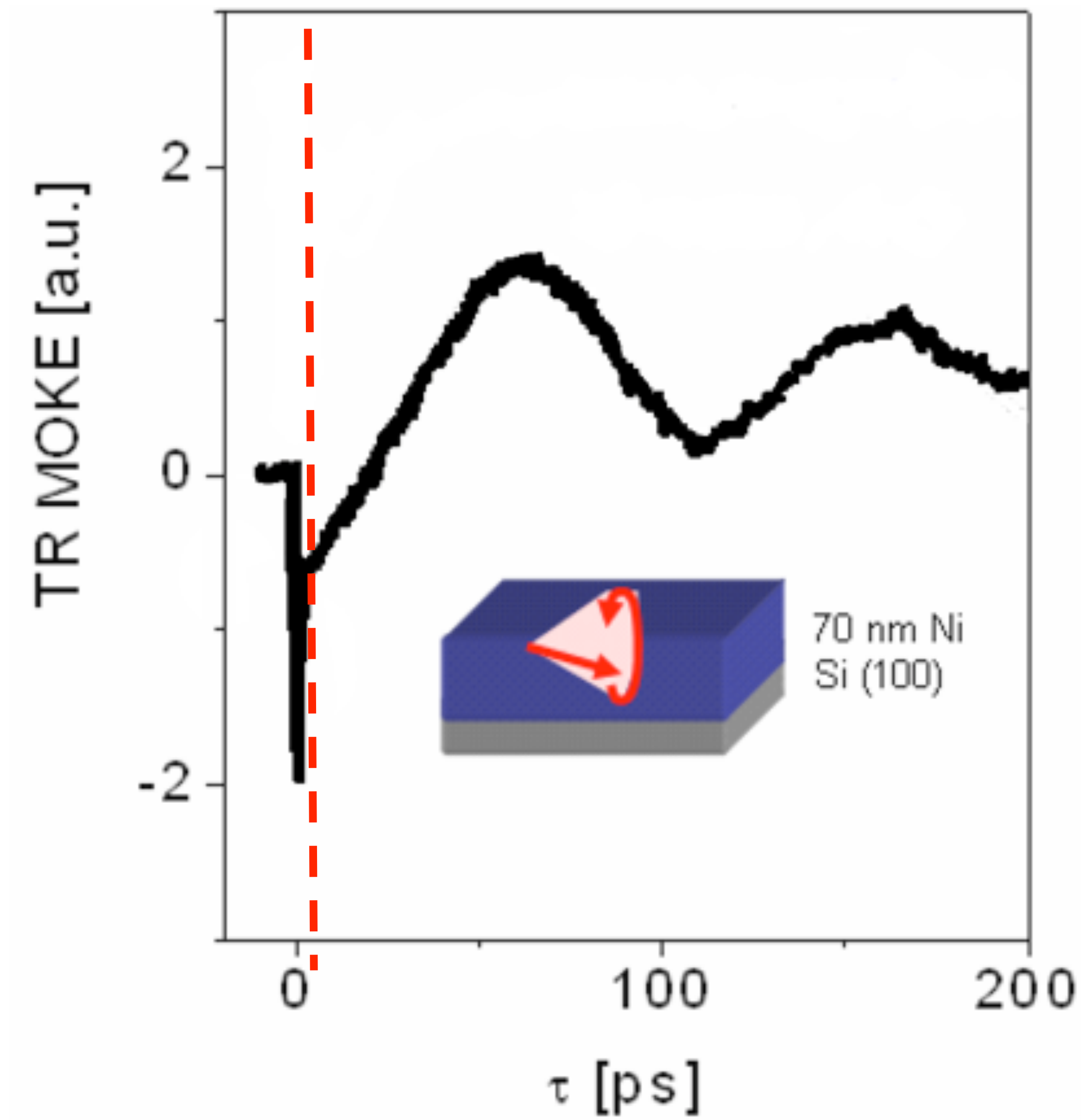


Stamm et al., Nature Materials **6**, 740 (2007)

Magnetization Dynamics on Different Time Scales

- Experimental TR-MOKE result on different time scales



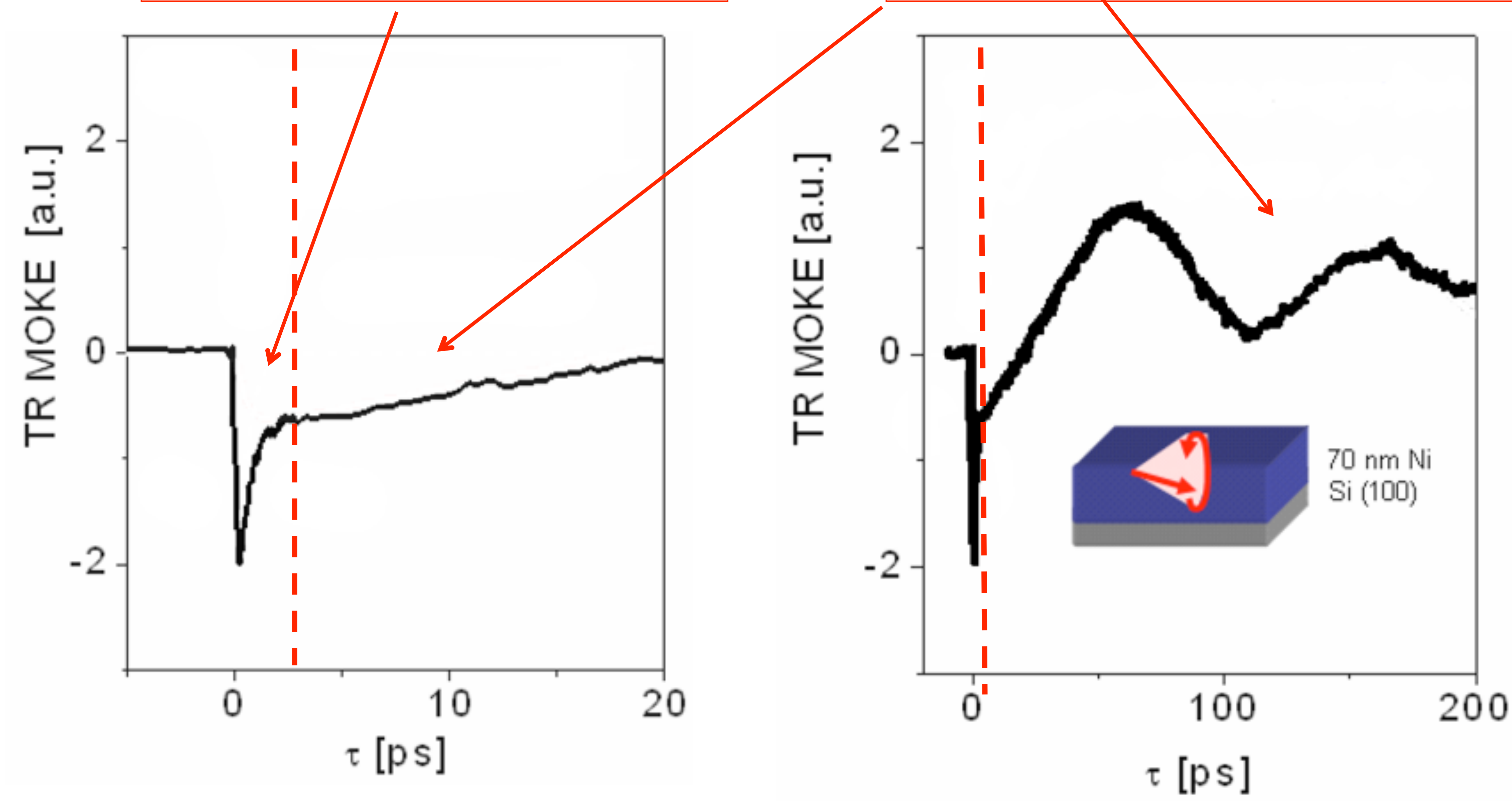


Djordjevic et al., phys. stat. sol. (c) 3, 1347 (2006)

Magnetization Dynamics on Different Time Scales

- non-equilibrium dynamics
- temperature not well defined

- quasi-equilibrium dynamics
- temperature $T=T(\mathbf{r},t)$



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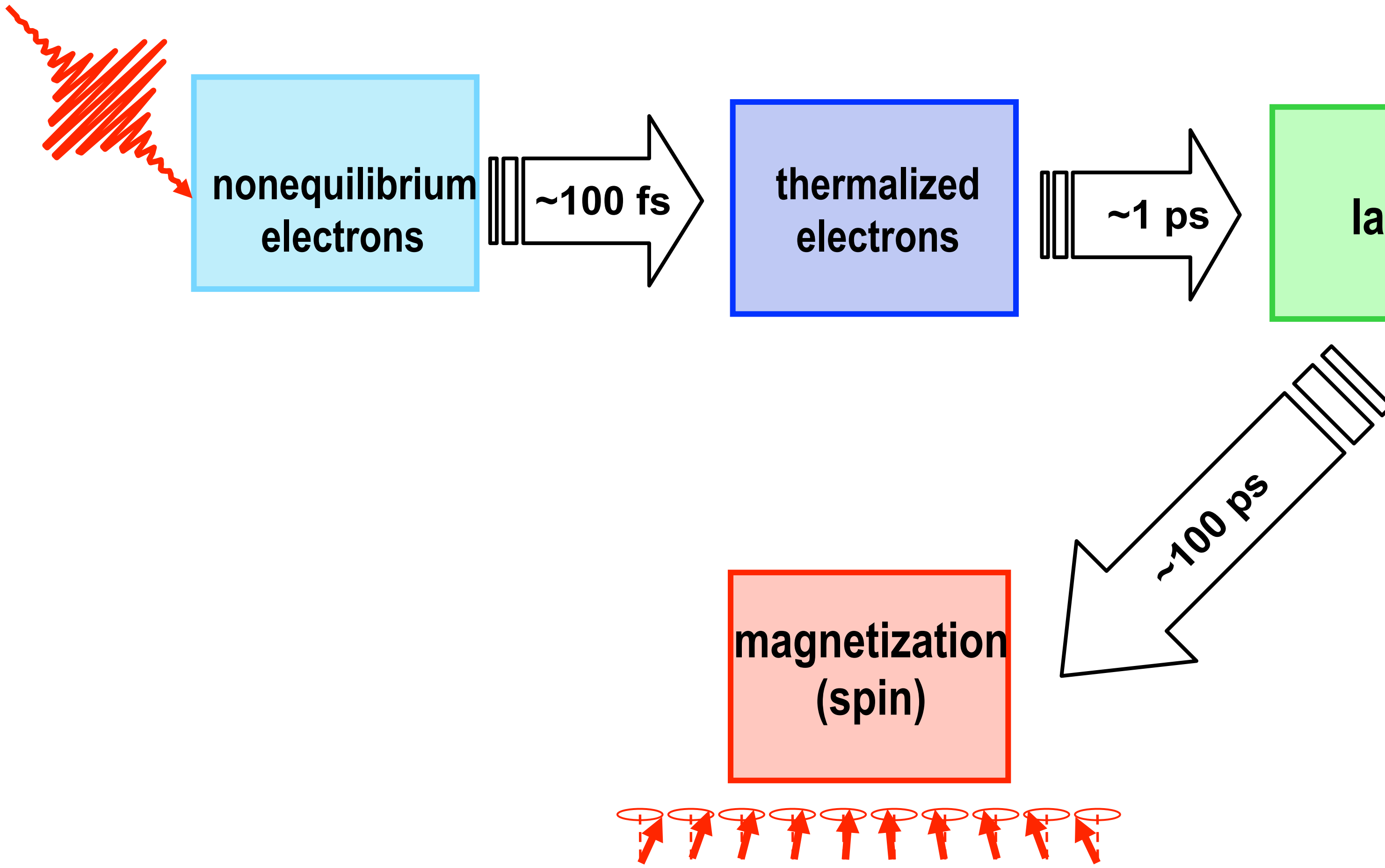
Time Scales of Magnetization Dynamics



- Coherent regime (~10 fs)
- Incoherent “thermalization” dynamics of nonequilibrium electrons (100 fs)
- Quasi-thermal regime: electron temperature, lattice temperature (1 ps)
- Spin-lattice equilibration (100 ps)

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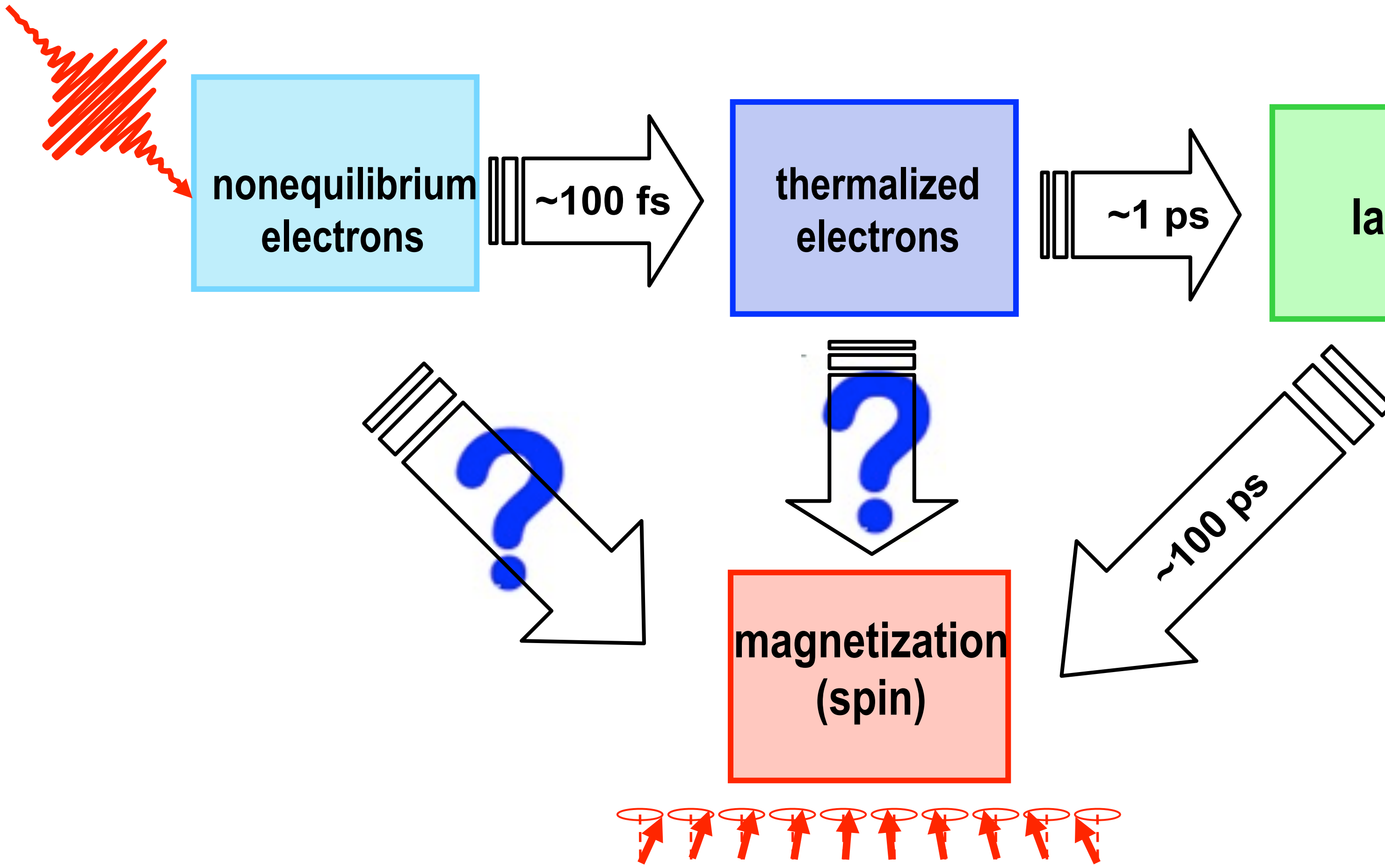

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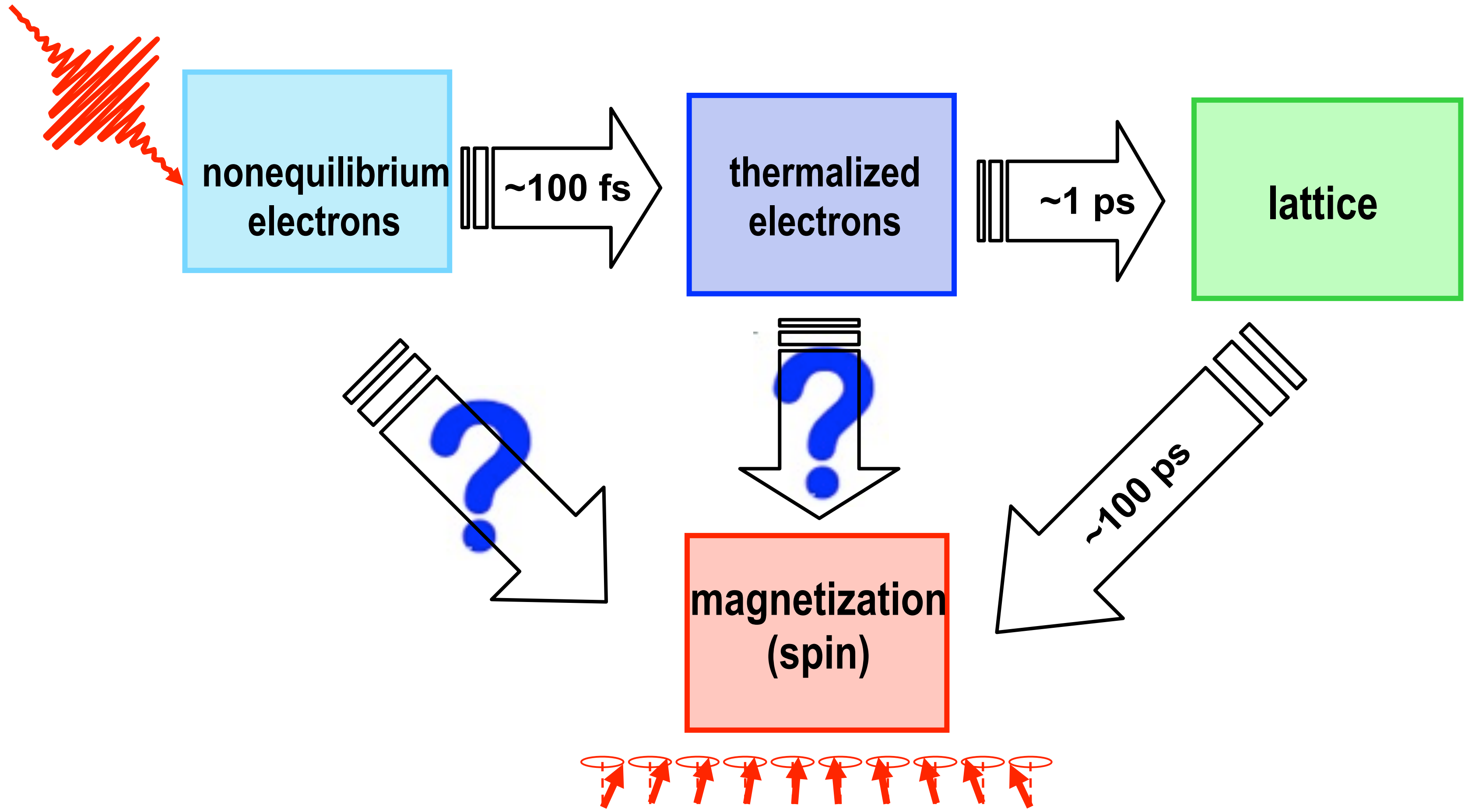

- Coherent regime (~10 fs)
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- Spin-lattice equilibration (100 ps)

- Ultrafast magnetization (spin) dynamics surprising!

Elliott (-Yafet) Mechanism for Depolarization

- Spin-orbit interaction: spin not a good quantum number

$$|\Psi_k\rangle = a_k|\uparrow\rangle + b_k|\downarrow\rangle$$

- Average spin of single particle states

$$|\langle\Psi|S_z|\Psi\rangle| \leq \frac{\hbar}{2}$$

- Spin diagonal scattering processes change average spin

$$|\Psi\rangle = a|\uparrow\rangle + b|\downarrow\rangle \longrightarrow |\Psi'\rangle = a'|\uparrow\rangle + b'|\downarrow\rangle$$

$$\langle\Psi|S_z|\Psi\rangle \neq \langle\Psi'|S_z|\Psi'\rangle$$

$$\boxed{\frac{d}{dt}\langle S_z\rangle \neq 0}$$

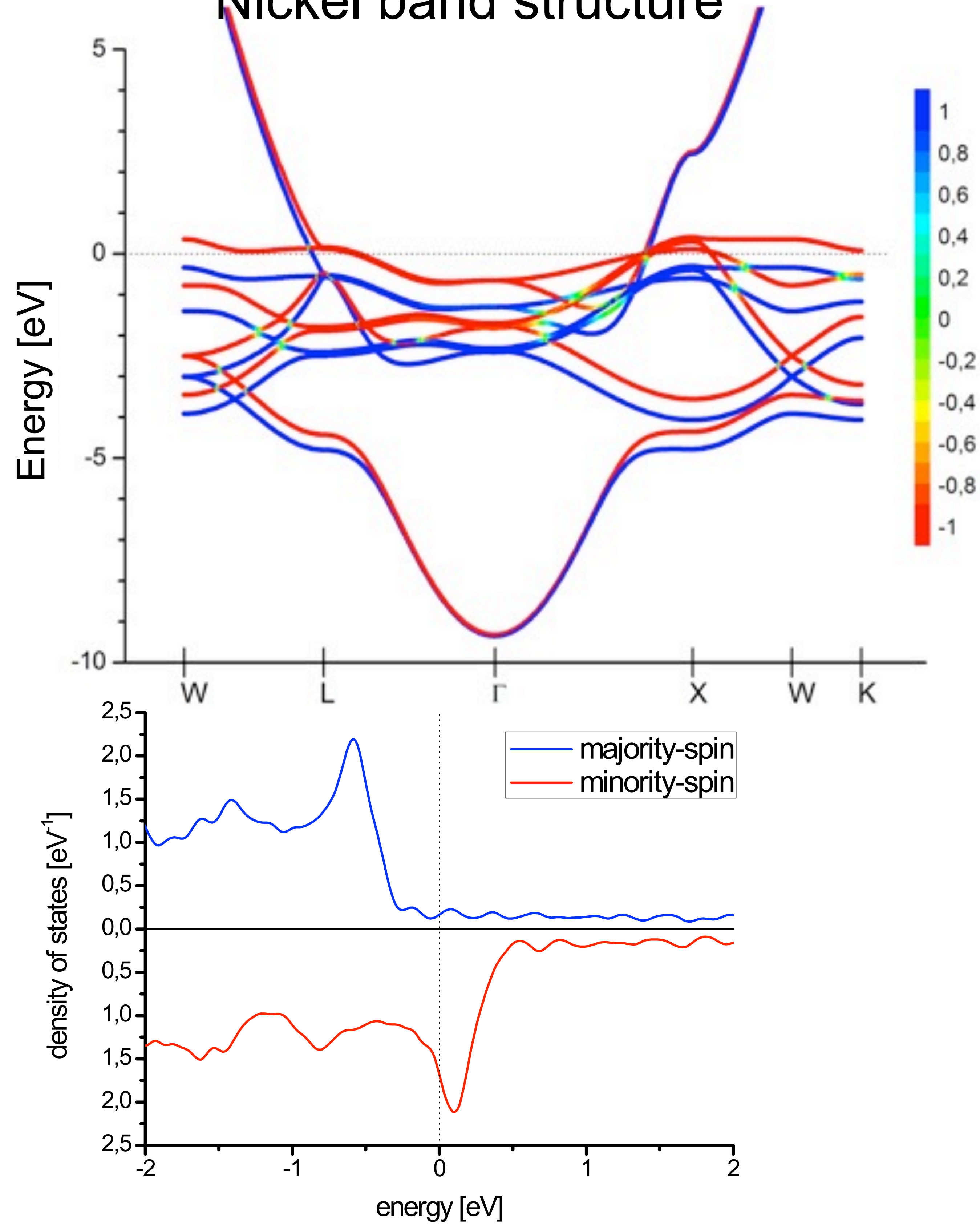
Transition Metals: Band Structure

- Spin mixing important for optical excitation and scattering
- Spin mixing anisotropic ("spin hot-spots")?

  Fabian & Das Sarma, Phys. Rev. Lett. **81**, 5624 (1998)

- Compute numbers for real experiments from a microscopic theory using ab-initio input (if possible)!

Nickel band structure



Other Approaches

- Coherent effects: Important for (few) localized levels with strong spin-orbit coupling

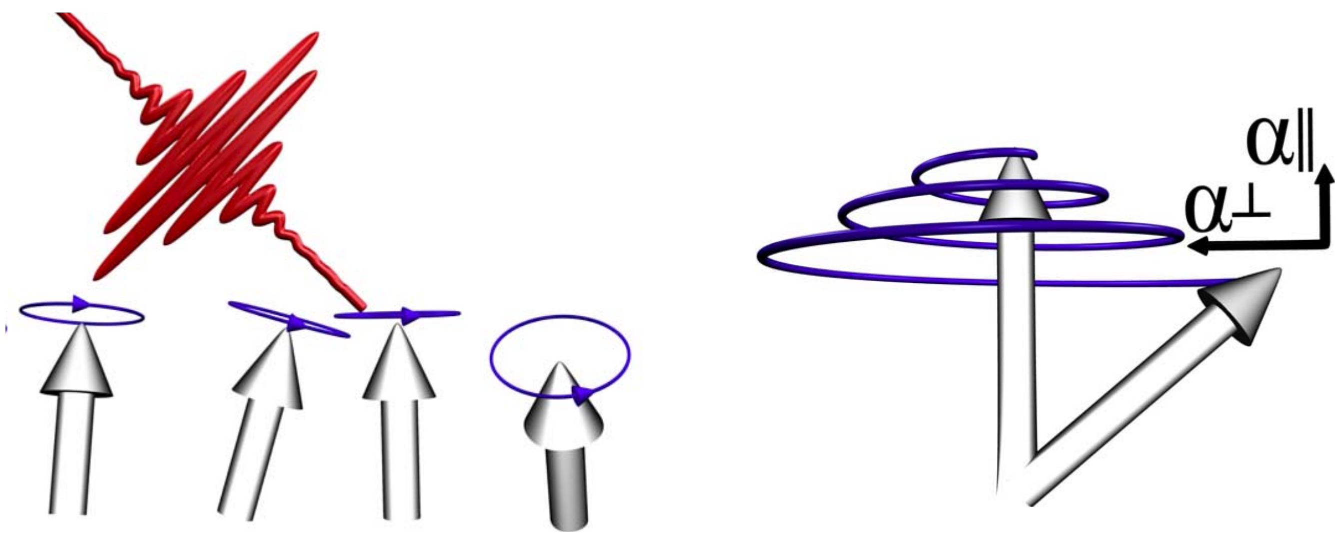
Zhang and Hübner, Phys. Rev. Lett. **85**, 3025 (2000)

Bigot, Vomir, Beaurepaire, Nature Phys. **5**, 515 - 520 (2009)

- Landau-Lifshitz-Bloch equations: spins coupled to bath; effective spin-orbit coupling includes spin-fluctuations (around T_c)

Chubykalo-Fesenko et al, Phys. Rev. B **74**, 094436 (2006)

Atxitia, Chubykalo-Fesenko, Walowski, Mann and Münzenberg Phys. Rev. B **81**, 174401 (2010)

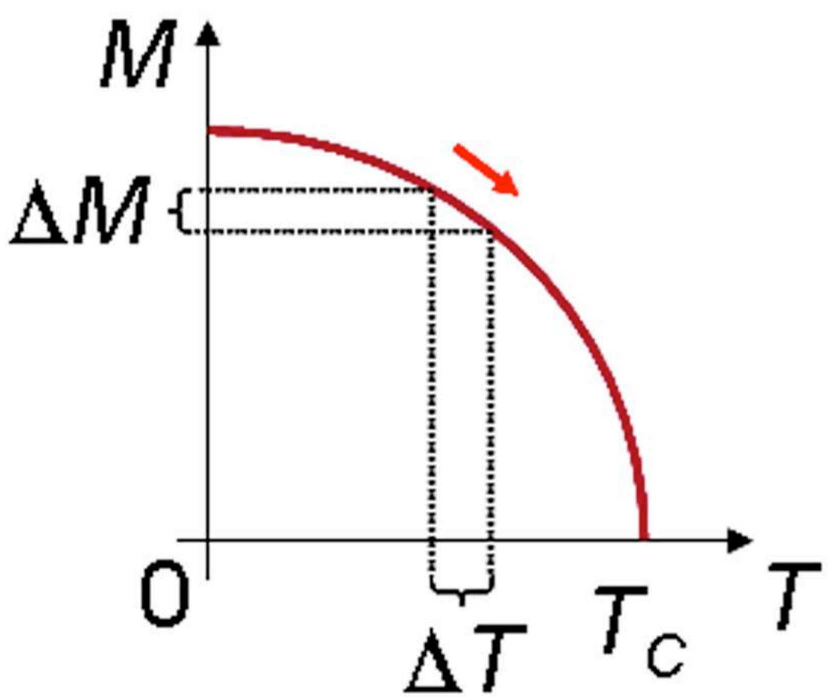


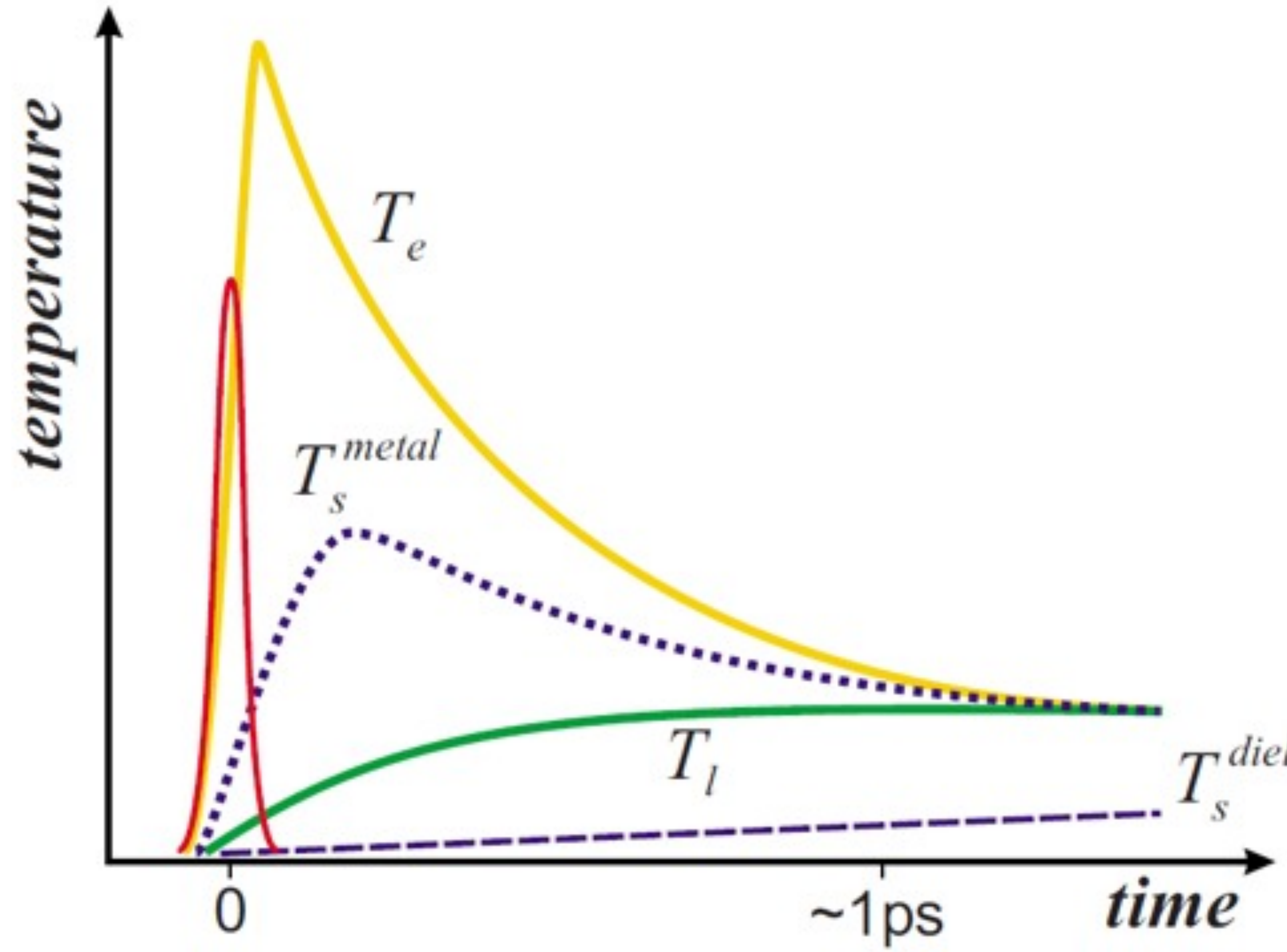
- Superdiffusive transport: electrons with different spin leave spot with different velocities

Battiato, Carva, and Oppeneer, PRL **105**, 027203 (2010)

(Phenomenological) Three-Temperature Model

- Three systems (electrons, lattice, and spins) in quasi-equilibrium: assign temperatures





from: Kirilyuk et al., Rev. Mod. Phys. **82**, 2731 (2010)

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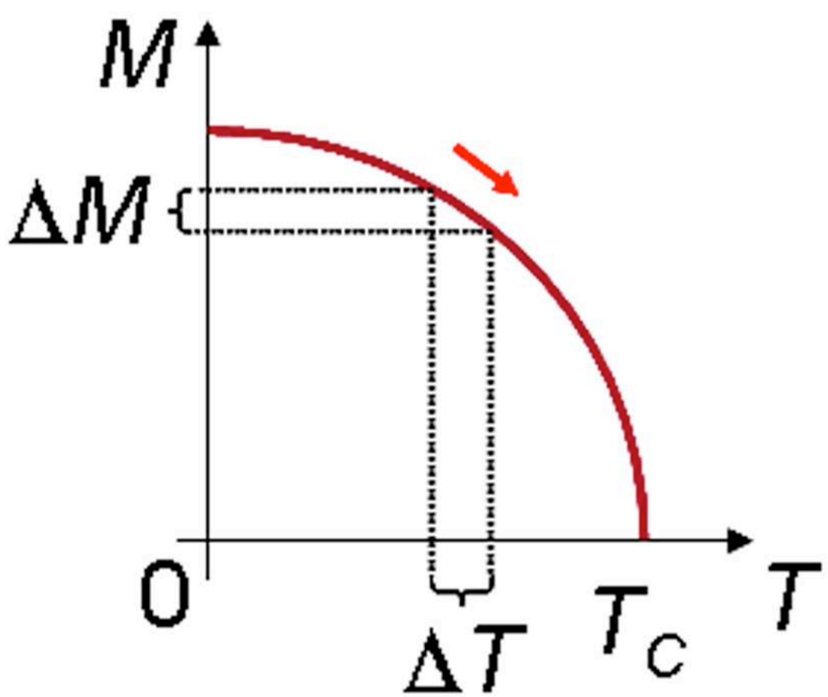

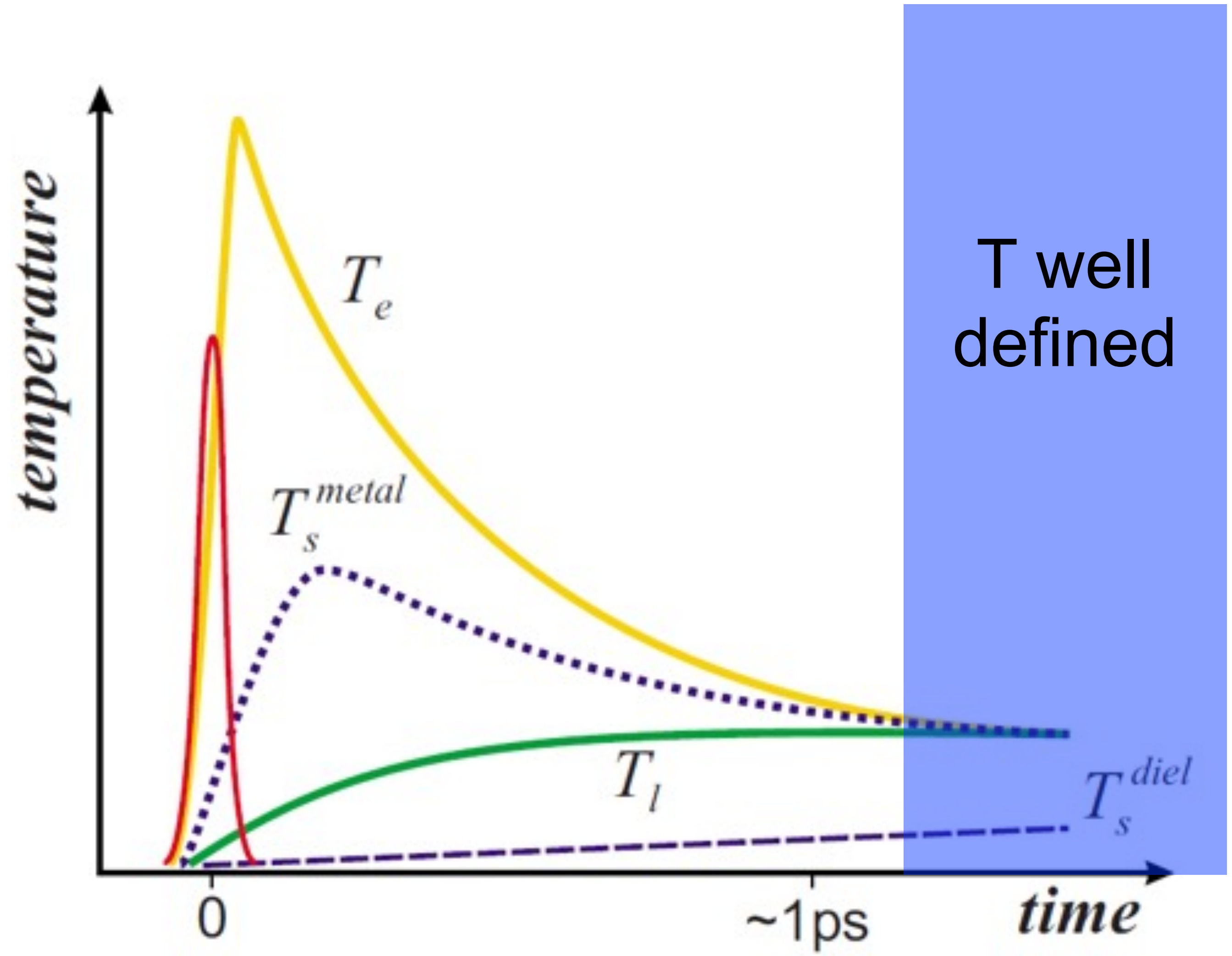

from: Kirilyuk et al., Rev. Mod. Phys. **82**, 2731 (2010)

- Separation and quasi-equilibrium assumption OK for picosecond time scale. But:

    How to describe ultrafast dynamics in the correlated electron system of the ferromagnet <u>microscopically</u>?

Outline

1. Ultrafast demagnetization in ferromagnets
2. Elliott-Yafet demagnetization due to electron-phonon scattering and optical excitation dynamics from ab-initio calculations
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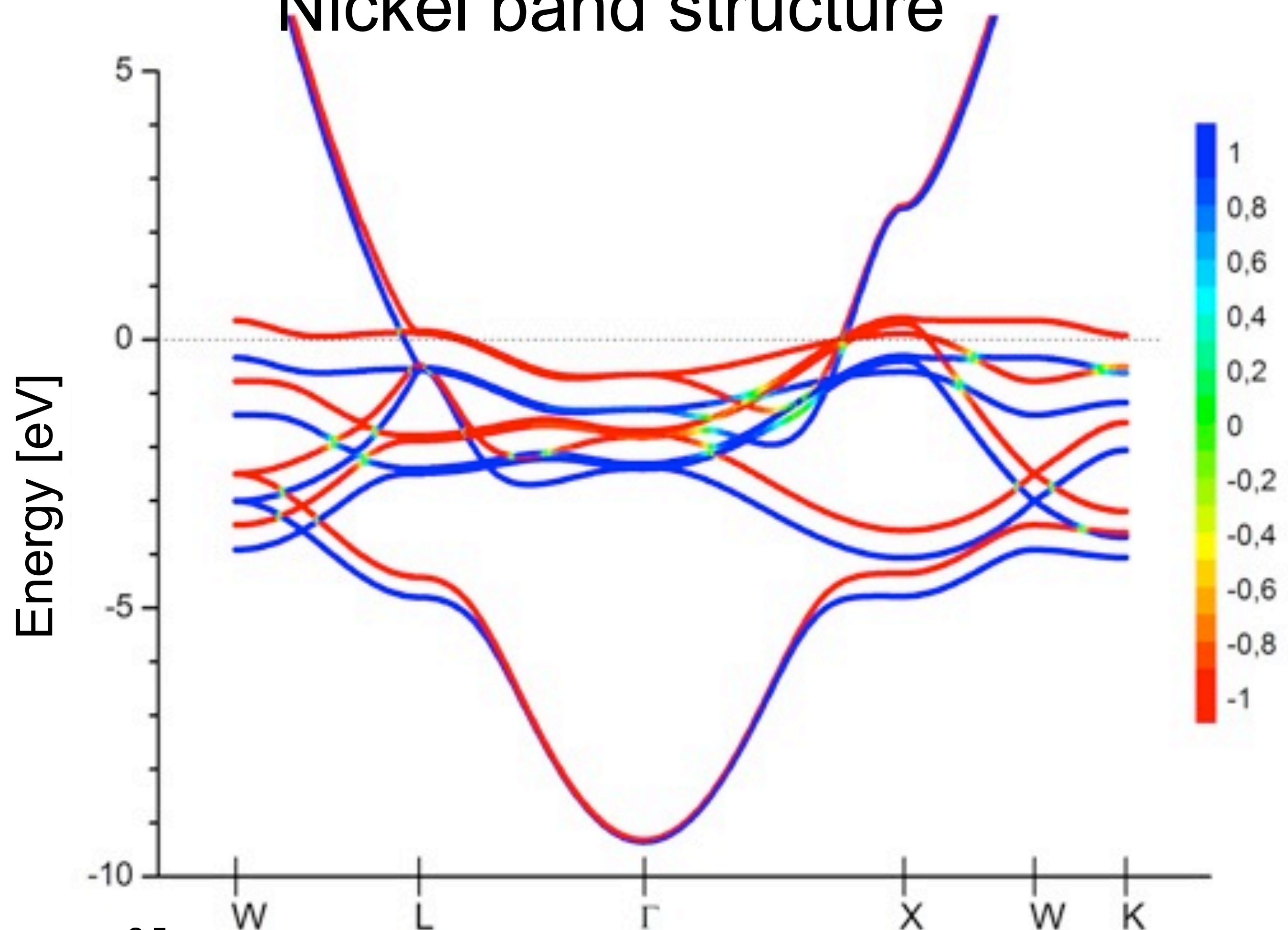
Scattering Dynamics in a Fixed Bandstructure

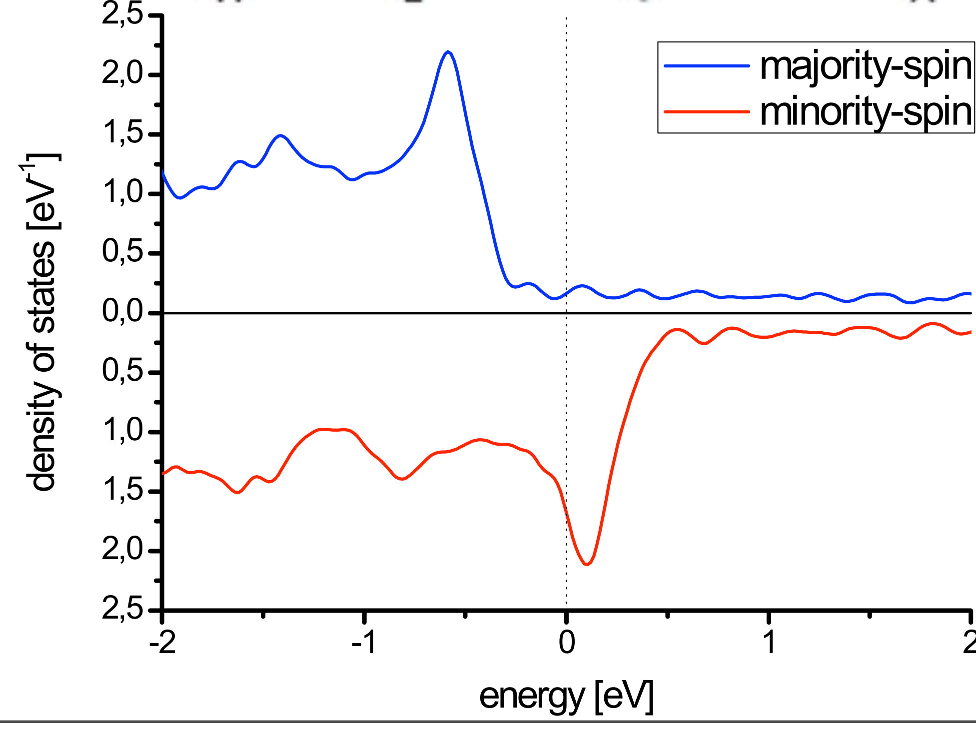
- Spin mixing important for optical excitation and scattering
- Spin mixing anisotropic ("spin hot-spots")?

  Fabian & Das Sarma, Phys. Rev. Lett. **81**, 5624 (1998)

- Keep band structure fixed!
- Parameter-free study of electronic dynamics due to electron-phonon scattering after ultrafast excitation!

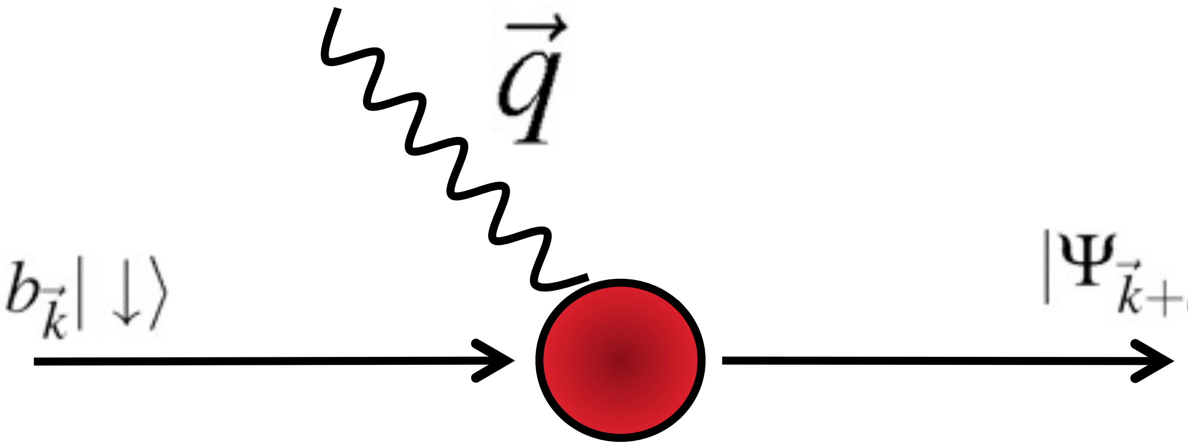
Nickel band structure





Elliott-Yafet Mechanism: Spin Relaxation due to Electron-Phonon Scattering

$|\Psi_{\vec{k}}\rangle = a_{\vec{k}}|\uparrow\rangle + b_{\vec{k}}|\downarrow\rangle$



$|\Psi_{\vec{k}+\vec{q}}\rangle = a_{\vec{k}+\vec{q}}|\uparrow\rangle + b_{\vec{k}+\vec{q}}|\downarrow\rangle$

Spin mixing + electron-phonon scattering = spin relaxation

$$\left\langle \Psi_{\vec{k}} \middle| \hat{S}_z \Psi_{\vec{k}} \right\rangle \neq \left\langle \Psi_{\vec{k}+\vec{q}} \middle| \hat{S}_z \Psi_{\vec{k}+\vec{q}} \right\rangle \Rightarrow \frac{d}{dt}\left\langle S_z \right\rangle \neq 0$$

- Phonons do <u>not</u> carry angular momentum (spin-diagonal interaction)

Yafet, Solid State Physics, **14** (1963)

- Extension of 3-temperature model: phonons with spin

Koopmans et al., Nature Mat. **9**, 256 (2010)

k-resolved Electron Scattering Dynamics

- Equation of motion for electronic dynamics

$$\frac{d}{dt} f^{\mu}(\vec{k}) = \frac{d}{dt} f^{\mu}(\vec{k})\Big|_{e-ph} + \frac{d}{dt} f^{\mu}(\vec{k})\Big|_{opt}$$

carrier distribution in
band μ with momentum k

- Optical excitation of carriers

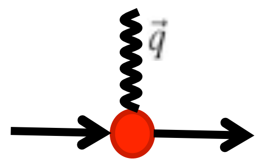
$$\frac{d}{dt} f^{\mu}(\vec{k})\Big|_{opt} = \frac{2\pi}{\hbar} \sum_{\nu \neq \mu} \left| \vec{d}_{\mu\nu} \cdot \vec{E} \right|^2 \left(f^{\nu}(\vec{k}) - f^{\mu}(\vec{k}) \right) g\left(\left| \epsilon^{\nu}(\vec{k}) - \epsilon^{\mu}(\vec{k}) \right| - \hbar\omega \right)$$

k-Resolved Electron-Phonon Scattering

- Electron-phonon Boltzmann scattering integrals

$$\frac{d}{dt} f^{\mu}(\vec{k}) = \sum_{\lambda} \sum_{\vec{q}} \left[w^{\lambda}_{\vec{k}+\vec{q},\mu' \to \vec{k},\mu} f^{\mu'}(\vec{k}+\vec{q}) \left(1 - f^{\mu}(\vec{k})\right) - w^{\lambda}_{\vec{k},\mu \to \vec{k}+\vec{q},\mu'} f^{\mu}(\vec{k}) \left(1 - f^{\mu'}(\vec{k}+\vec{q})\right) \right]$$

$$w^{\lambda}_{\vec{k},\mu \to \vec{k}+\vec{q},\mu'} = \frac{2\pi}{\hbar} \Bigg|$$



$$\Bigg|^2 \left[\tilde{n}^{\lambda}_{q} \delta\left(\epsilon^{\mu'}(\vec{k}+\vec{q}) - \epsilon^{\mu}(\vec{k}) - \hbar\omega^{\lambda}_{\vec{q}}\right) + \left(\tilde{n}^{\lambda}_{-\vec{q}} + 1\right) \delta\left(\epsilon^{\mu'}(\vec{k}+\vec{q}) - \epsilon^{\mu}(\vec{k}) + \hbar\omega^{\lambda}_{-\vec{q}}\right) \right]$$

S. Essert & H. C. Schneider,
Phys. Rev. B **84**, 224405 (2011)

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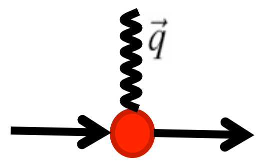

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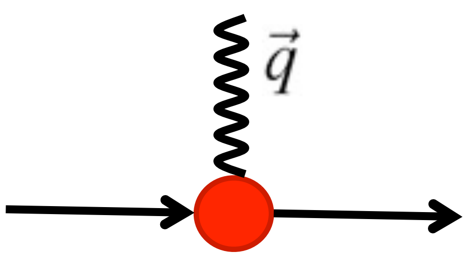
- Two contributions to spin-flip matrix element



$$\propto \sum_{\vec{R}} e^{i\vec{q}\cdot\vec{R}} \left\langle \psi_f \middle| \vec{\epsilon}_{\vec{q}} \cdot \nabla_{\vec{R}} \left(V + \frac{\hbar}{4m^2c^2} (\nabla_{\vec{r}} V \times \vec{p}) \cdot \vec{\sigma} \right) \middle| \psi_i \right\rangle$$

S. Essert & H. C. Schneider,
Phys. Rev. B **84**, 224405 (2011)

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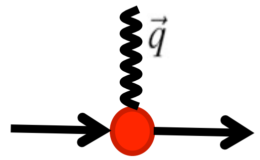

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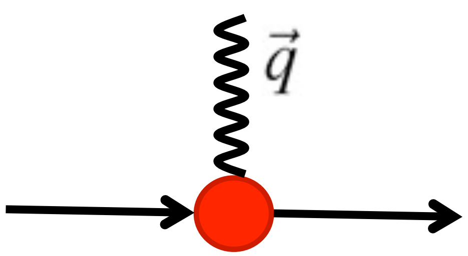

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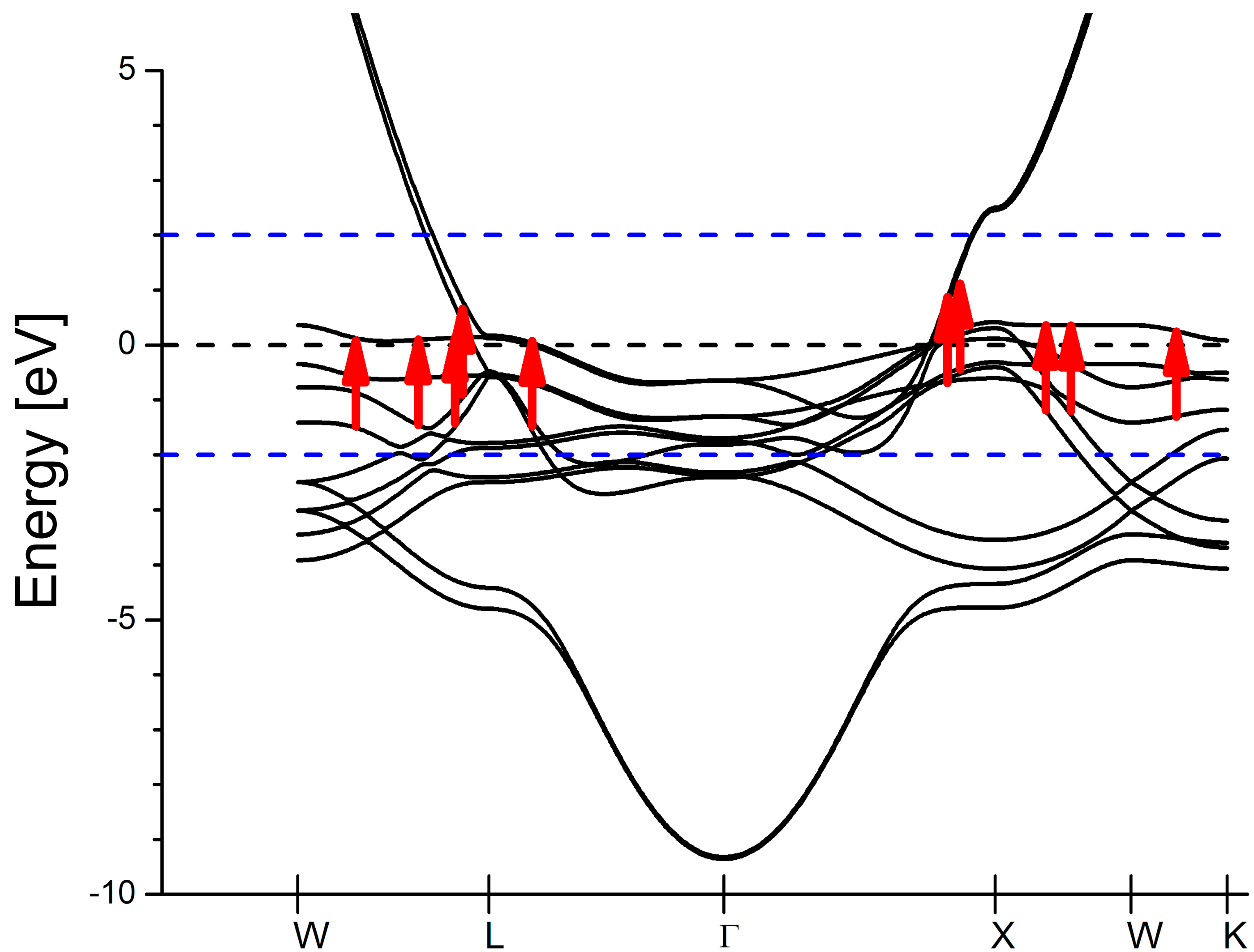
ab-initio input

S. Essert & H. C. Schneider, Phys. Rev. B **84**, 224405 (2011)

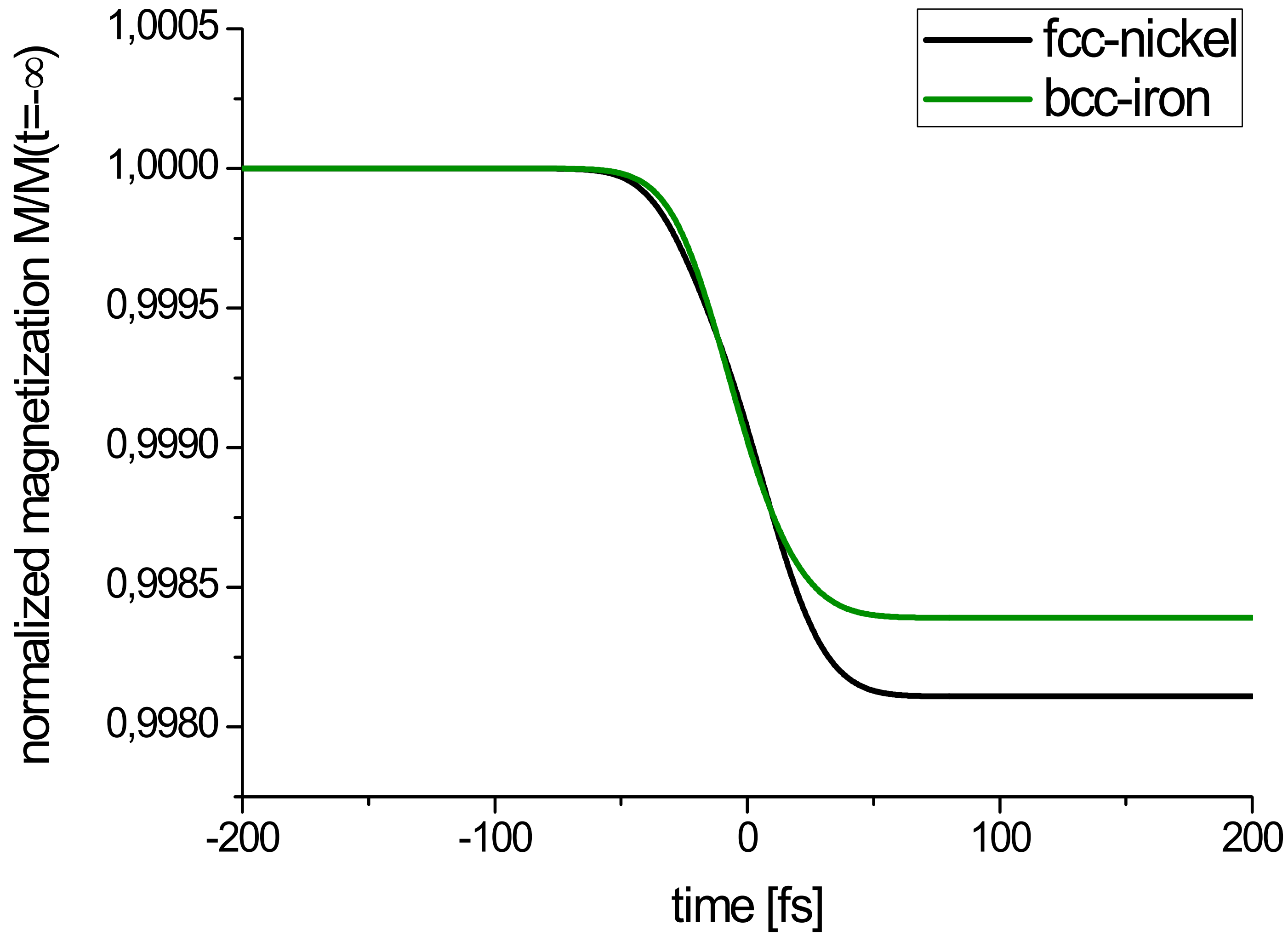
- Band structure @ T = 0K: $\epsilon^{\mu}(\vec{k})$
- Phonon dispersion $\omega^{\lambda}_{\vec{q}}$
- Transition dipole matrix elements $\vec{d}_{\mu\nu}$
- Electron-phonon matrix elements $M^{\lambda}_{\vec{k},\mu;\vec{k}',\mu'}$

Optical Excitation: Dipole Transitions in Nickel

- Dipole transitions with photon energy 1.55 eV in different regions of the Brillouing zone



Optical Excitation (2)

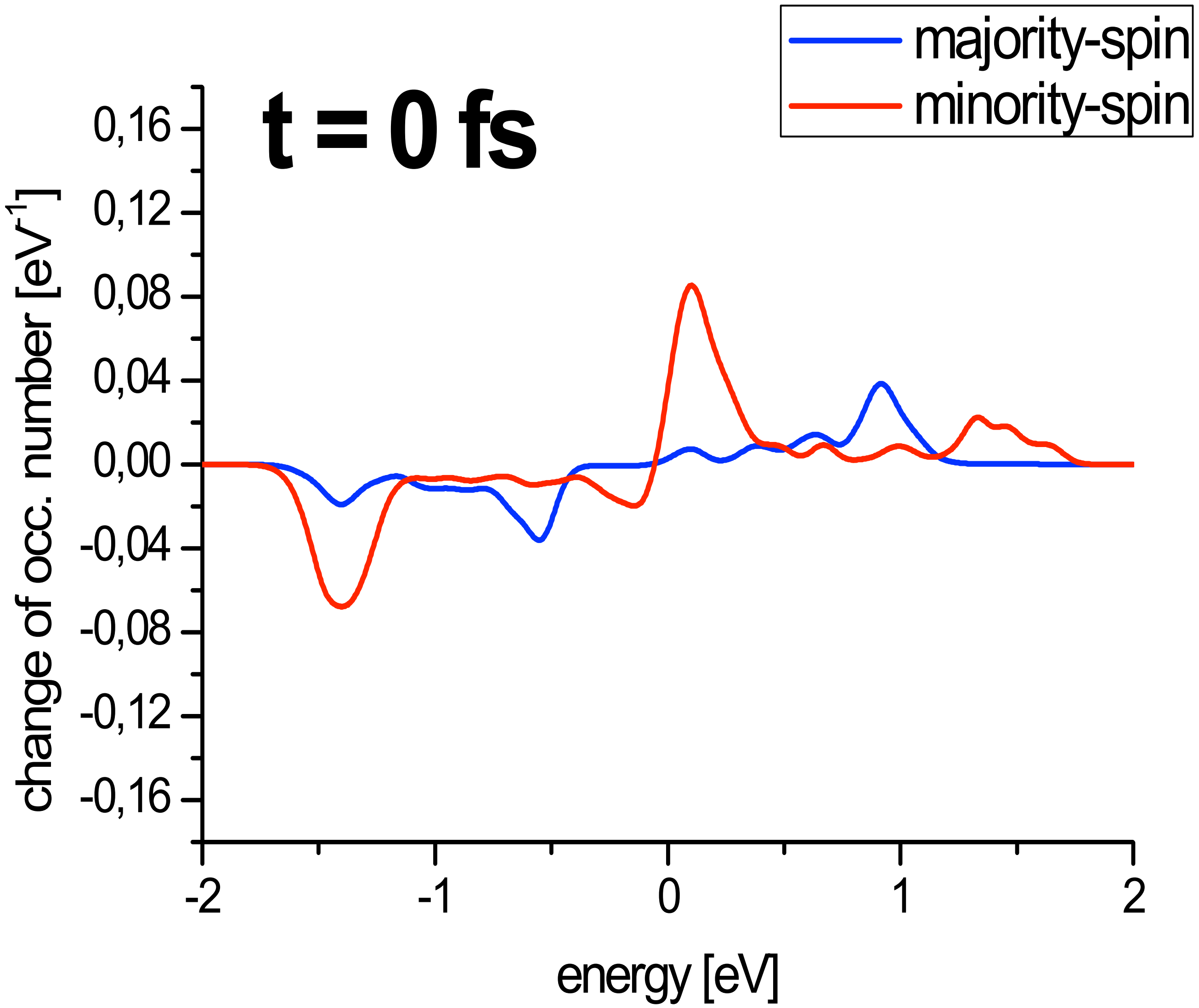


- Optical excitation using ultrashort pulse (1.55 eV, 50fs, 4 mJ/cm^{-2})
- Demagnetization is not caused by spin mixing during optical excitation

S. Essert & H. C. Schneider,
Phys. Rev. B **84**, 224405 (2011)

Optical Excitation in Nickel

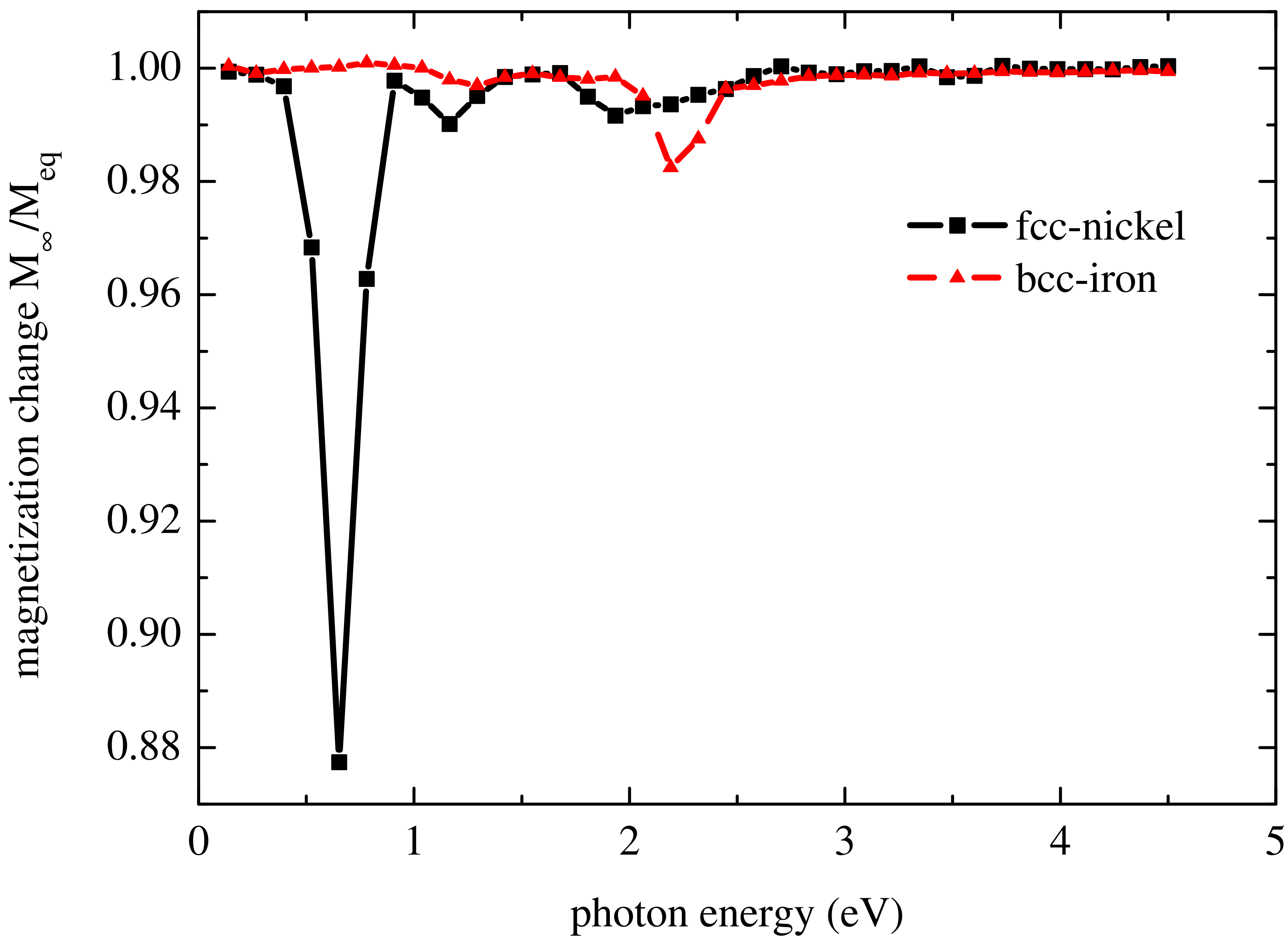
- Energy resolved change in carrier occupation
- Optical excitation using ultrashort pulse (1.55 eV, 50fs, 4 mJ/cm^{-2})
- Mainly minority electrons (and holes!) excited



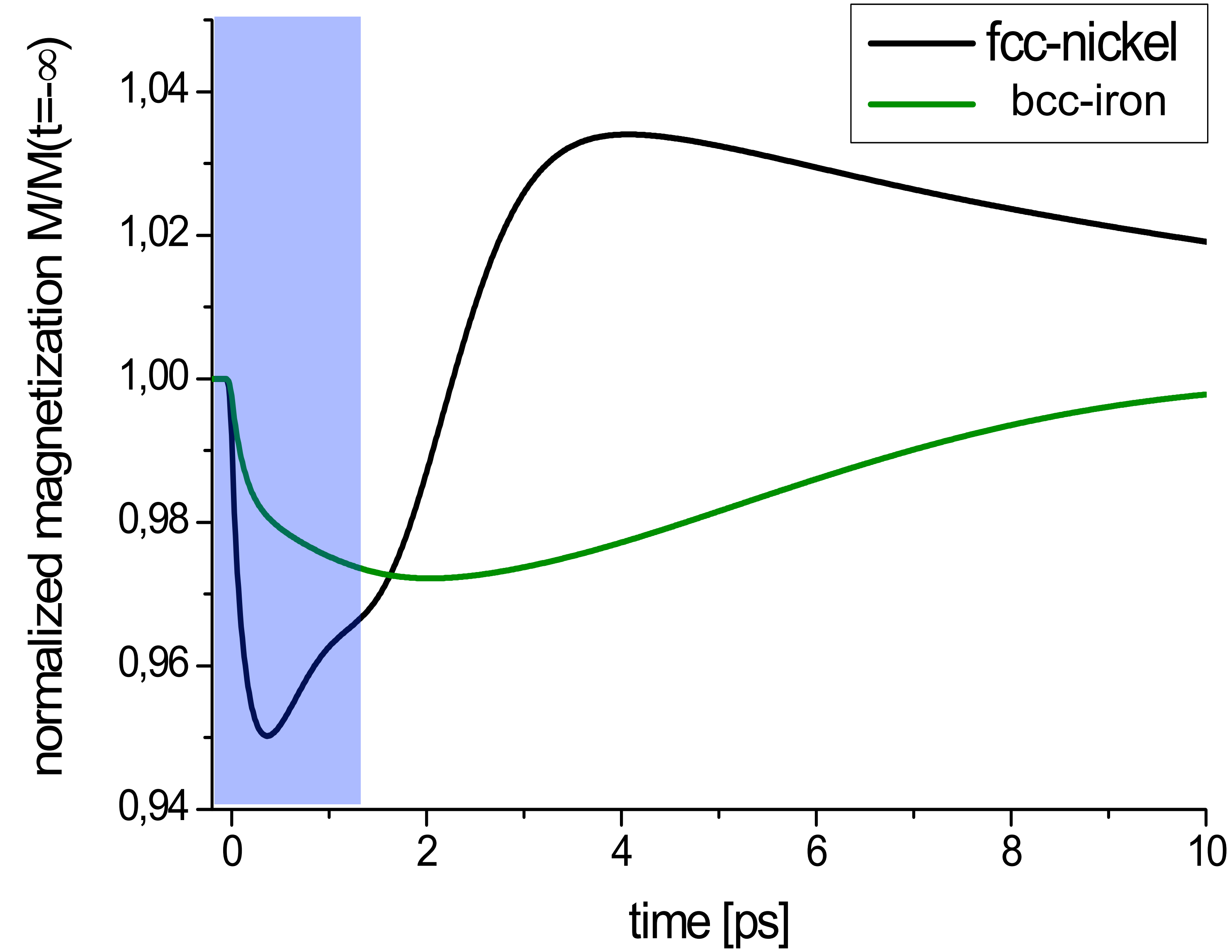
$$\Delta N_\tau(E) = \sum_{\vec{k}} \sum_{\mu} \delta(E - \varepsilon_k^\mu) \langle \sigma_\tau \rangle_k^\mu \left[f_{\vec{k}}^\mu(t) - F(\varepsilon_{\vec{k}}^\mu, T_0) \right]$$

Optical Excitation: Frequency Dependence

- Influence of band structure/spin-mixing on optical excitation



Magnetization Dynamics after Optical Excitation



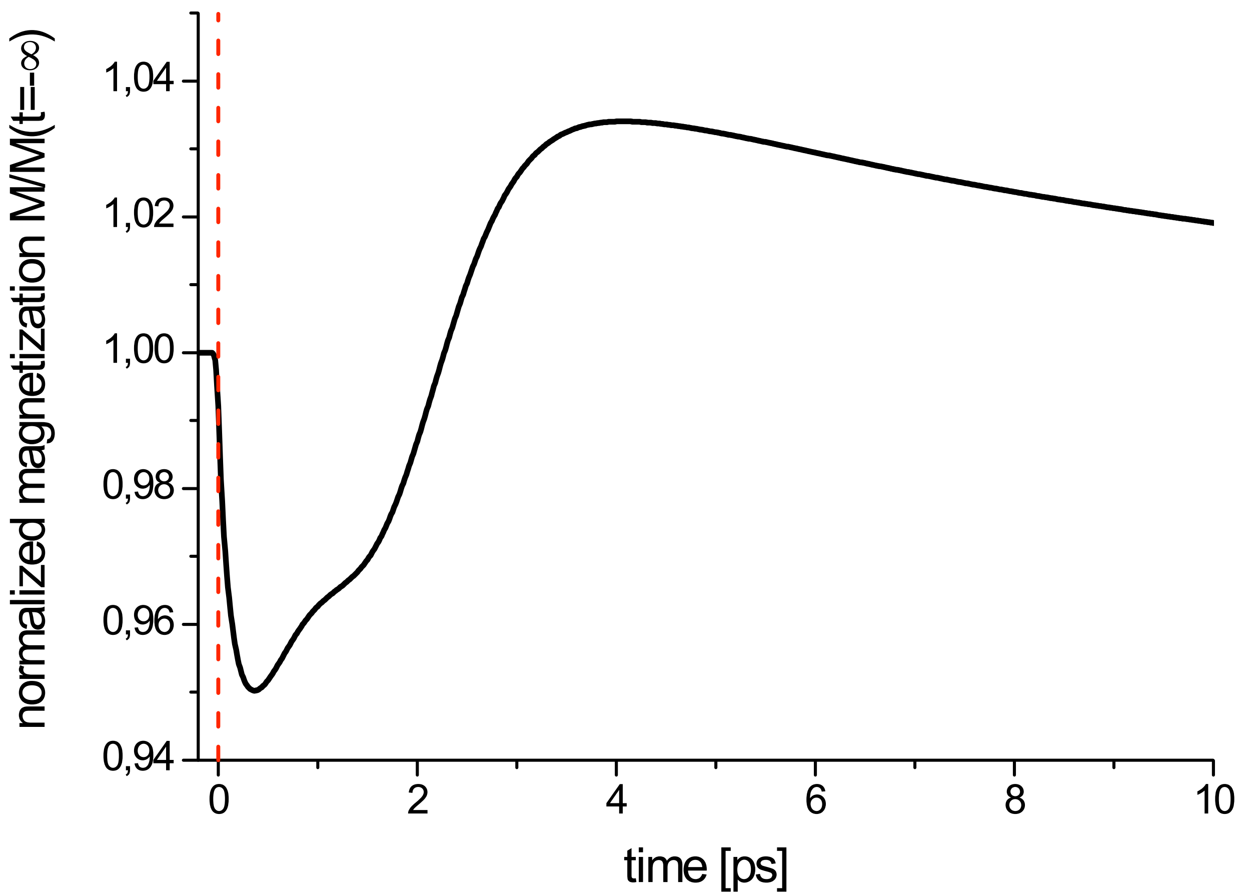
Essert & Schneider, Phys. Rev. B **84**, 224405 (2011)

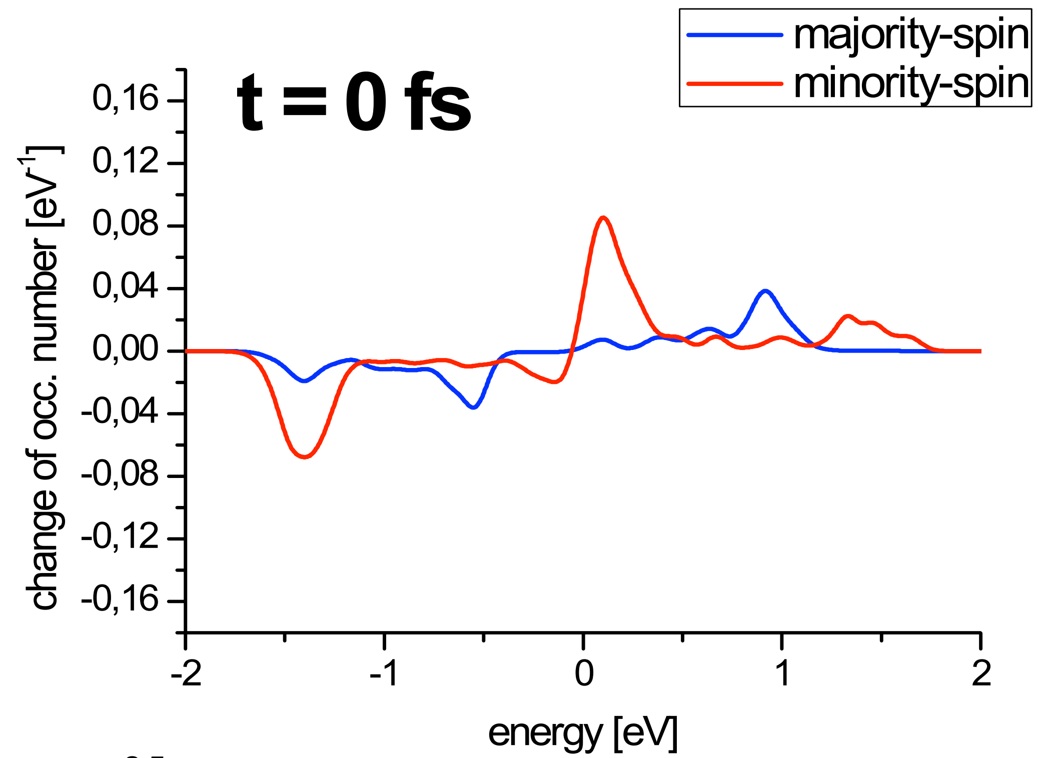
agreement with Carva, Battiato and Oppeneer, PRL **107**, 207201 (2011)

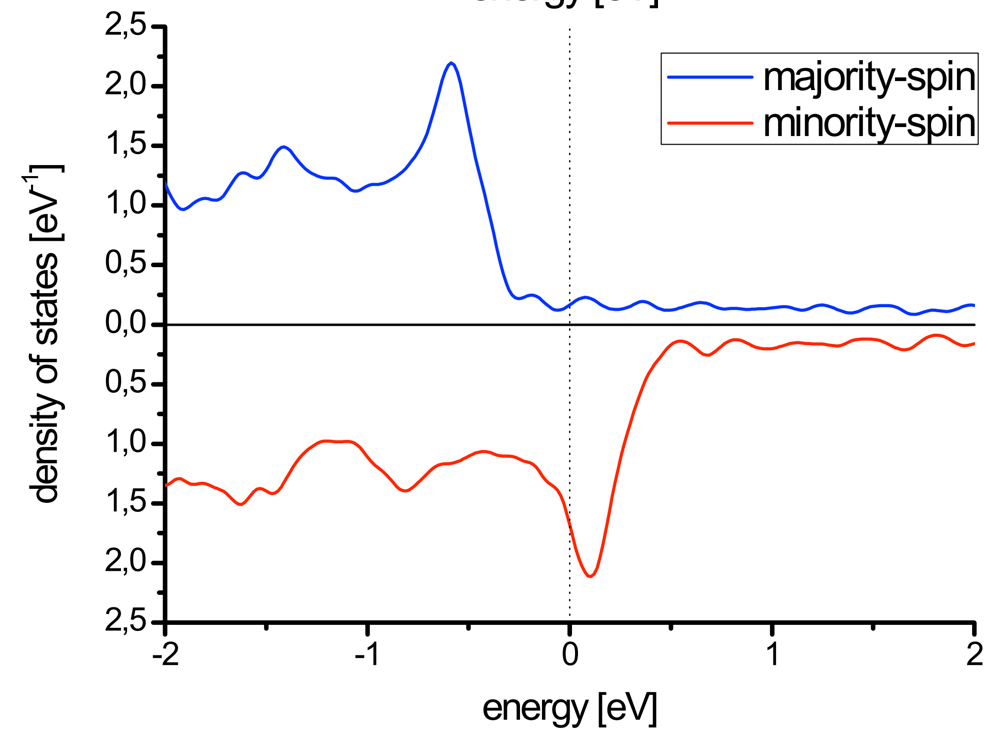
- Demagnetization mainly due to hole scattering
- Optical excitation and electron-phonon-scattering cannot explain the observed demagnetization
- Other scattering mechanisms?

Energy-Resolved Dynamics: fcc nickel

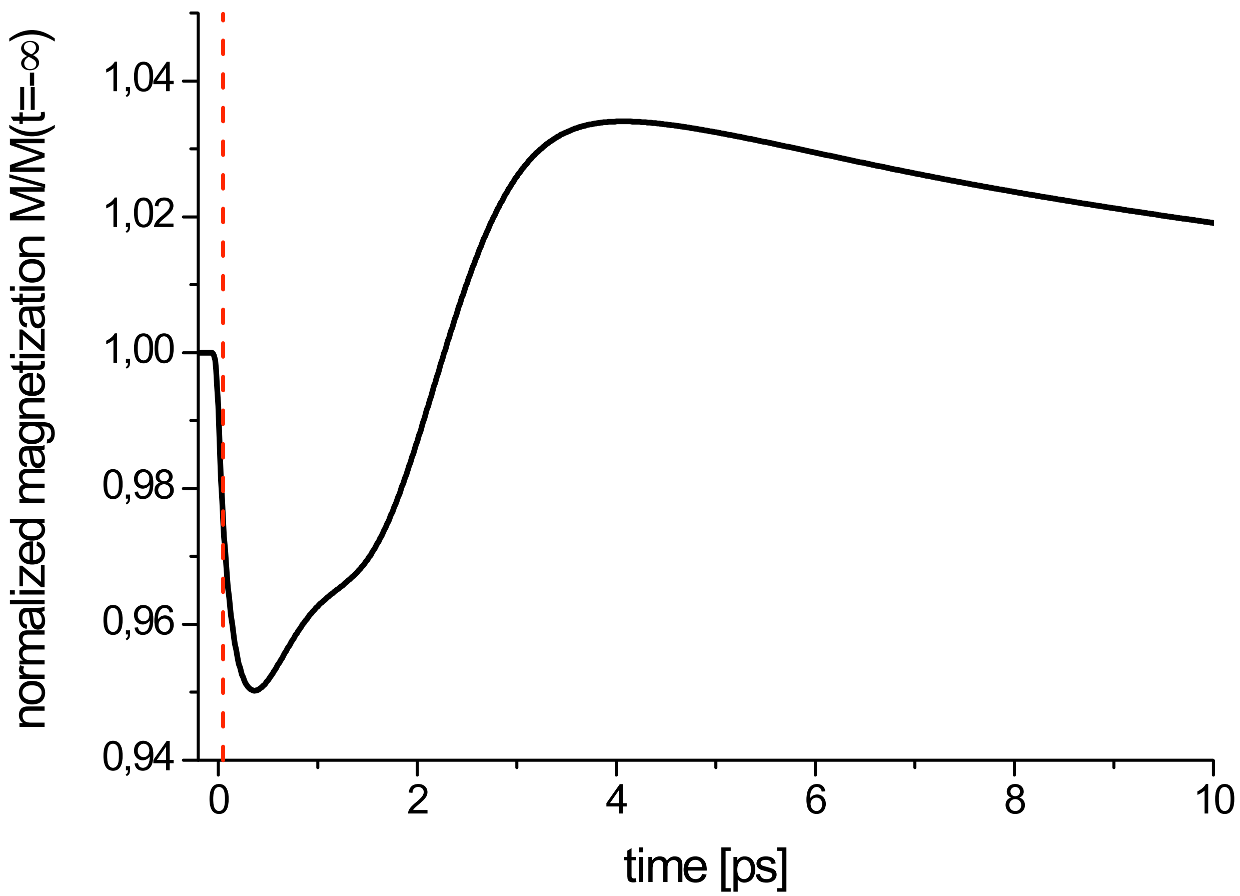
- Microscopic energy resolved dynamics

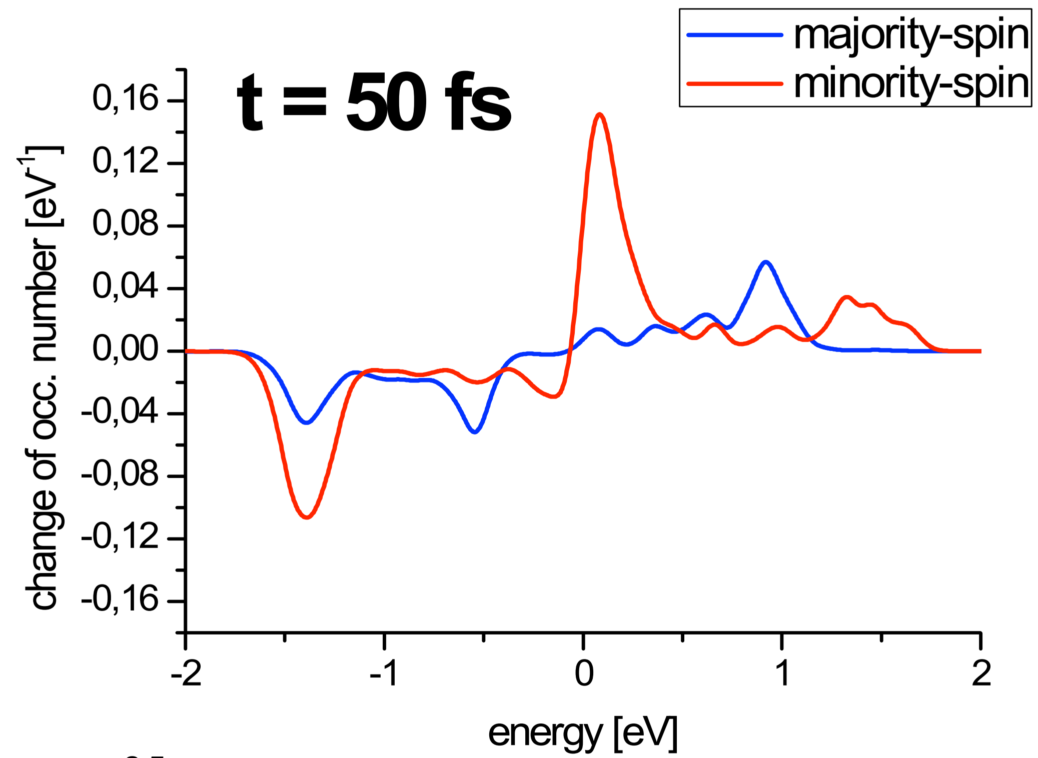


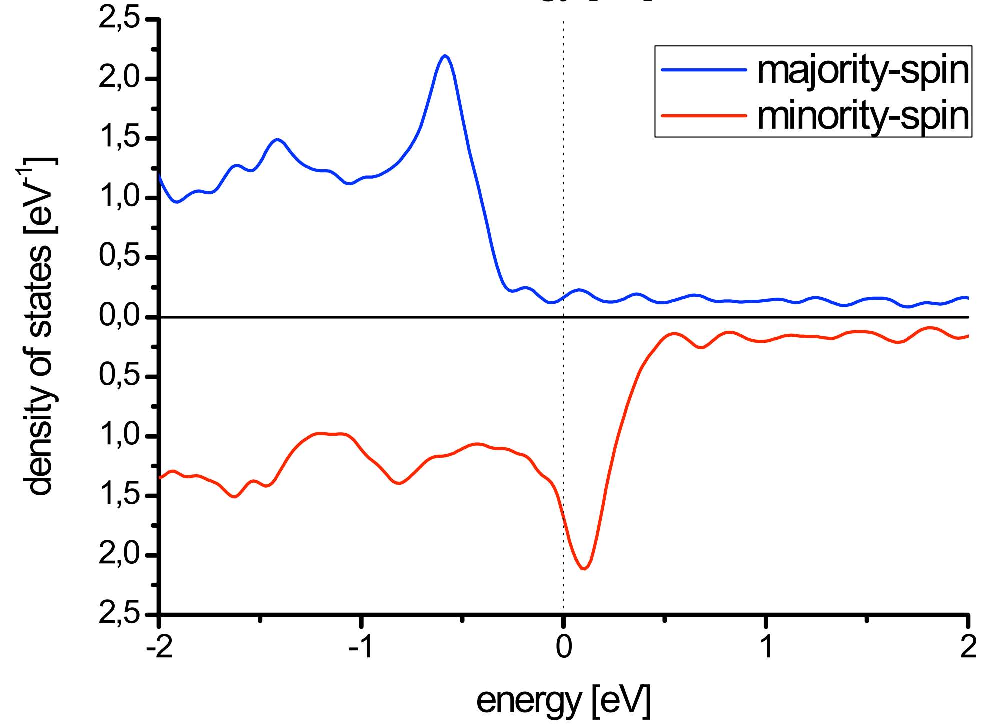




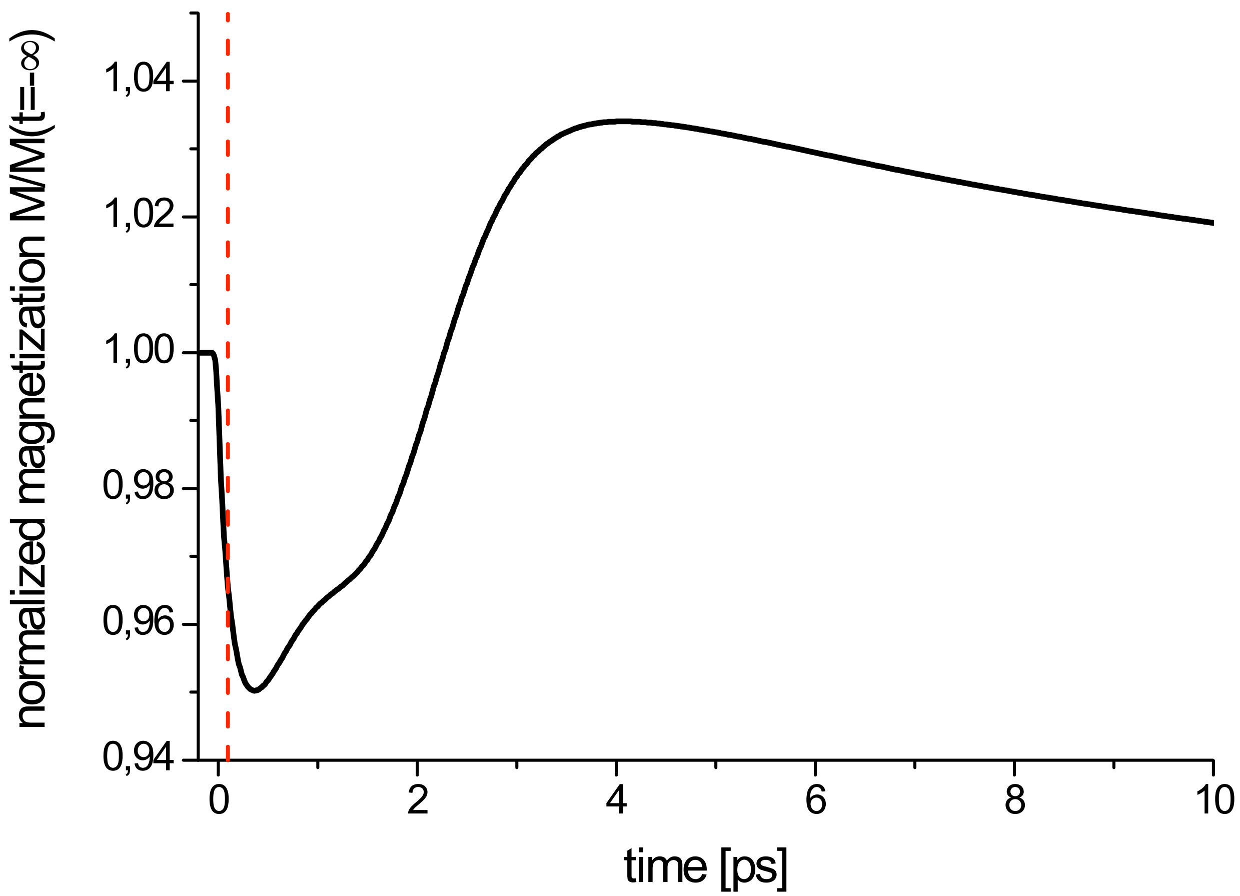
Energy-Resolved Dynamics: fcc nickel

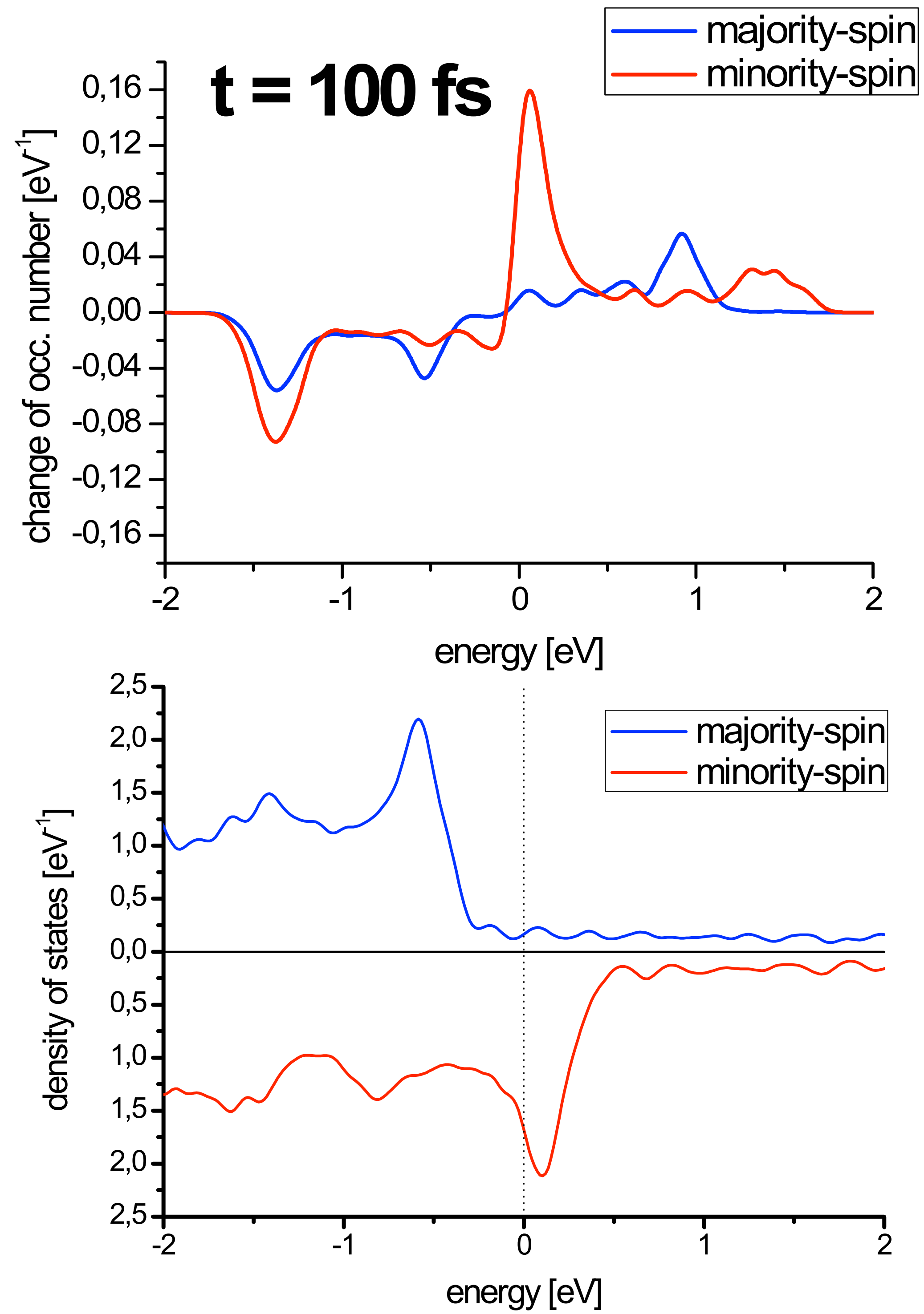




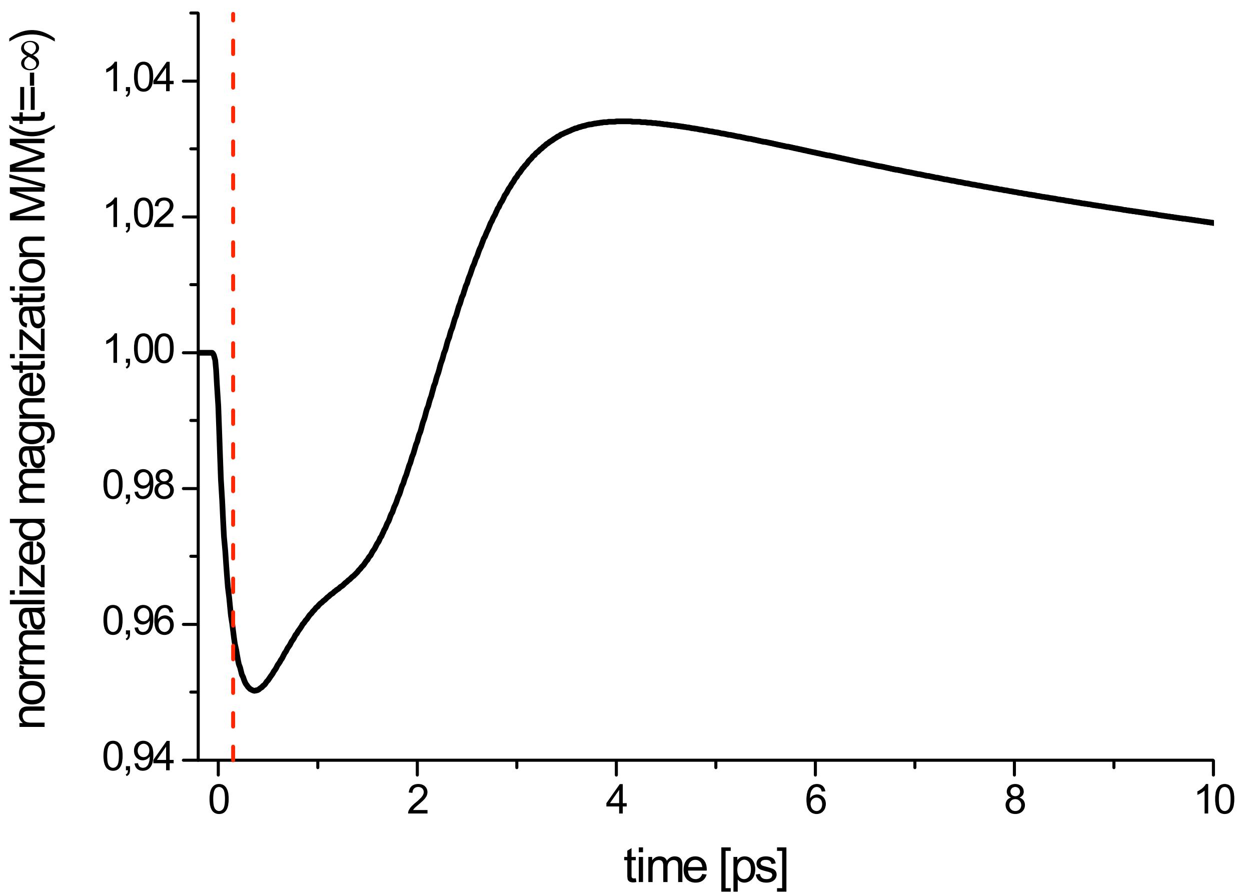


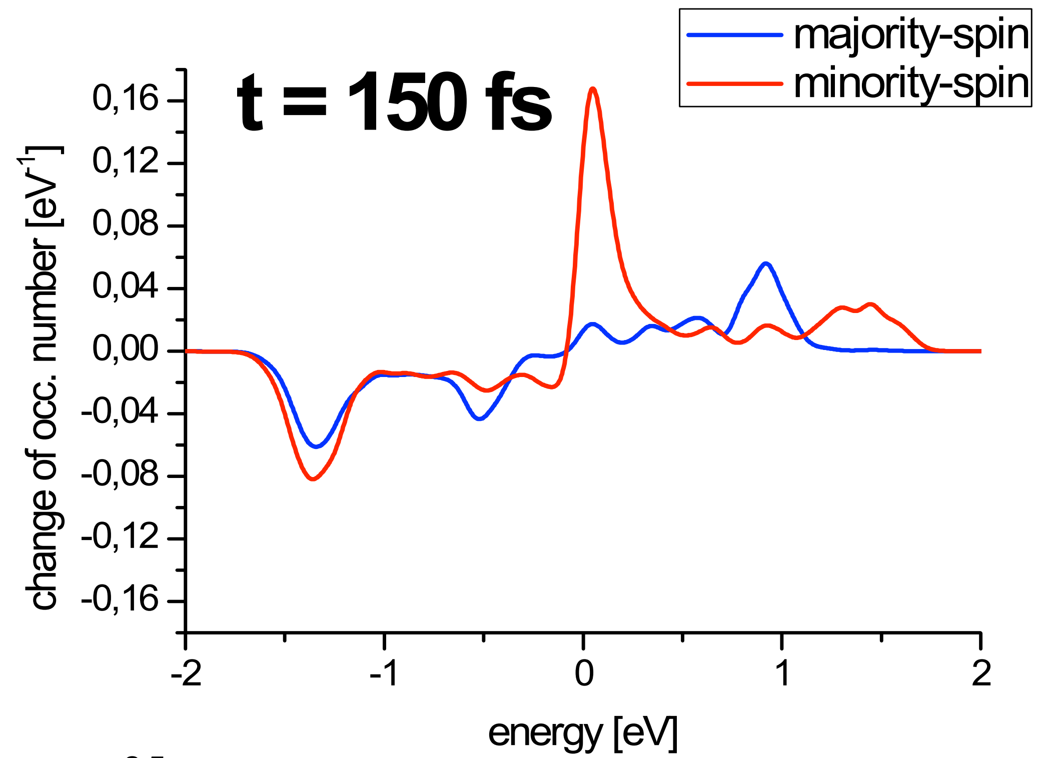
Energy-Resolved Dynamics: fcc nickel

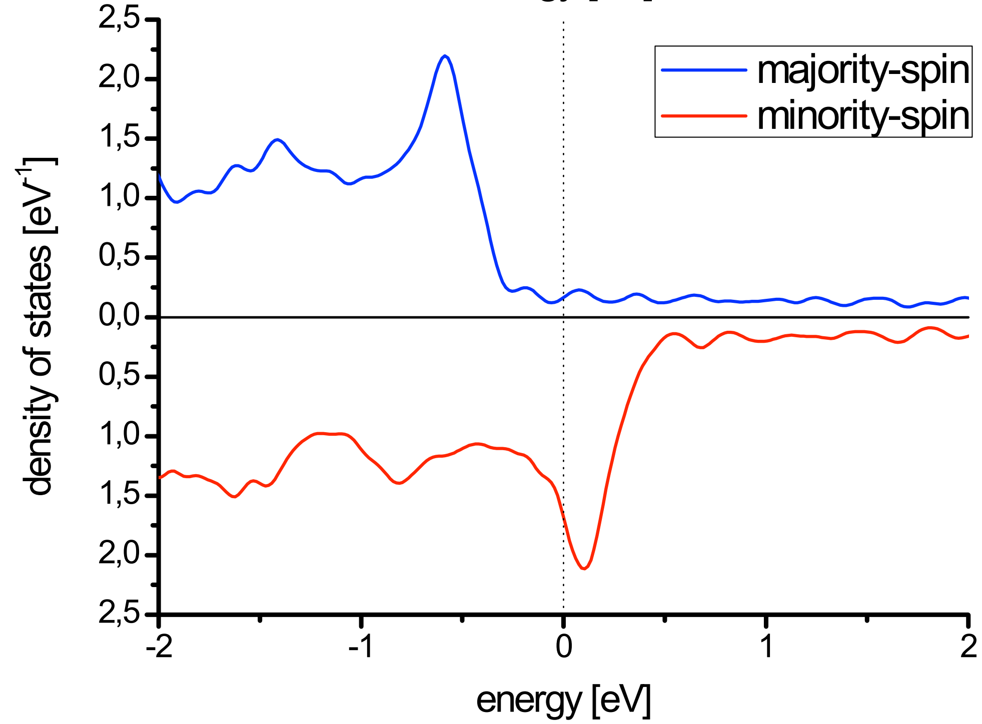




Energy-Resolved Dynamics: fcc nickel







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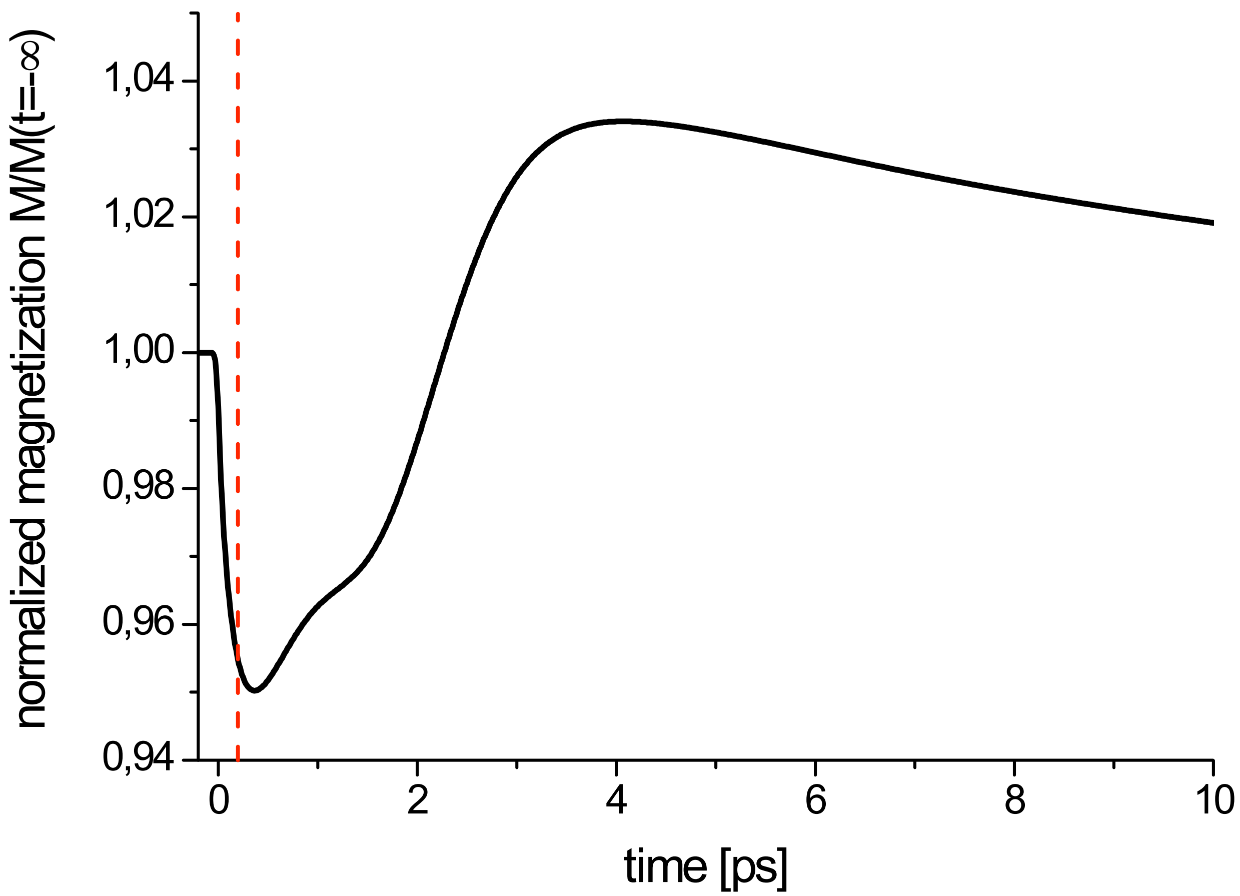
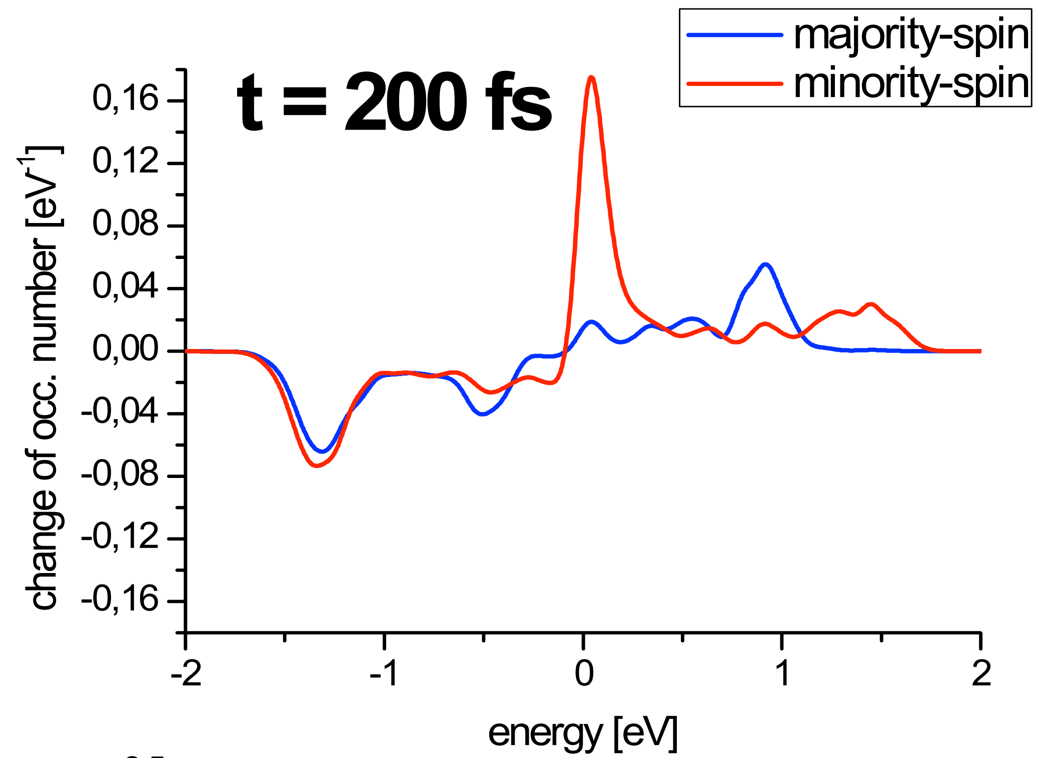
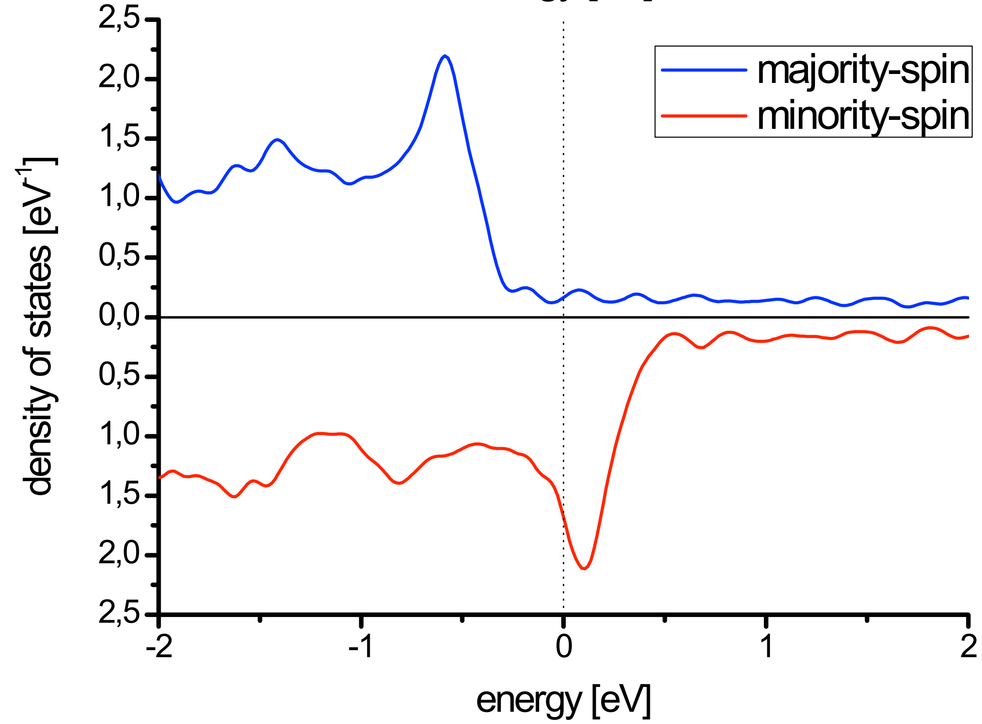
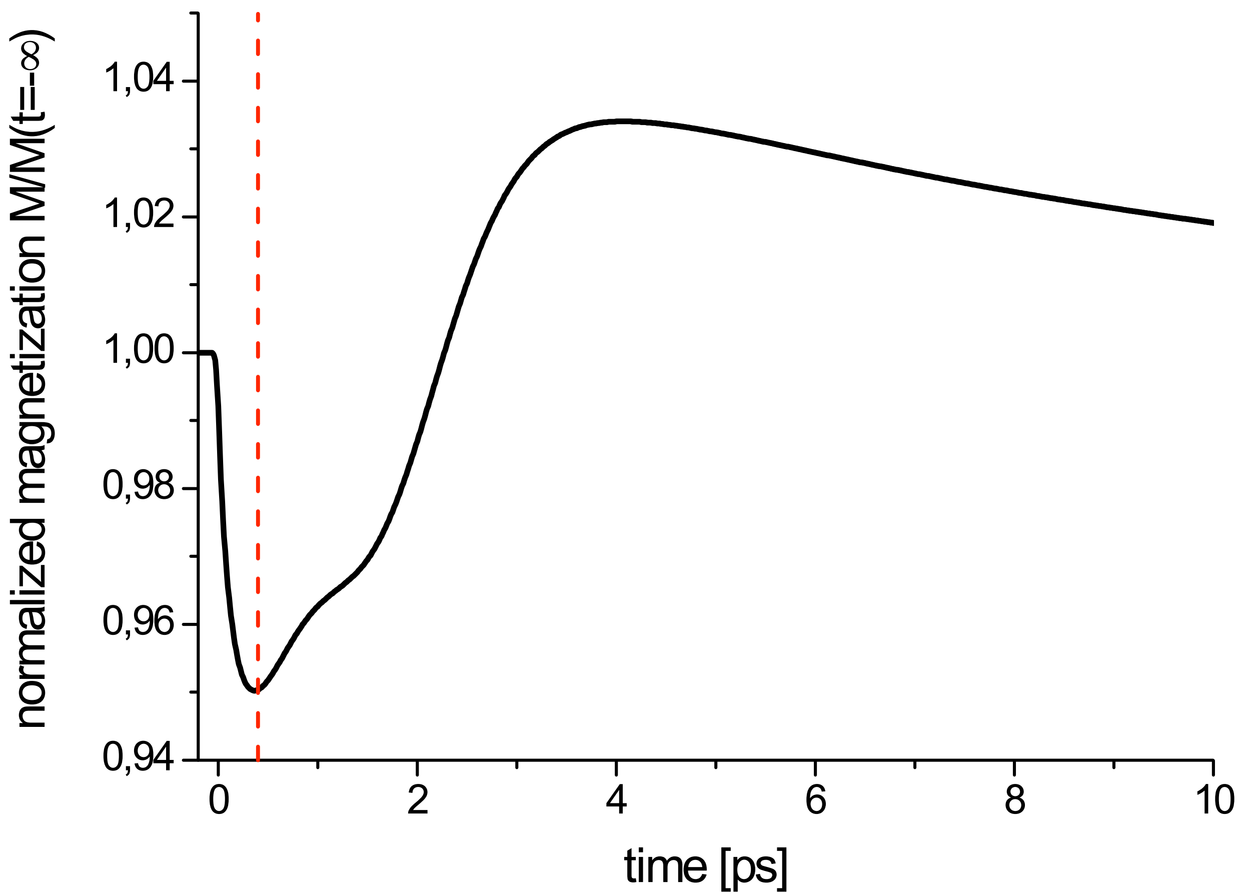
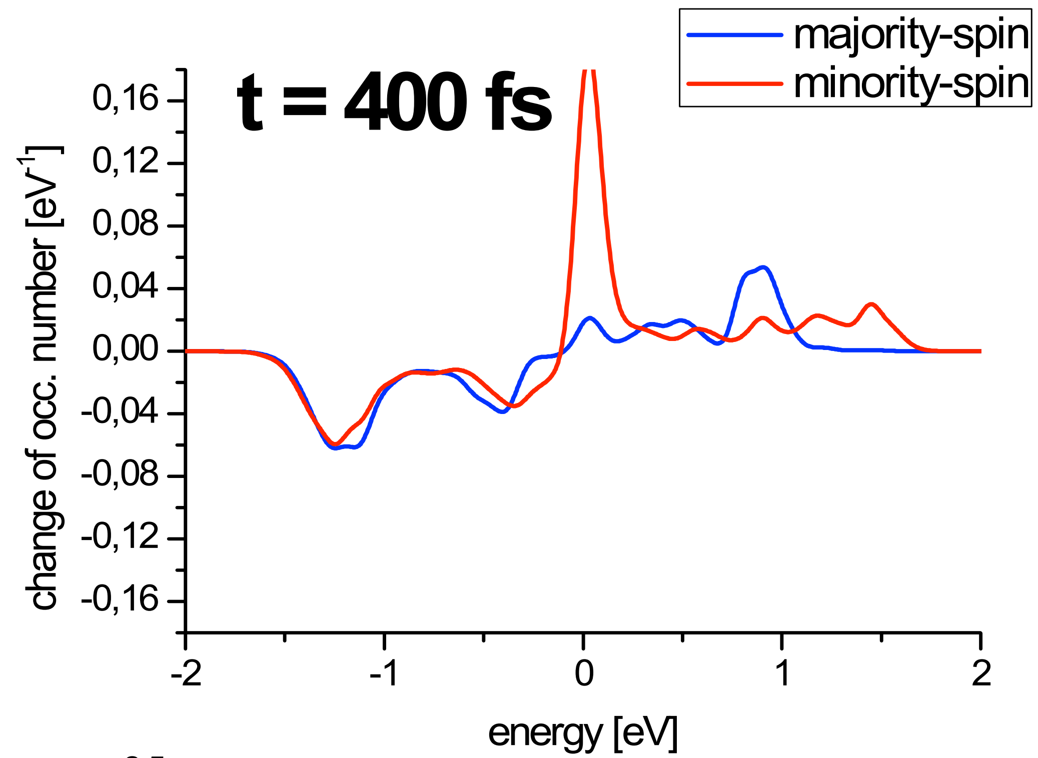


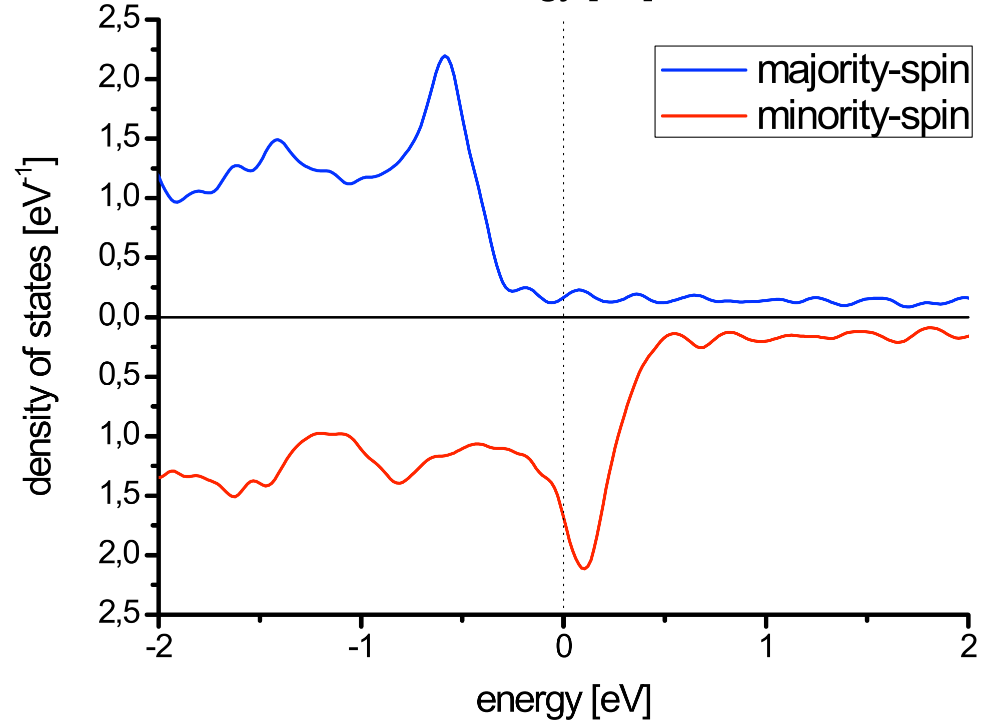




Energy-Resolved Dynamics: fcc nickel







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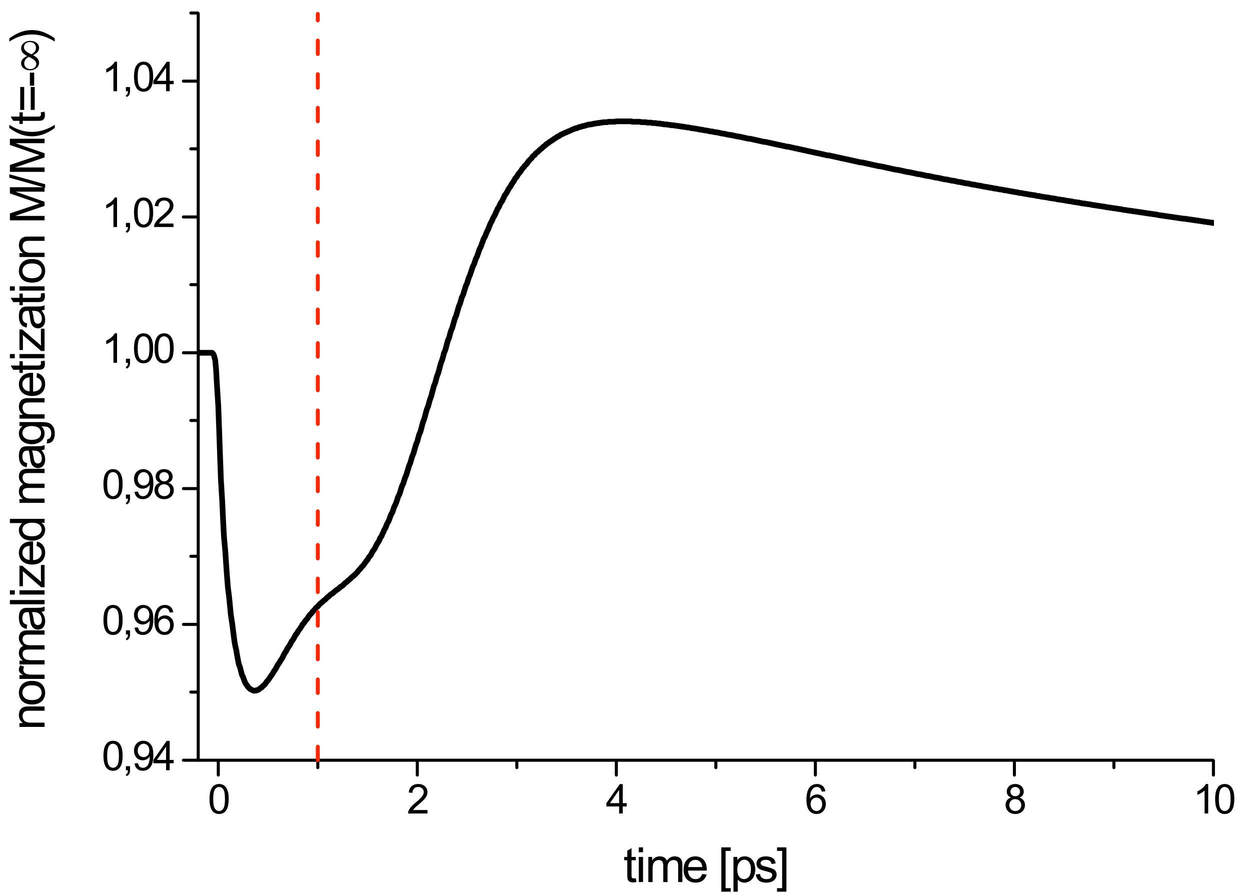
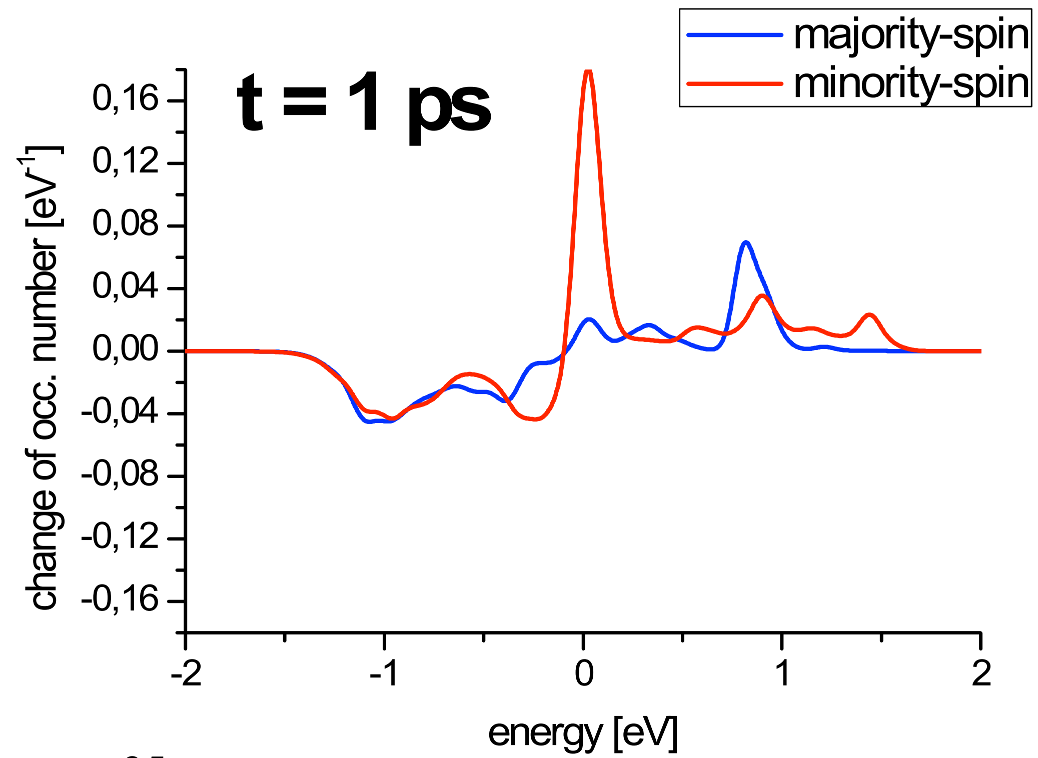
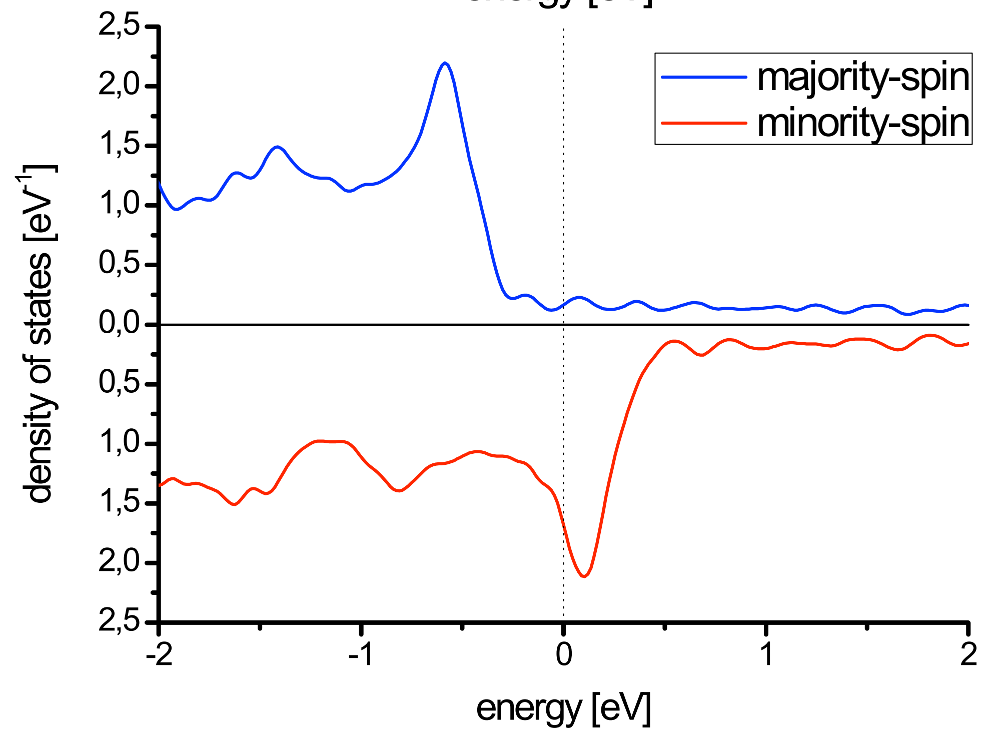
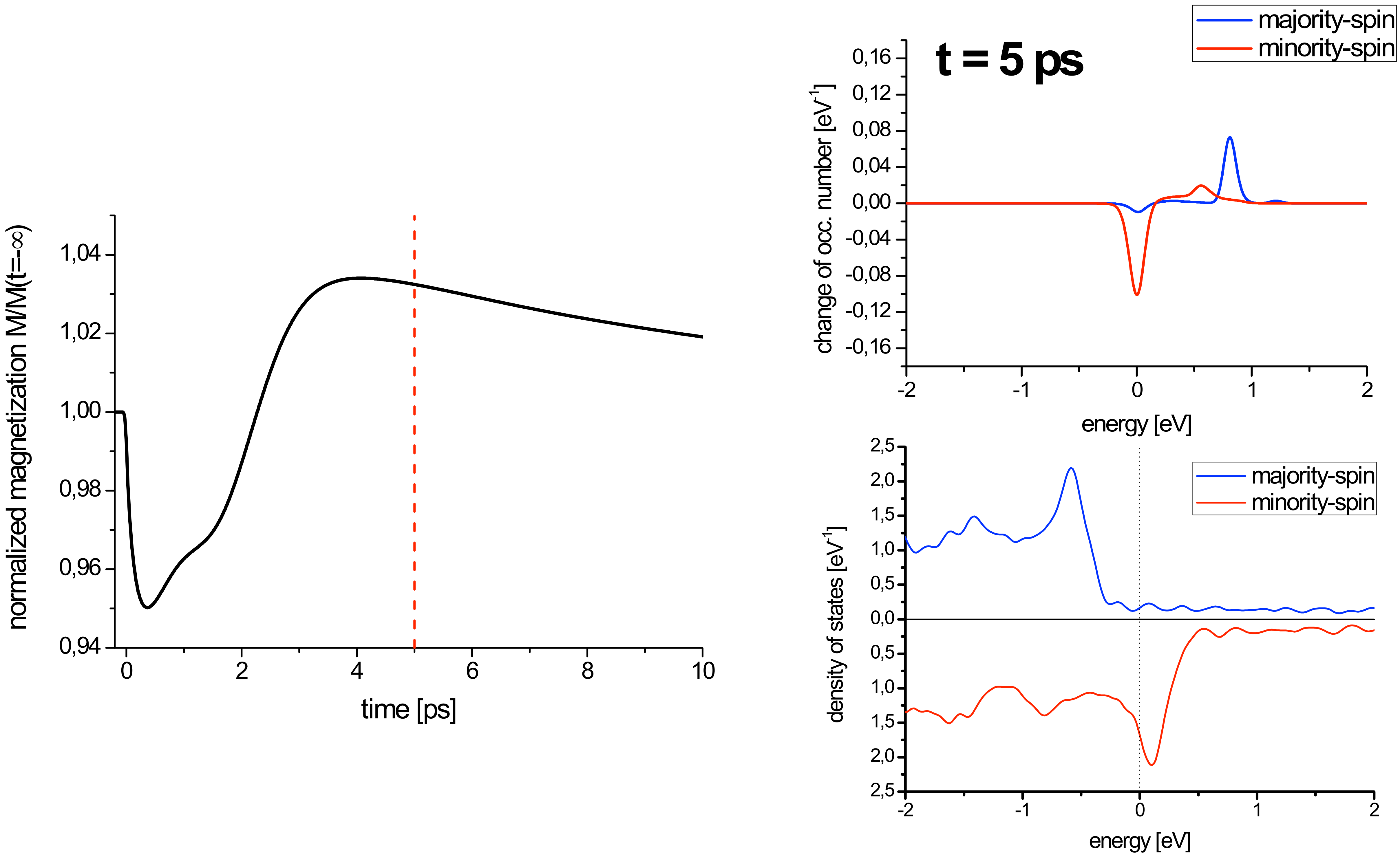






Energy-Resolved Dynamics: fcc nickel



Heating of the Lattice

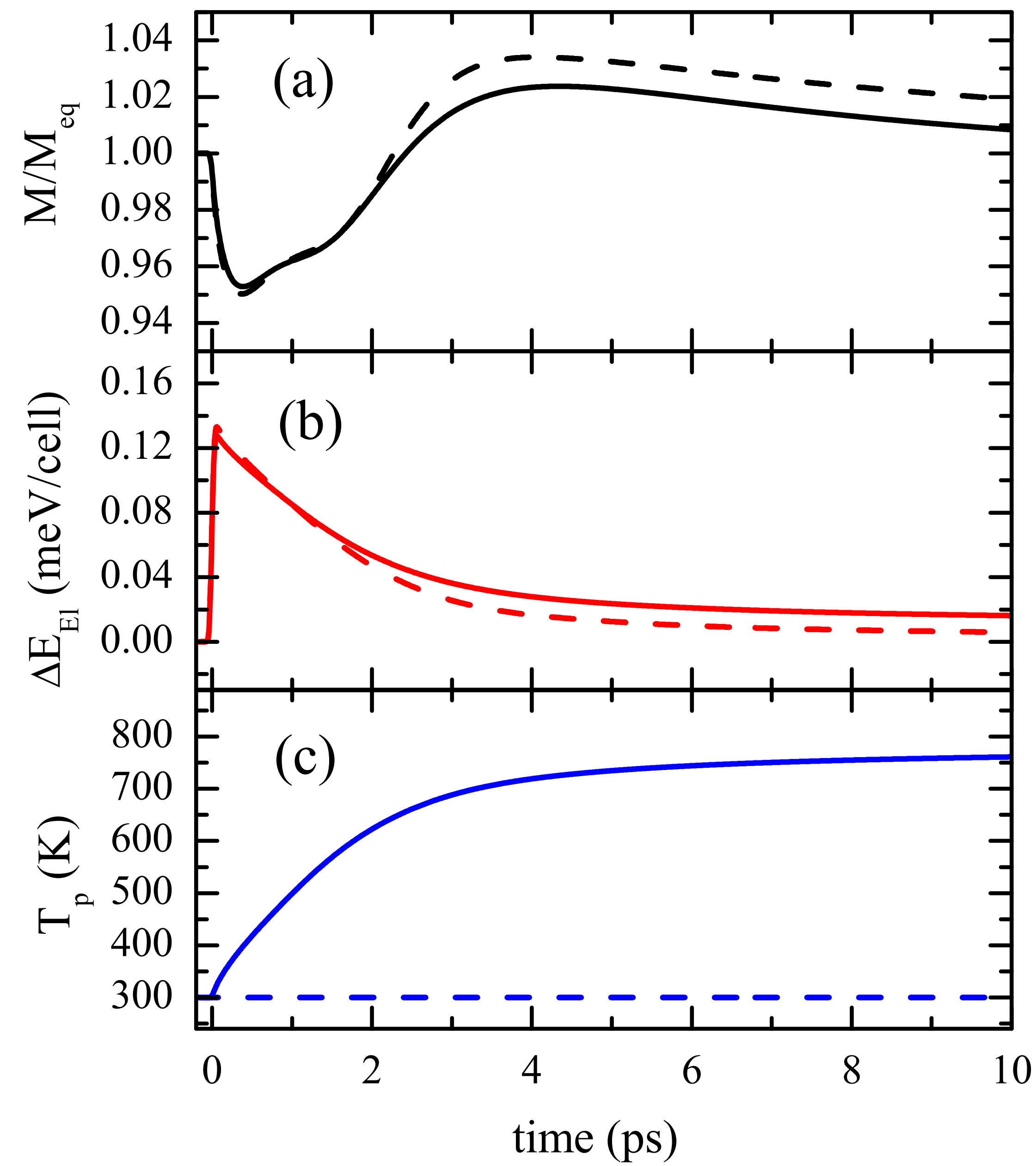
- Include dynamical lattice temperature

$$\frac{\partial}{\partial t}T_{\mathrm{p}} = \frac{1}{C_{\mathrm{p}}(T_{\mathrm{p}})}\frac{\partial E_{\mathrm{p}}}{\partial t} = -\frac{1}{C_{\mathrm{p}}(T_{\mathrm{p}})}\left.\frac{\partial E_{\mathrm{e}}}{\partial t}\right|_{\text{e-p}}$$
$$= -\frac{1}{C_{\mathrm{p}}(T_{\mathrm{p}})}\sum_{\mu,\vec{k}}\epsilon^{\mu}_{\vec{k}}\left.\frac{\partial}{\partial t}n^{\mu}_{\vec{k}}\right|_{\text{e-p}},$$

- Heat capacity

$$C_{\mathrm{p}}(T_{\mathrm{p}}) = \frac{\partial E_{\mathrm{p}}(T_{\mathrm{p}})}{\partial T_{\mathrm{p}}} = \sum_{\vec{q},\lambda}\hbar\omega^{\lambda}_{\vec{q}}\frac{\partial\tilde{n}^{\lambda}_{\vec{q}}(T_{\mathrm{p}})}{\partial T_{\mathrm{p}}}$$

- Change of scattering phase space: No qualitative difference!



Essert & Schneider J. App. Phys. **111**, 07C514 (2012)

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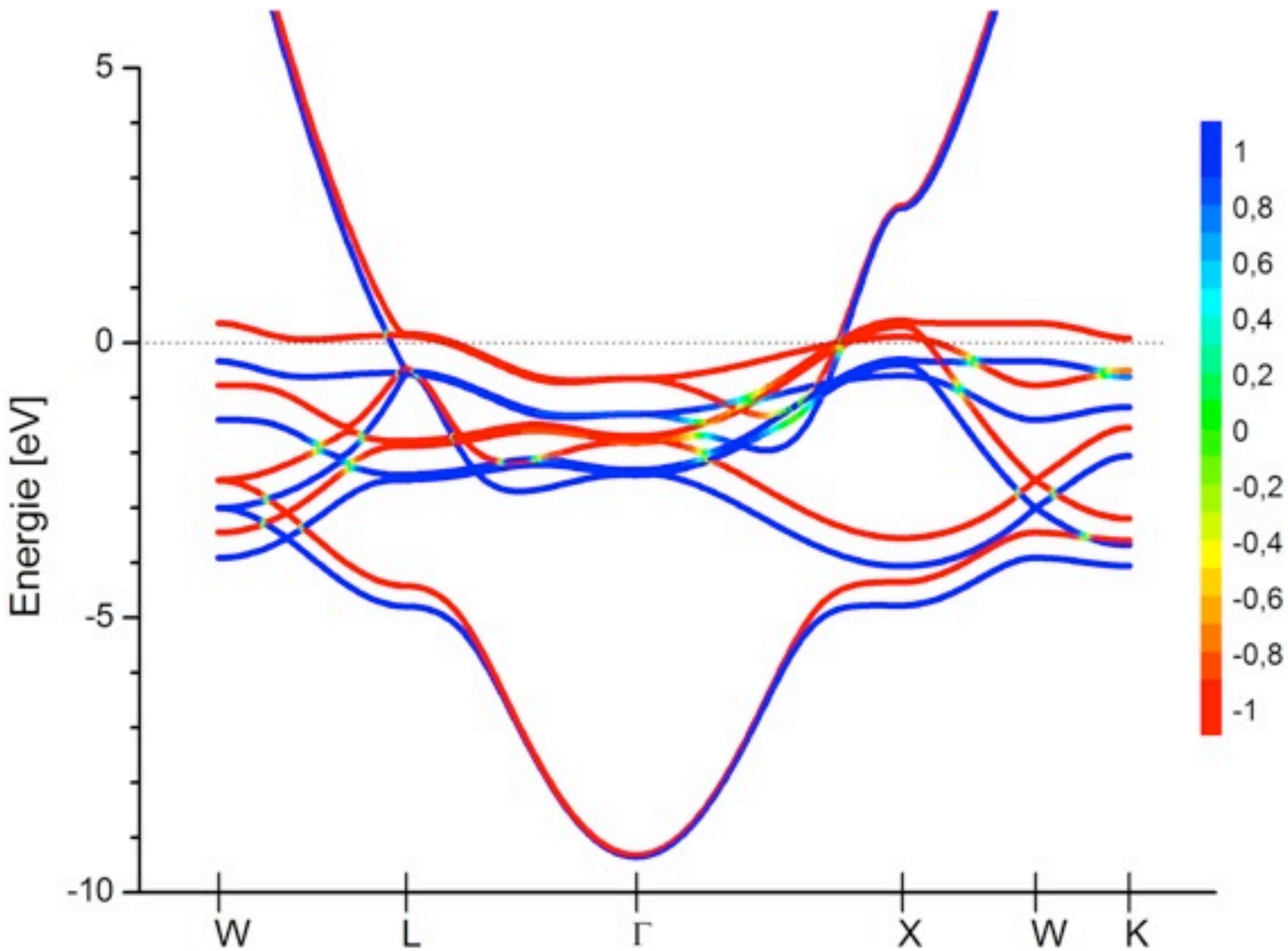
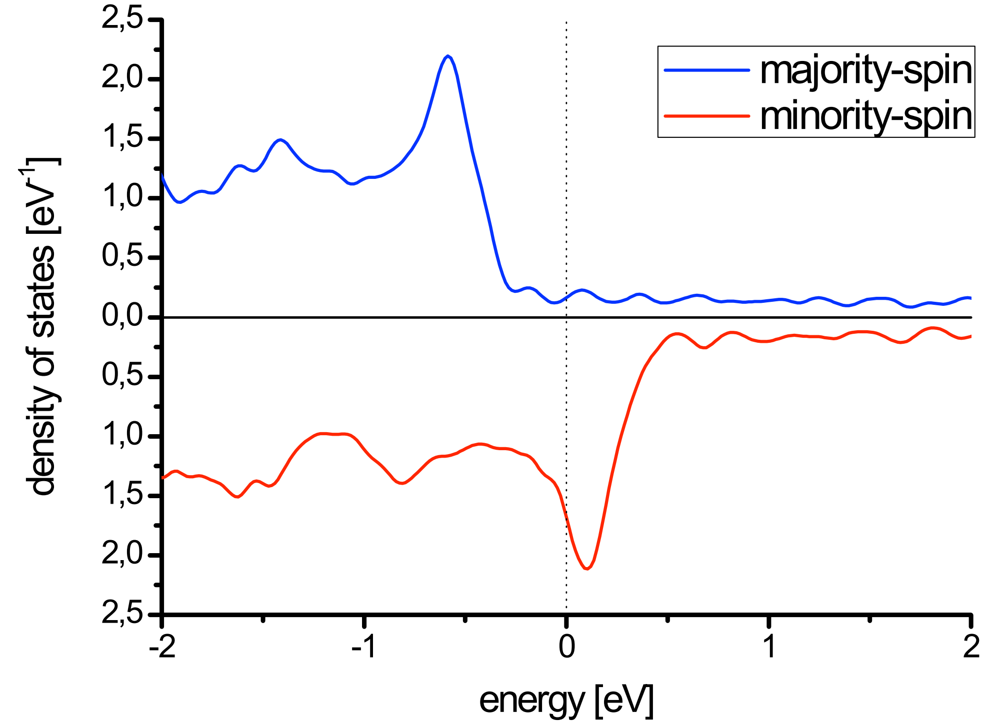
Band Structure Properties

Nickel band structure @ T = 0K
with "spin hot-spots"



Nickel Density of States
@ T=0K



- Demagnetization requires energy (delivered by pulse)
- <u>Any</u> scattering process = dynamical redistribution of excited carriers

How Accurate Can Scattering in a Fixed Band Structure Be?

- Minimal magnetization (maximal demagnetization) by “optimization” for the energy deposited by laser pulse in a fixed band structure

$$\min_{\{n^{\mu}_{\vec{k}}:0\leq n^{\mu}_{\vec{k}}\leq 1\}} \sum_{\vec{k}}\sum_{\mu} n^{\mu}_{\vec{k}} \langle S_z\rangle^{\mu}_{\vec{k}}$$

- Constraints

$$\sum_{\vec{k}}\sum_{\mu} n^{\mu}_{\vec{k}} = N_{\mathrm{eq}}$$

$$\sum_{\vec{k}}\sum_{\mu} n^{\mu}_{\vec{k}}\epsilon^{\mu}_{\vec{k}} \leq E_{\mathrm{eq}} + \Delta E$$

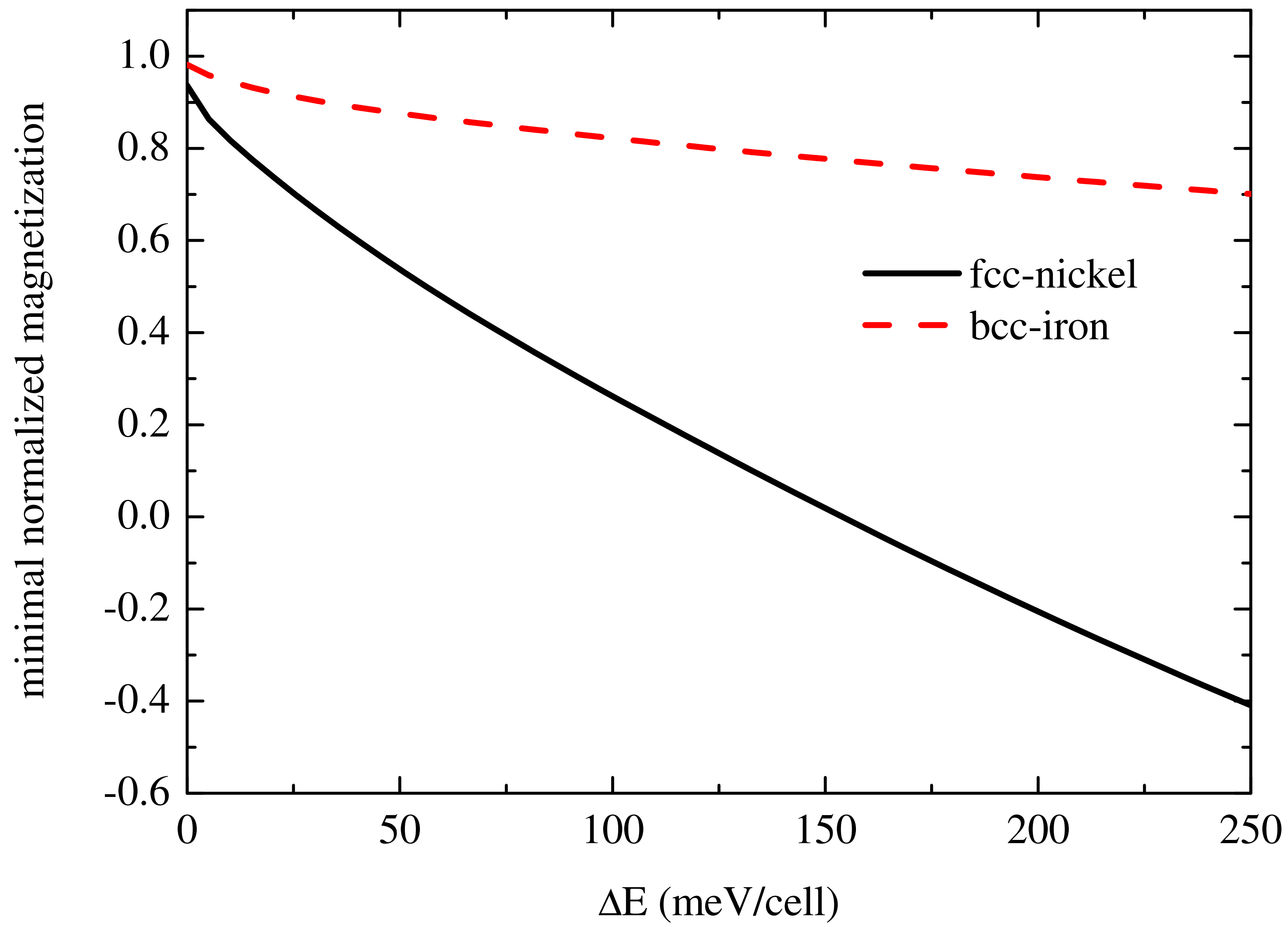
- Deposited energy

$$\Delta E = \int_{300\,\mathrm{K}}^{T(5\,\mathrm{ps})} dT C_{\mathrm{p}}(T)$$

Essert & Schneider, Phys. Rev. B **84**, 224405 (2011)

How Accurate Can Scattering in a Fixed Band Structure Be?

- Minimal magnetization (maximal demagnetization) by “optimization” for the energy deposited by laser pulse in a <u>fixed</u> <u>band structure</u>



Essert & Schneider, Phys. Rev. B **84**, 224405 (2011)

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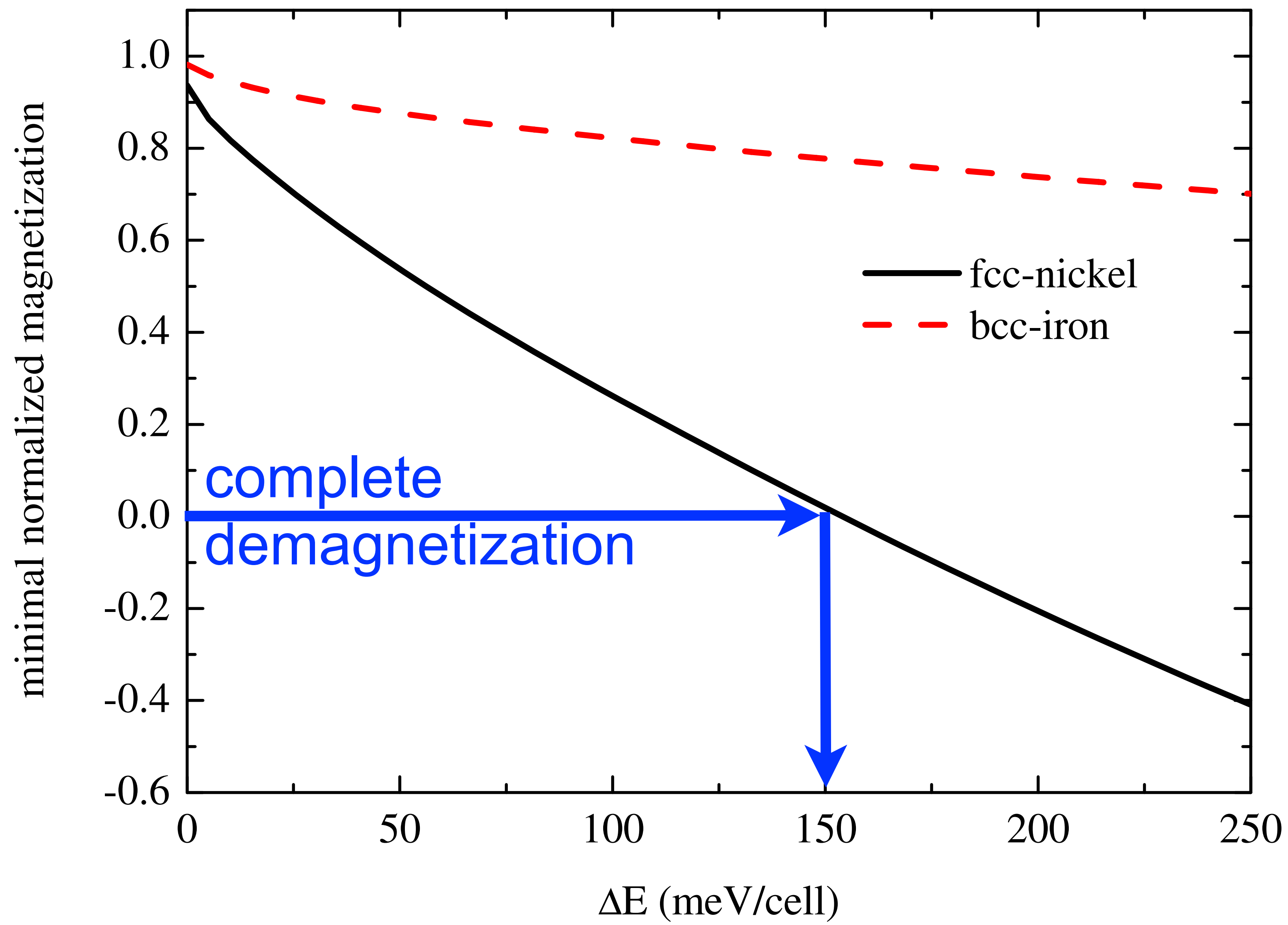

Essert & Schneider, Phys. Rev. B **84**, 224405 (2011)

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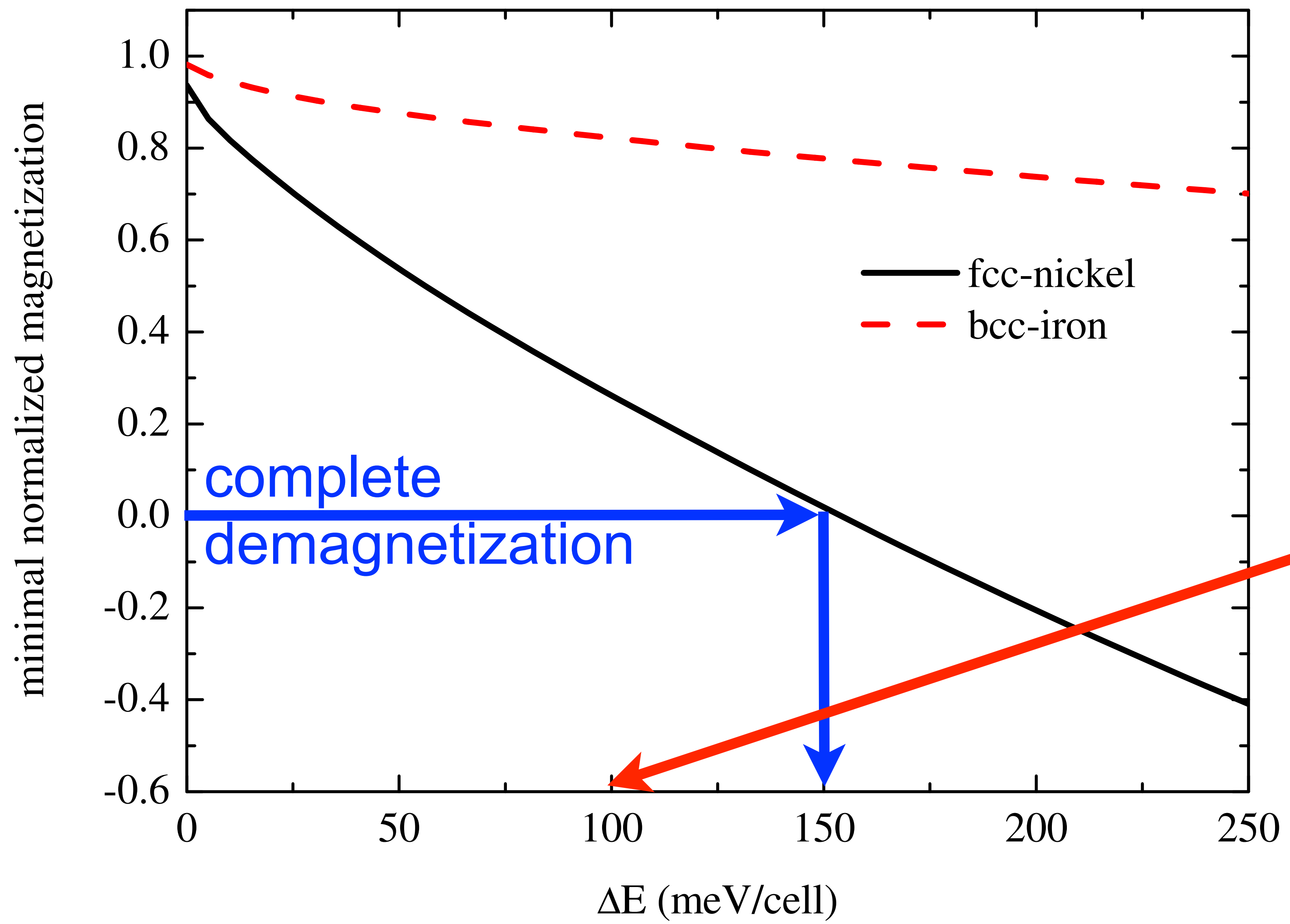


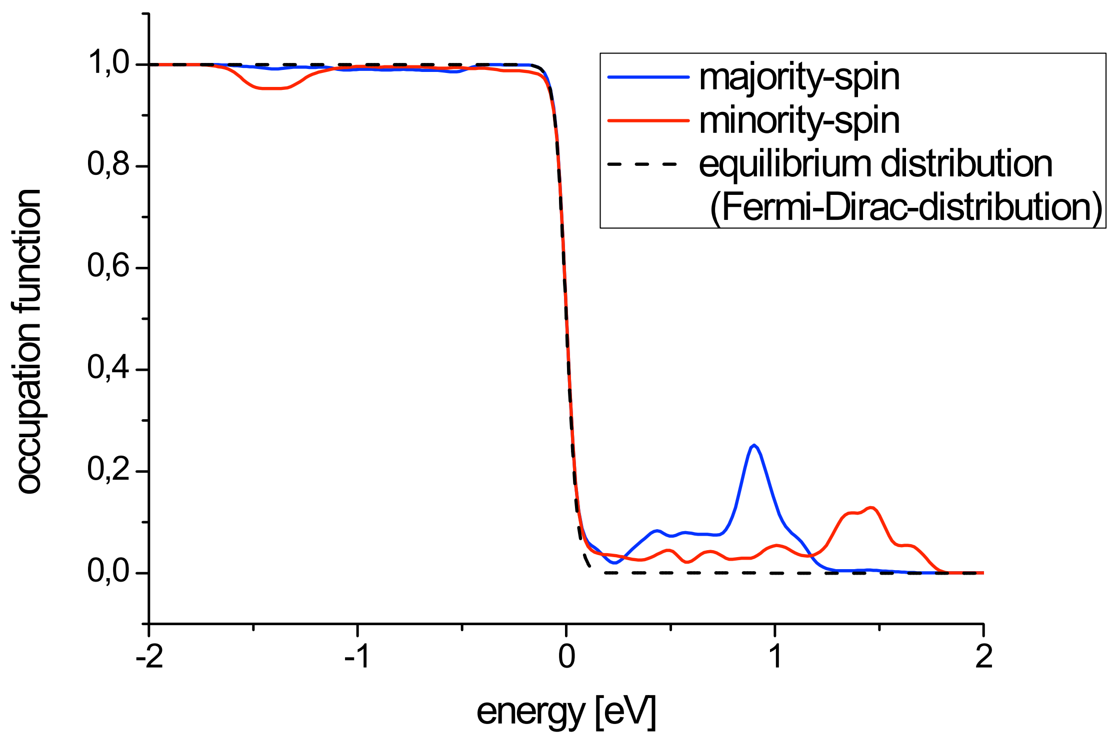
Essert & Schneider, Phys. Rev. B **84**, 224405 (2011)

ΔE from experiment for complete demagnetization:

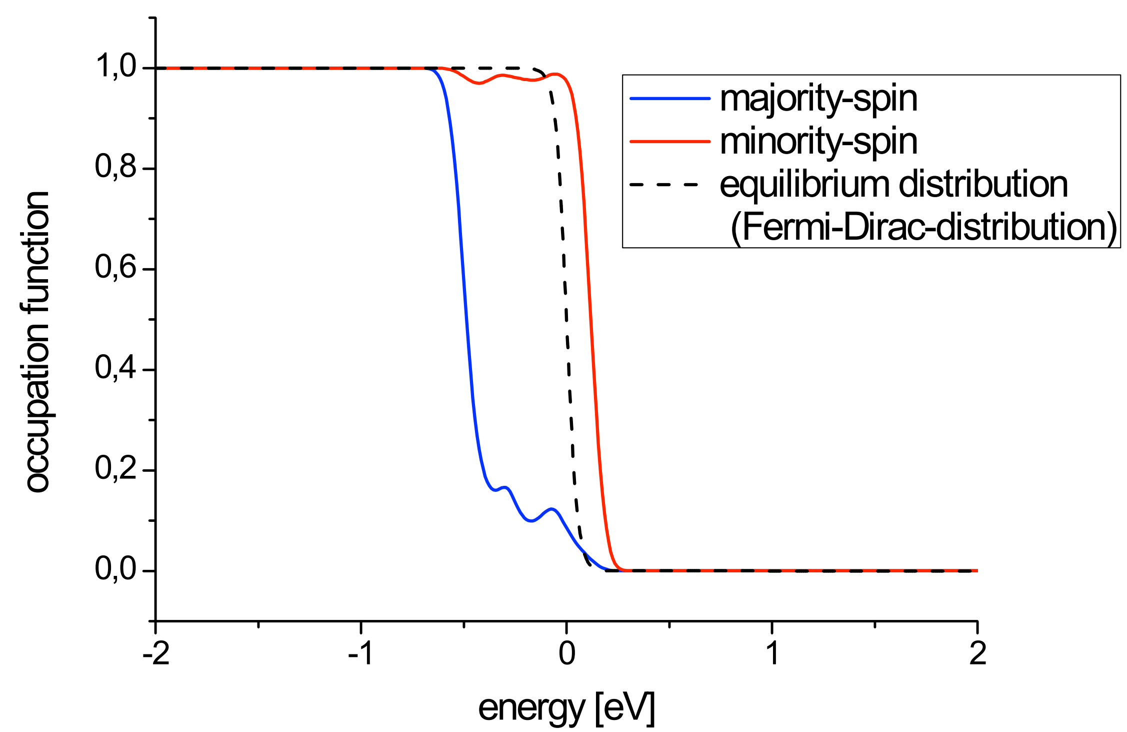
inconsistent with experiment!!!

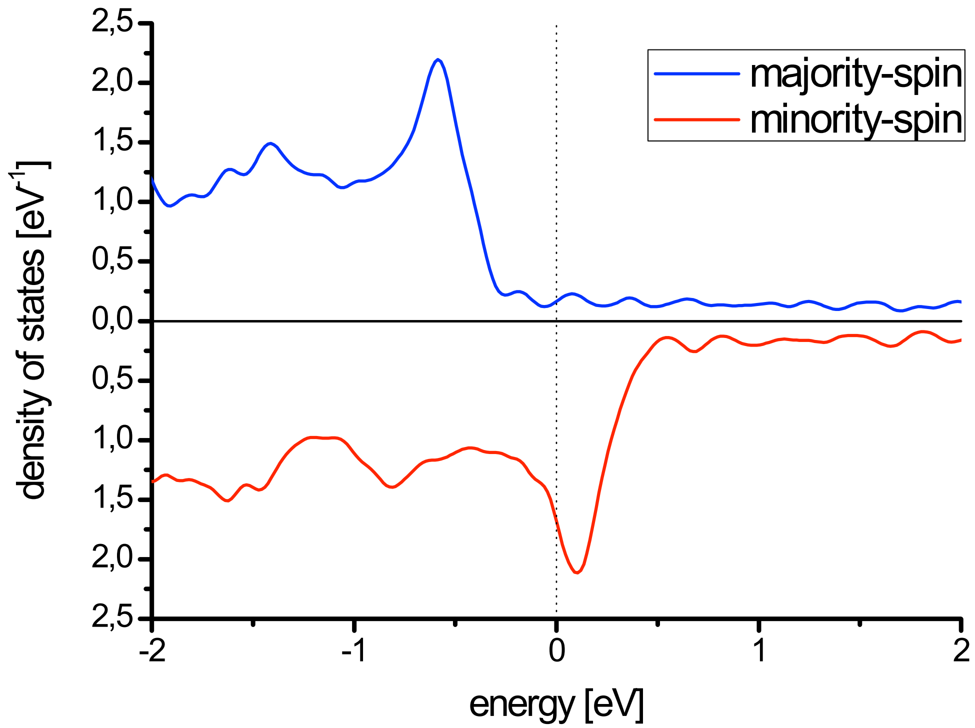
Distribution Functions

after optical excitation



minimal magnetization

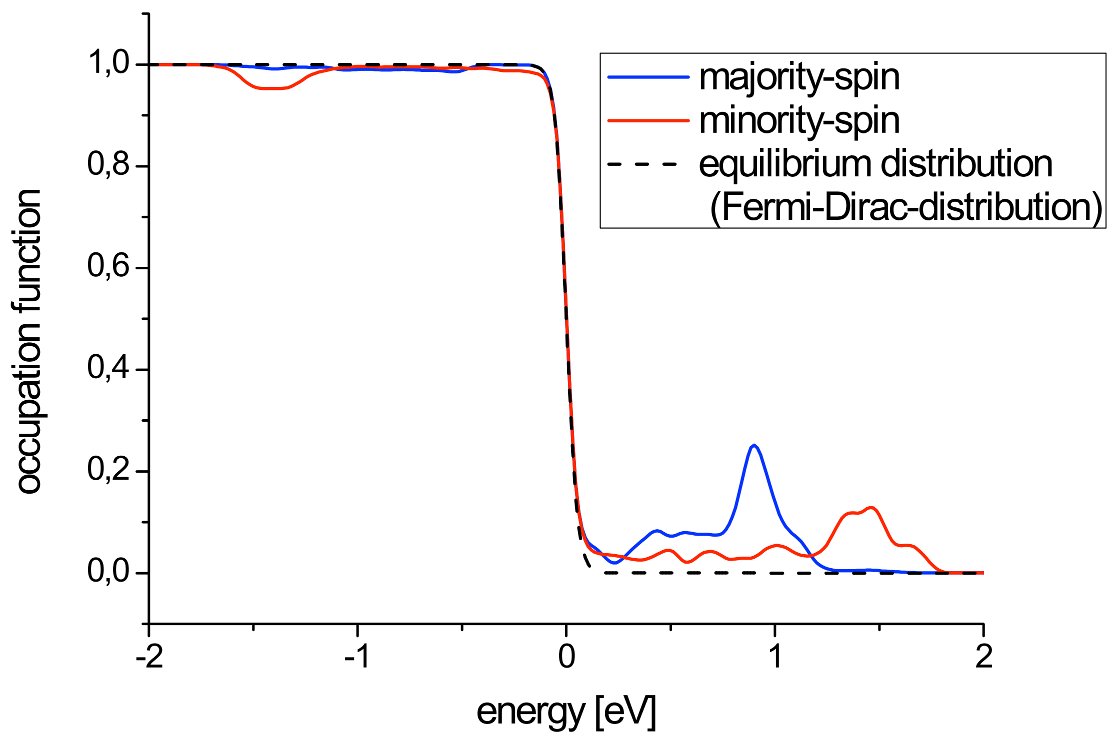




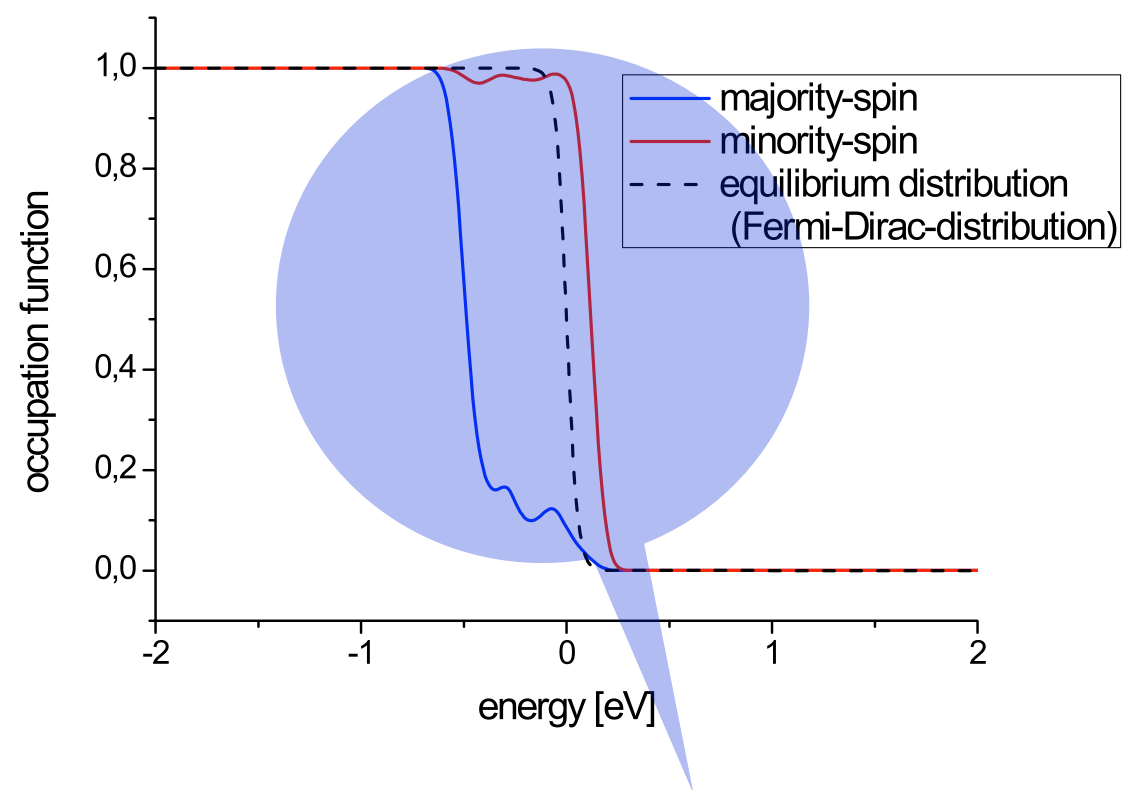
Hans Christian Schneider, TU Kaiserslautern

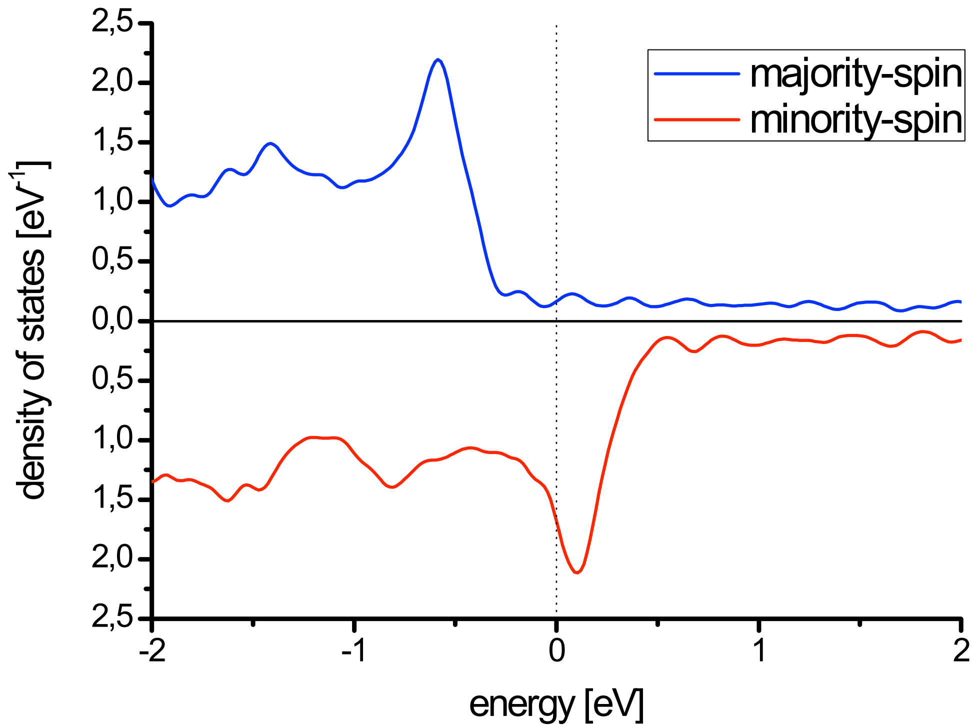
Distribution Functions

after optical excitation



minimal magnetization

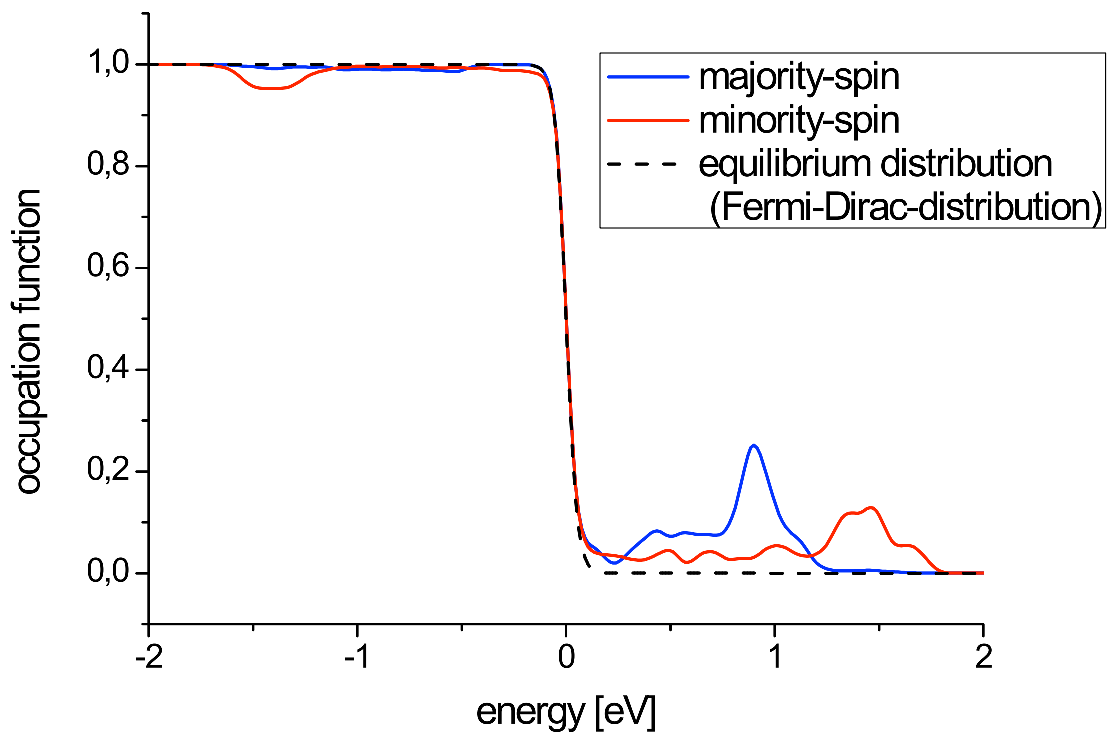




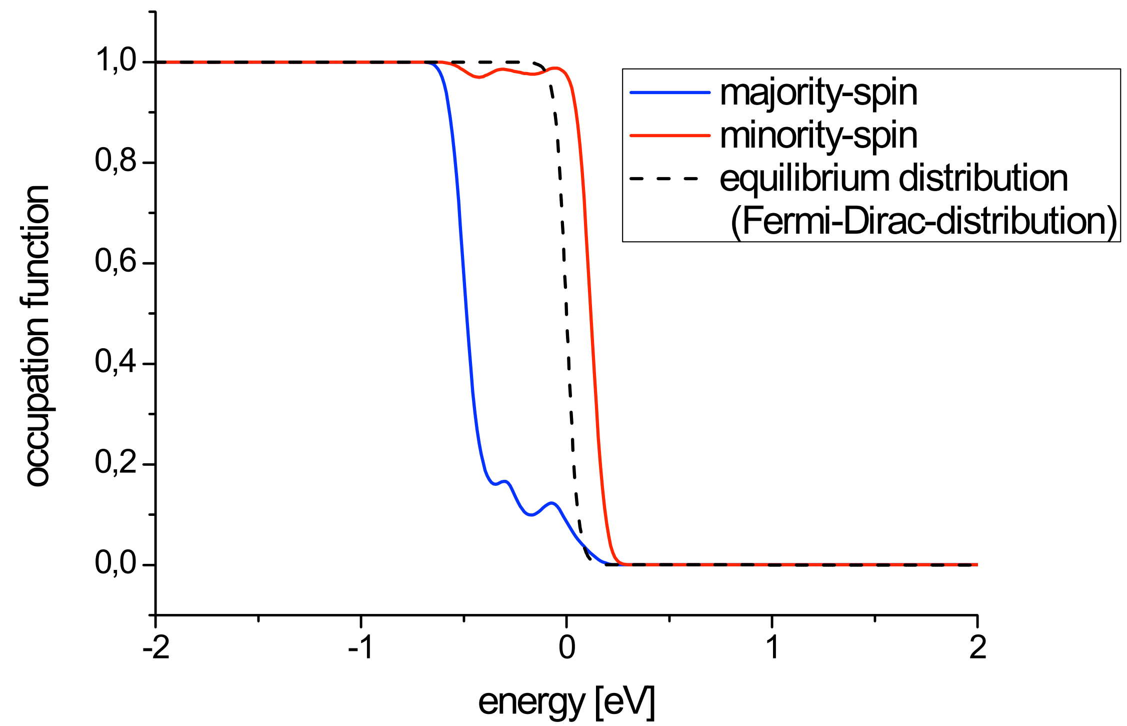
unlikely to be reached by physical scattering processes

Distribution Functions

after optical excitation



minimal magnetization



- Scattering in DFT band structure in general not sufficient to explain demagnetization
- Exchange splitting change/spin fluctuations must occur on ultrafast timescale in addition to scattering

agreement with Carva, Battiato and Oppeneer, PRL **107**, 207201 (2011)

Rhie et al., Phys. Rev. Lett. **90**, 247201 (2003)

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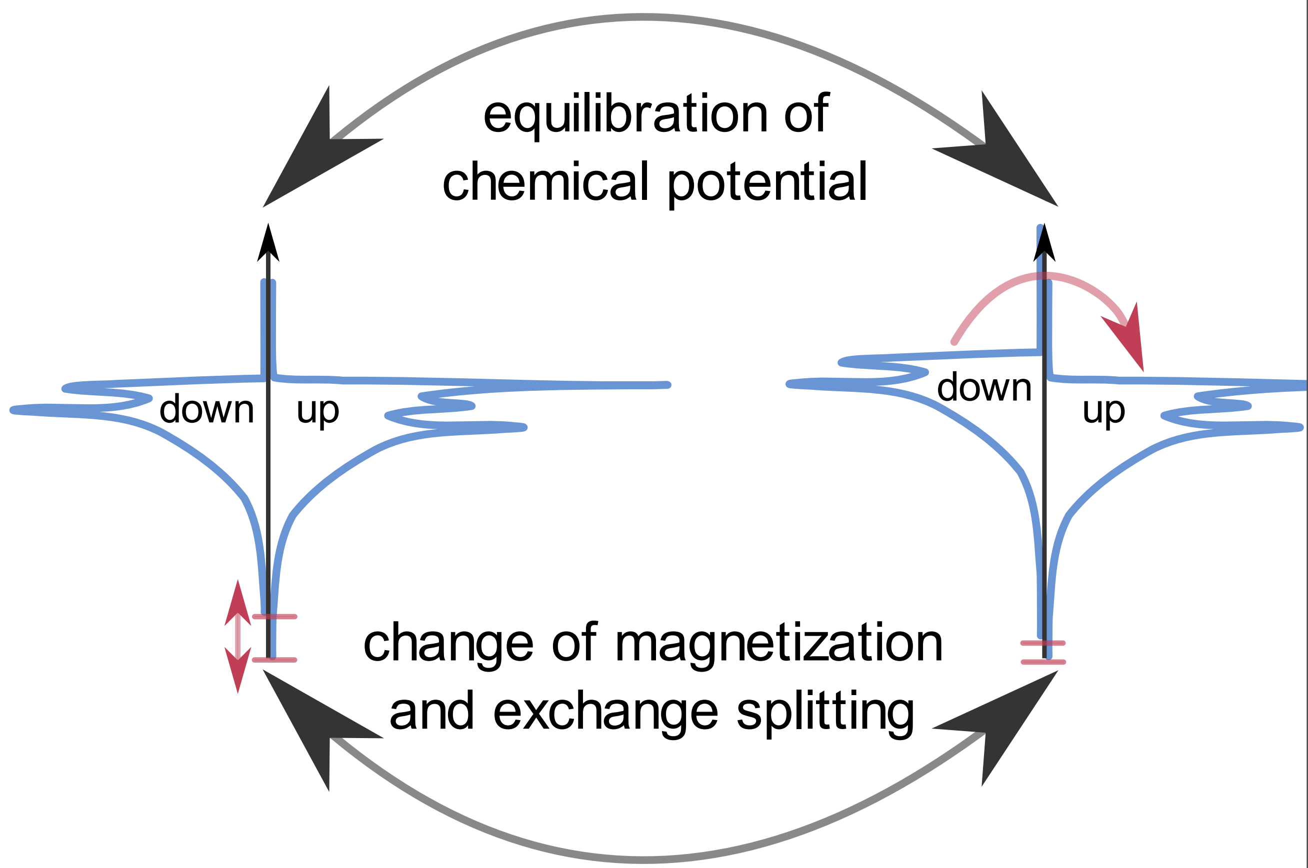
Dynamical Exchange-Splitting: Model

- Stoner Model for exchange splitting $\Delta = U_{\text{eff}}(n_\uparrow - n_\downarrow)$
- spin-dependent DOS

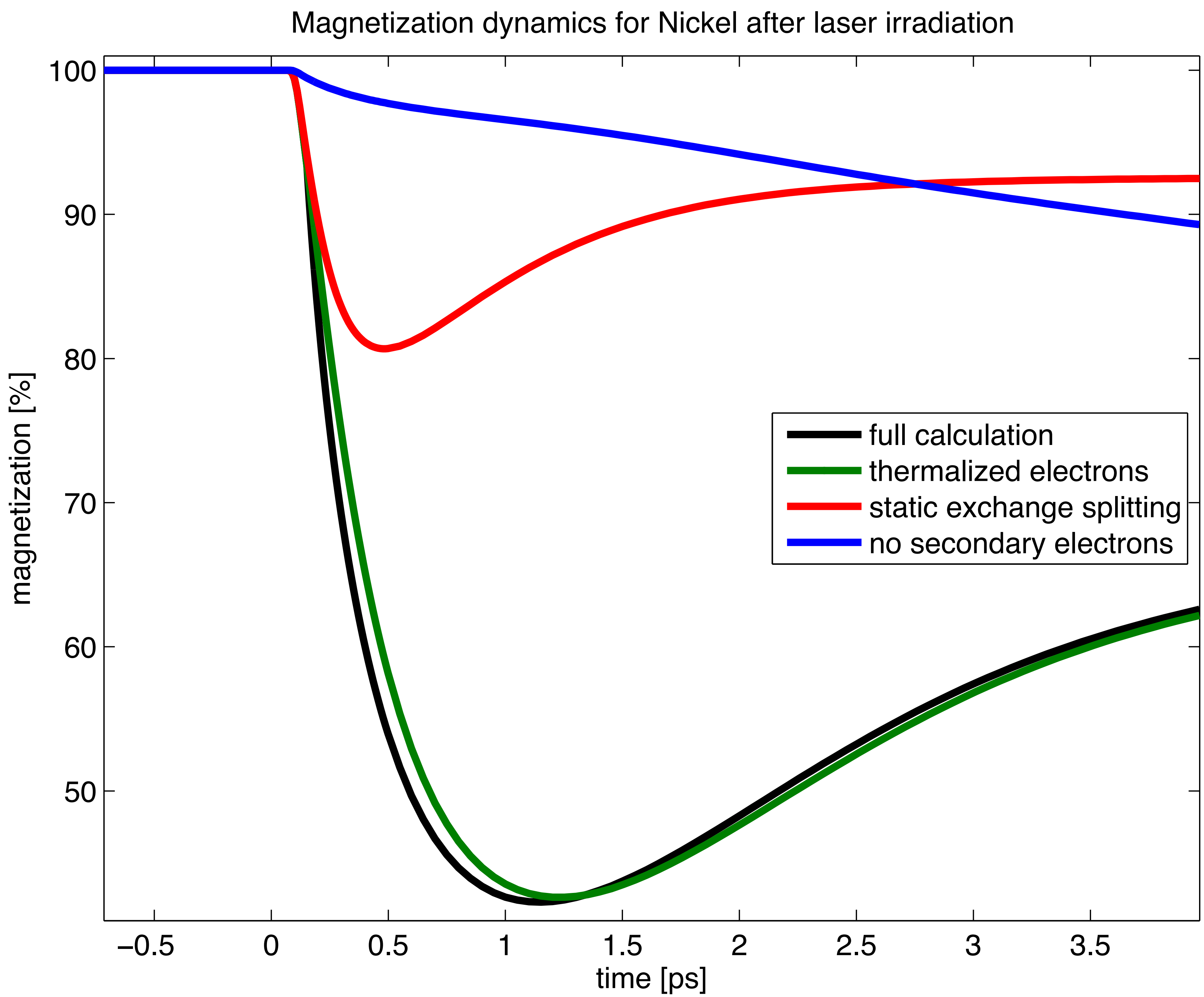
  $\mathcal{D}_\sigma(\epsilon) = \mathcal{D}_\sigma^{(0)}(\epsilon \pm \Delta)$
- electron-electron and electron-phonon scattering
- band structure (spin-orbit interaction, matrix elements, optical excitation) not ab-initio



Dynamics in 2-Band Model

- Compare influence of scattering mechanisms and exchange splitting

- Exchange splitting important
- Electron-electron scattering to a lesser extent



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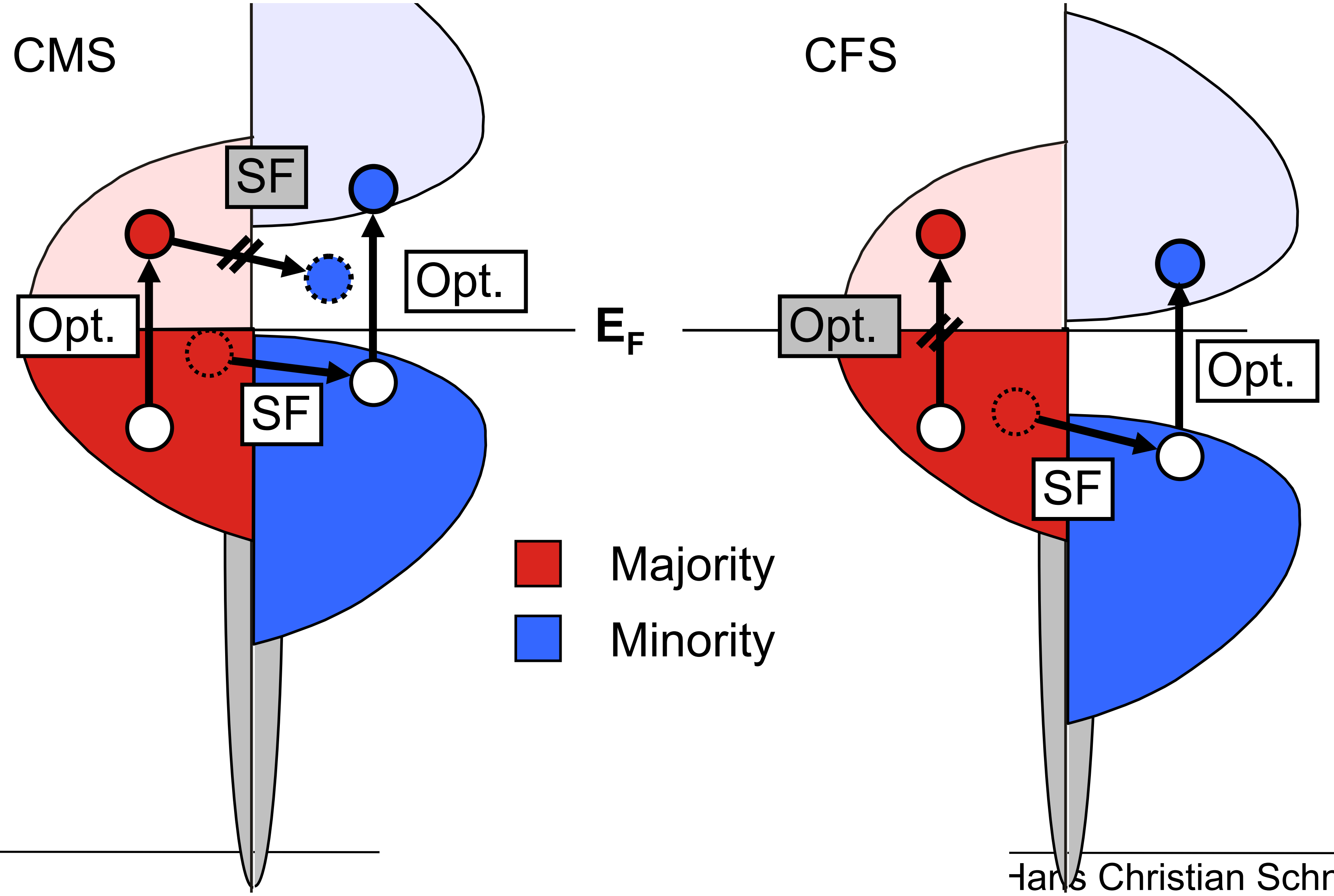
Heusler Alloys: Generalities

- Composition X_2YZ (X,Y = transition metals; Z = main group element)
- Here: Co_2MnSi (CMS) and Co_2FeSi (CFS)

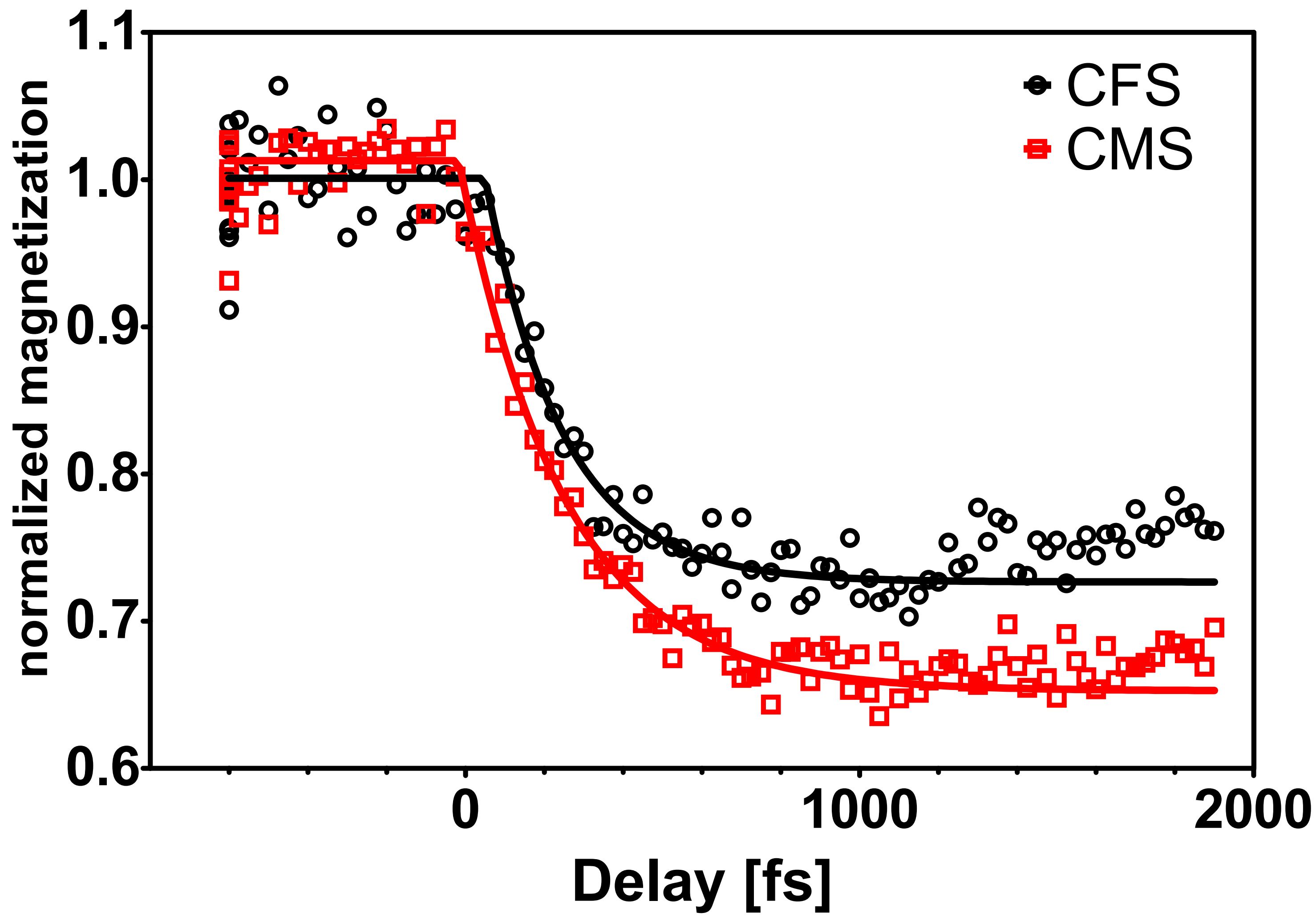
- Band structure engineering: $Co_2Mn_{1-x}Fe_xSi$
- Half-metals ("tunable" gap in minority bands @ Fermi energy): Materials with high spin polarization
- Exact determination of half-metallicity via theory/ experiment difficult

Heusler Alloys: CMS and CFS

- Half metals with different line-up of gap in minority channel
- CMS: “minority-state blocking” = no empty minority-spin states for spin-flip transitions above Fermi energy
- CFS: empty minority-spin states available at the Fermi energy
- Expect different demagnetization after optical excitation



Demagnetization Dynamics for CFS and CMS

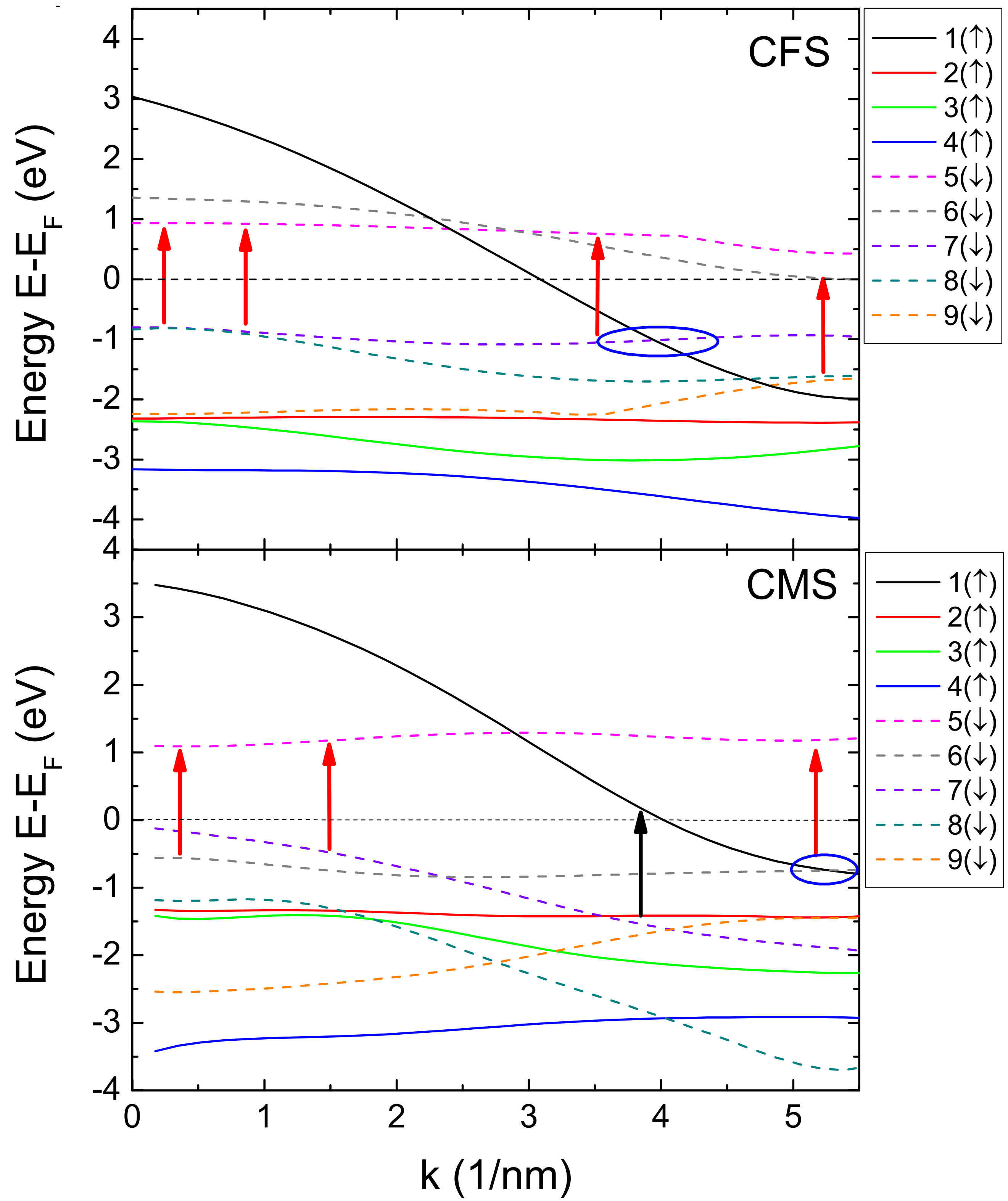


- epitaxial CFS, CMS samples
- optical excitation @800 nm, 50 fs pulses
- τ_M = 198 fs (CFS)
- τ_M = 256 fs (CMS)

- Experimental MOKE spectra show similar demagnetization dynamics
- No signature of minority-state blocking in CMS
- Possible explanation: defect states in the band gap
- Here: trace scattering pathways in dynamical model

Band Structure and Dynamics

- Band structure in Γ-X direction
- Optical excitation in CFS only in minority channel (no electronic demagnetization!)
- CMS: minority and majority electrons are excited
- Possibility of majority-minority spin transitions below E_F in both cases

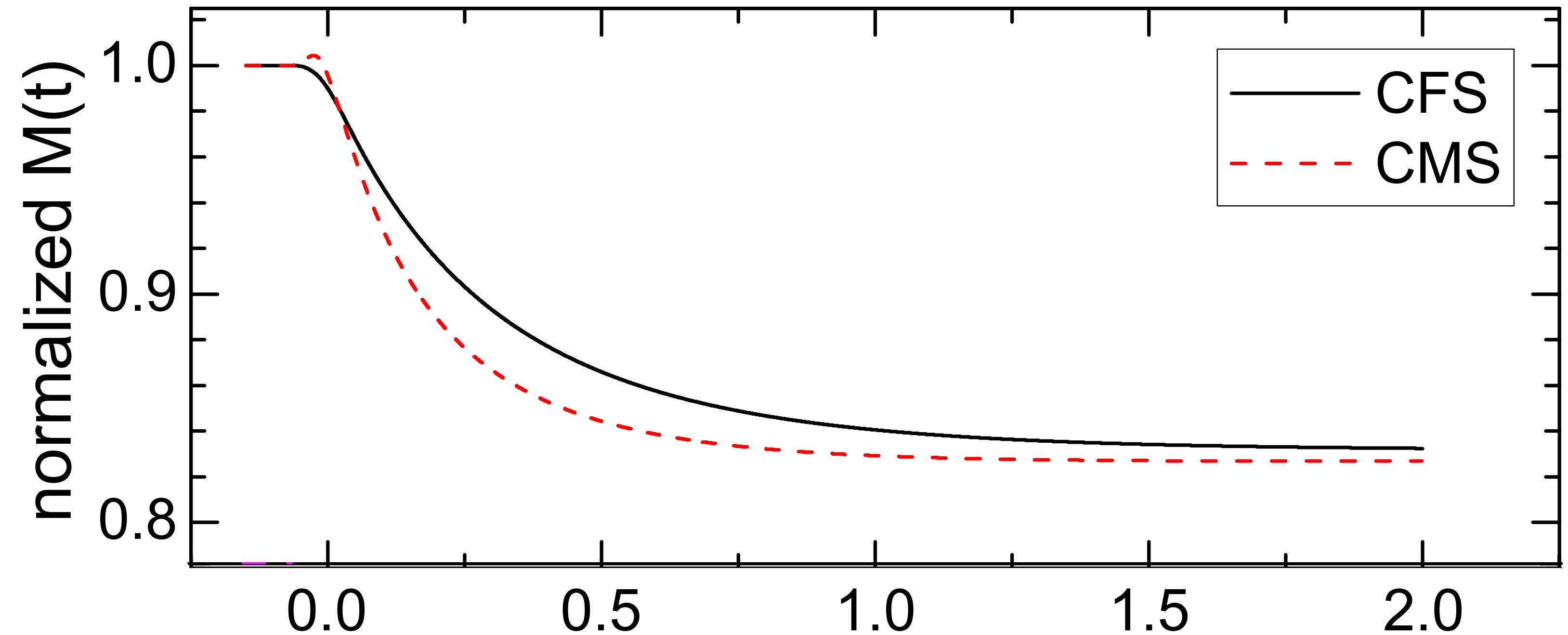


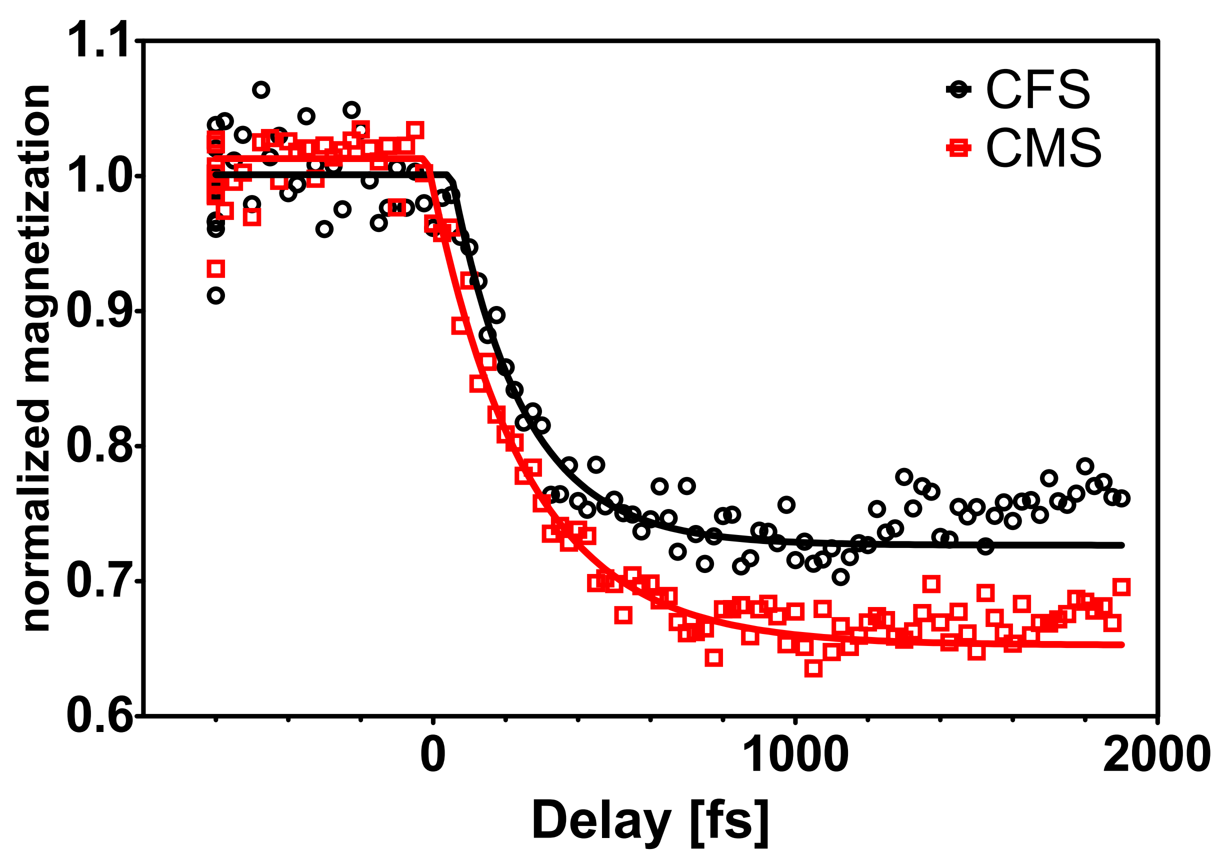
Steil, D. *et al.* B *Phys. Rev. Lett.* **105**, 217202 (2010).

Krauß, M. *et al.*. *Phys. Rev. B* **80**, 180407 (2009).

Calculated Demagnetization Dynamics

- Good agreement with experiment for time constants
- “Quenching” somewhat different





Steil, D. *et al.* B *Phys. Rev. Lett.* **105**, 217202 (2010).

Krauß, M. *et al.*. *Phys. Rev. B* **80**, 180407 (2009).

Conclusions (1)

- Dynamical calculation of momentum resolved distribution functions w/ excitation and Boltzmann scattering integrals including spin-orbit interaction
- DFT (T = 0K) band structure and electron-phonon coupling matrix elements including electron-phonon scattering <u>OR</u>
- Simplified band structure (DOS) and more (bands/scattering mechanisms/dynamical exchange splitting)

- Classical Elliott-Yafet spin-flip scattering occurs mainly for holes!
- In Heusler alloys, this explains the observed characteristics
- In ferromagnets, it cannot explain observed magnetization quenching
- Dynamical exchange splitting (together with electronic redistribution) seems to improve results!

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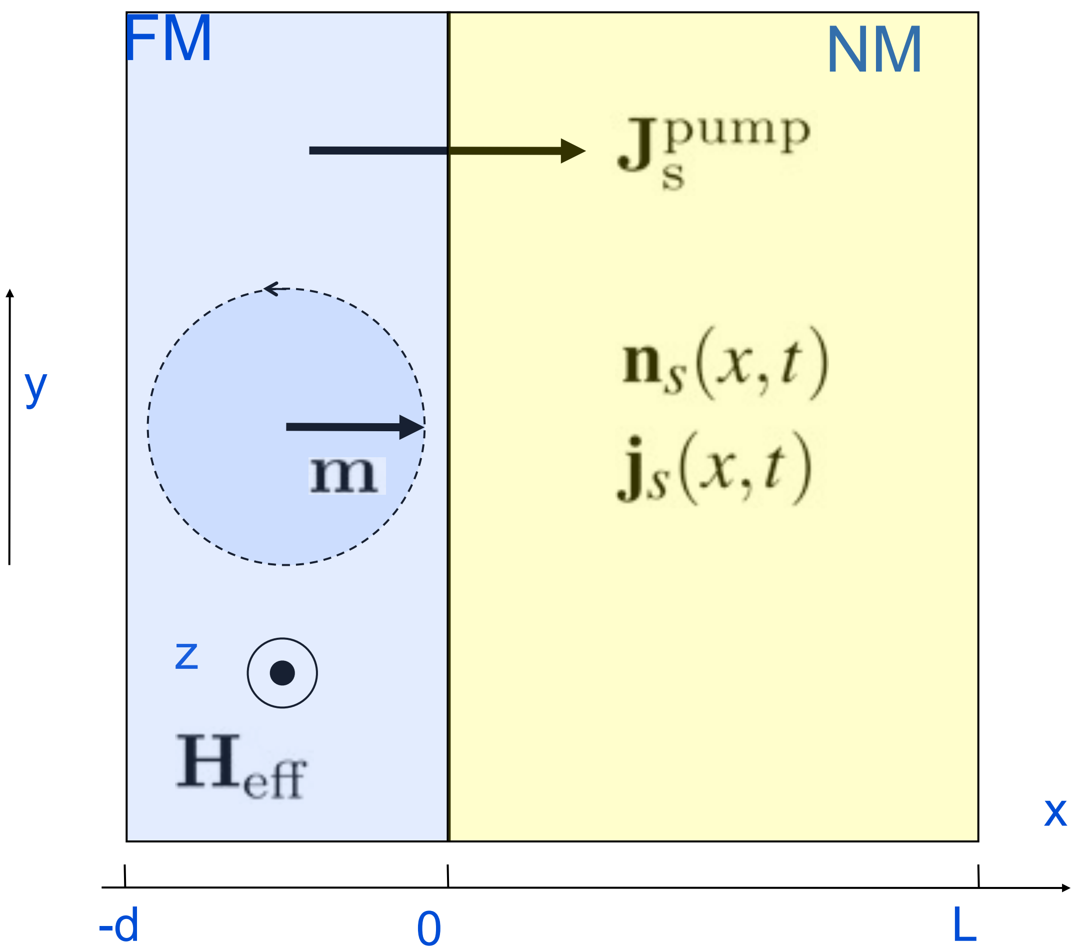
"Experimental" Problems

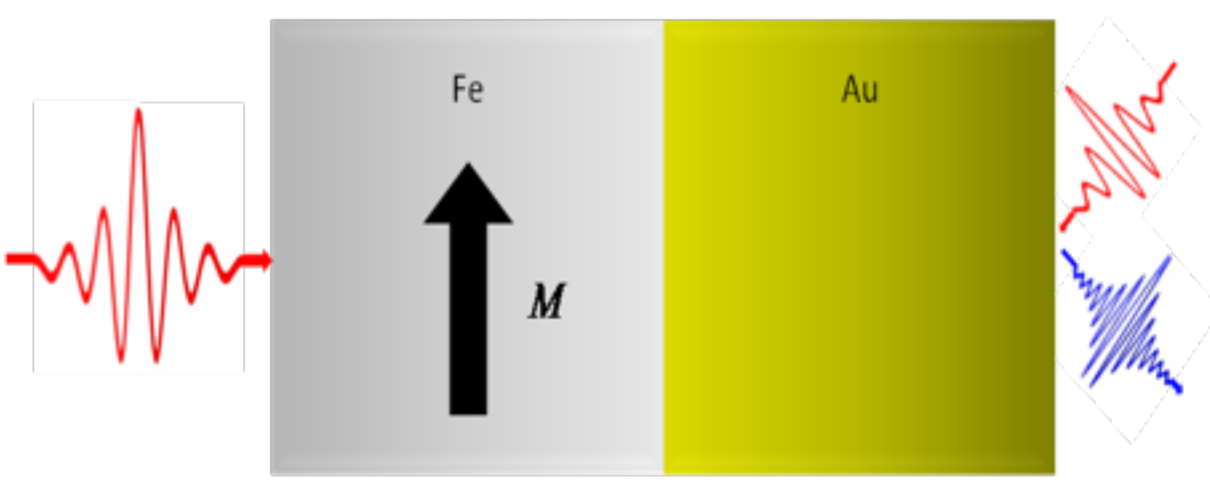
Spin current pumped by FM precessing around magnetic field



$$\vec{j}_s^{\,pump} = \frac{1}{2\pi}\frac{g^{\uparrow\downarrow}}{S}\left[\vec{m}\times\frac{d\vec{m}}{dt}\right]$$

Tserkovnyak et al, Phys. Rev. Lett. **88**, 117601 (2002); Phys. Rev. B **66**, 224403 (2002)

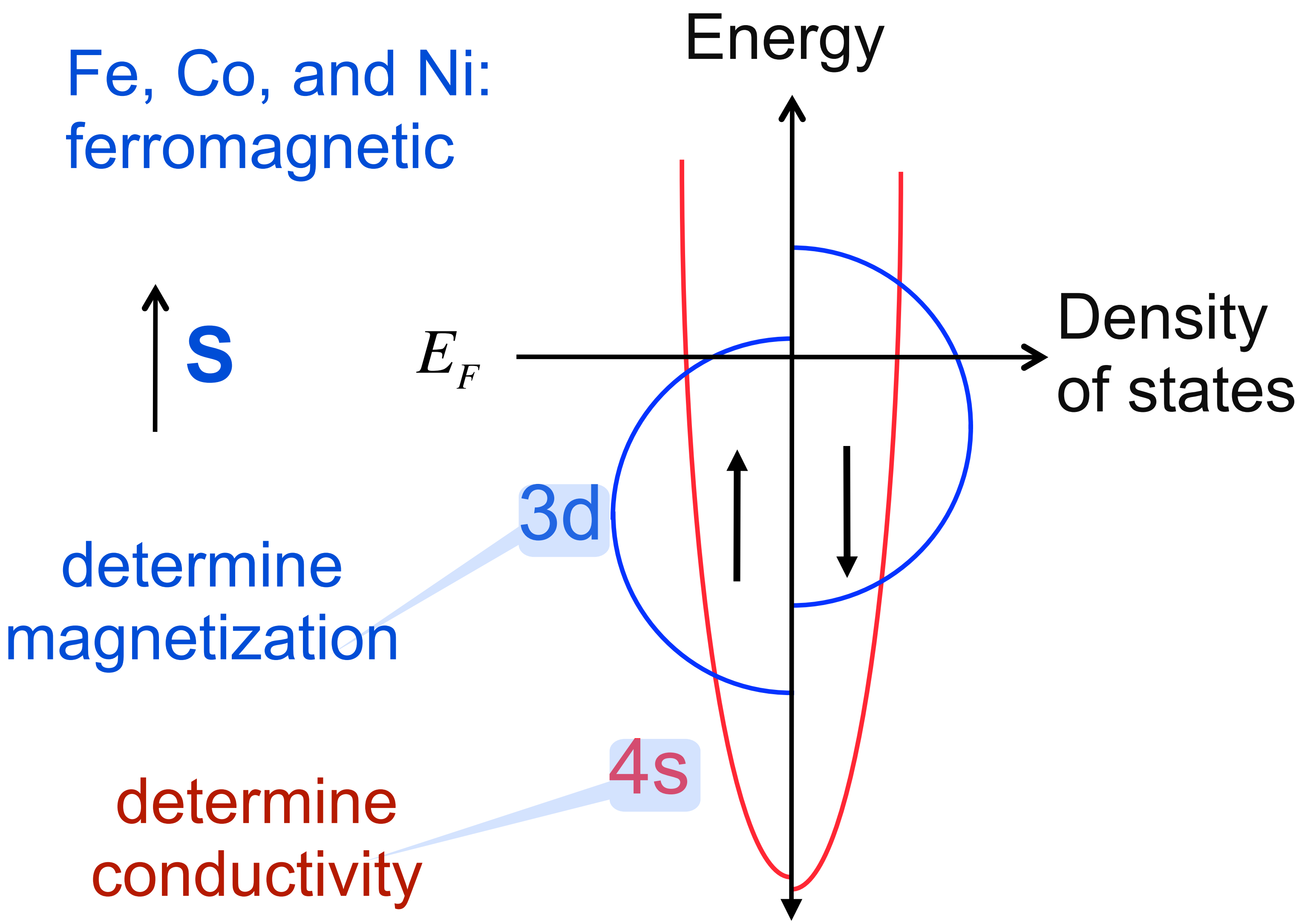
Spin current exited in FM by ultrashort optical pulse



Basics (1): Two-Current Model

- Current carried by “spin-up” and “spin-down” electrons; spin flips rare

Fe, Co, and Ni:
ferromagnetic



Conductivity $\sigma_{\uparrow} > \sigma_{\downarrow}$

N. F. Mott, Proc. Roy. Soc. A **153**, 699 (1936)

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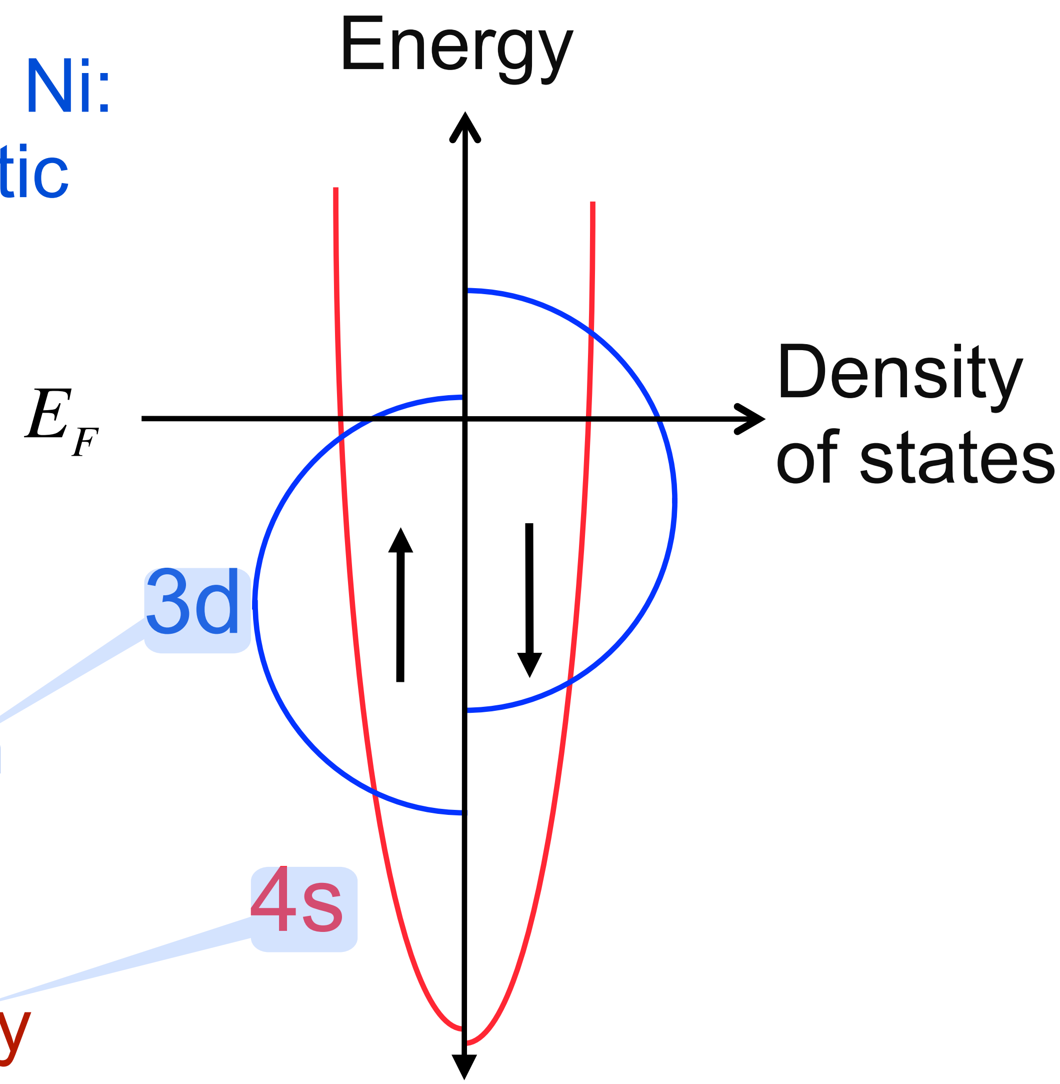
- Current carried by “spin-up” and “spin-down” electrons; spin flips rare

Fe, Co, and Ni: ferromagnetic

↑ **S**

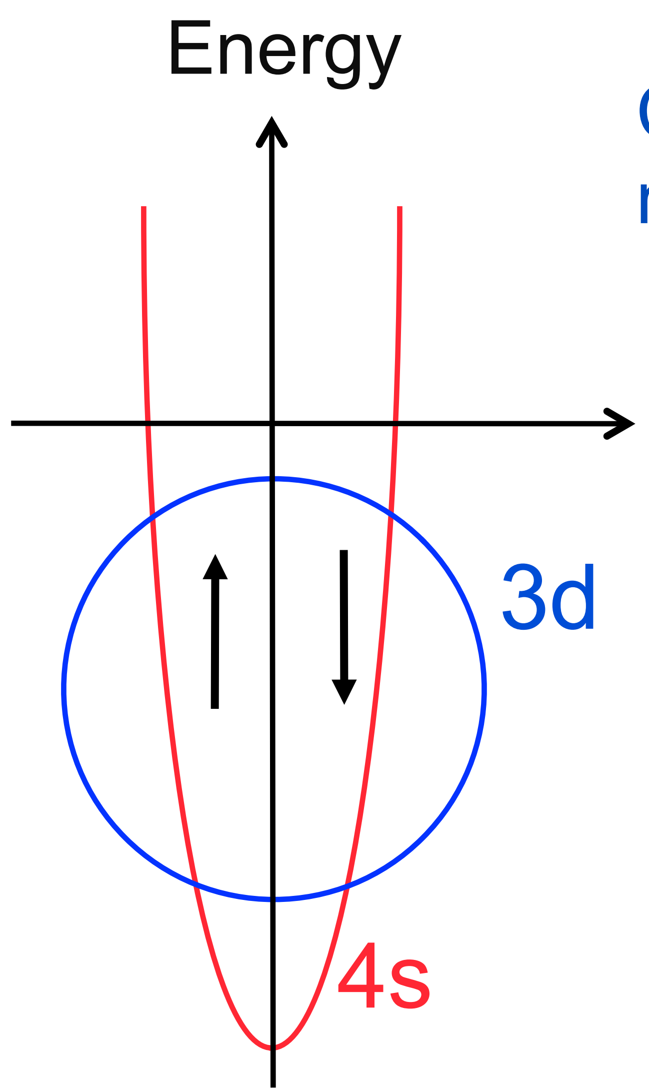


determine magnetization

determine conductivity

Conductivity $\sigma_{\uparrow} > \sigma_{\downarrow}$

Cu: nonmagnetic

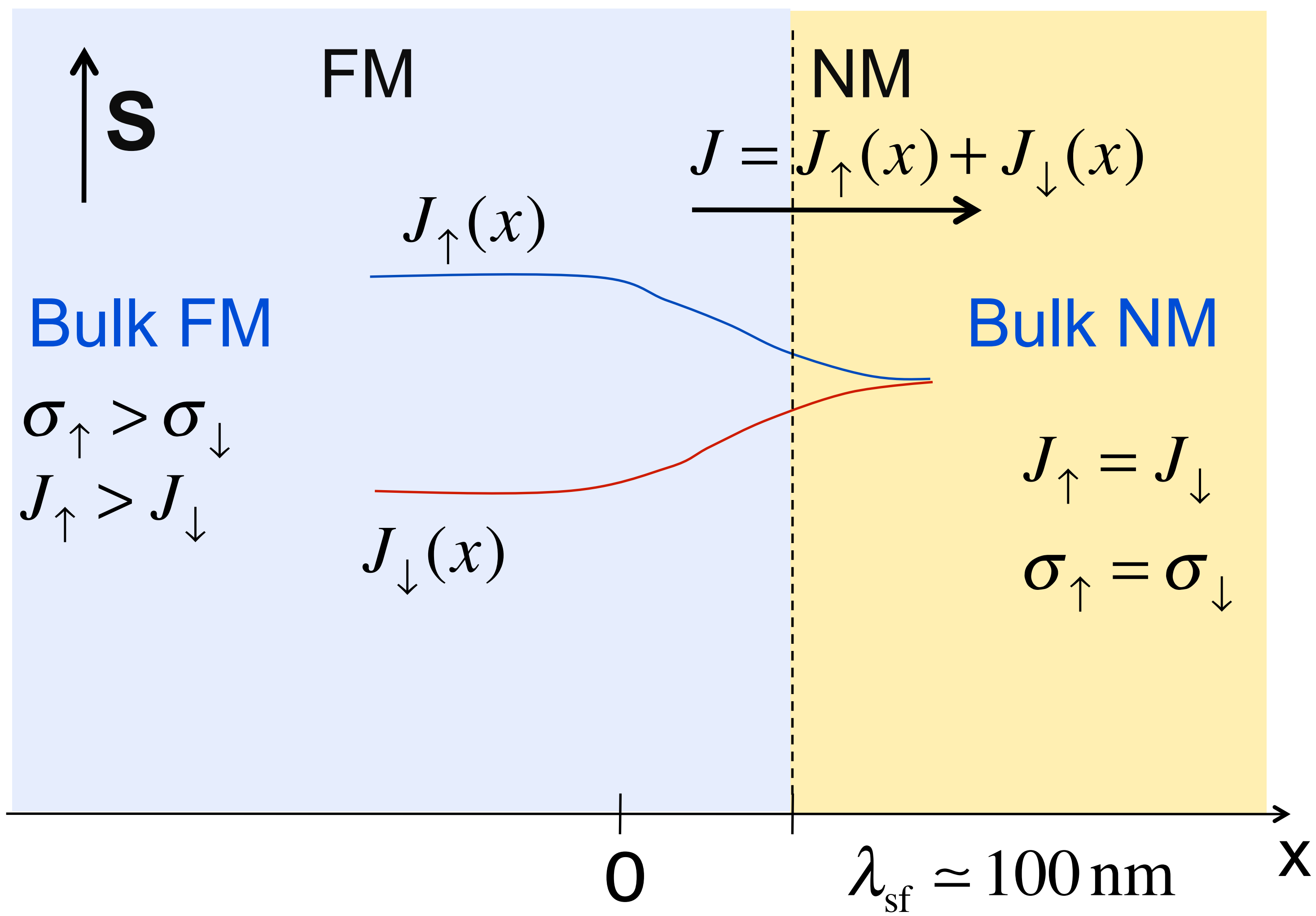


Conductivity $\sigma_{\uparrow} = \sigma_{\downarrow}$

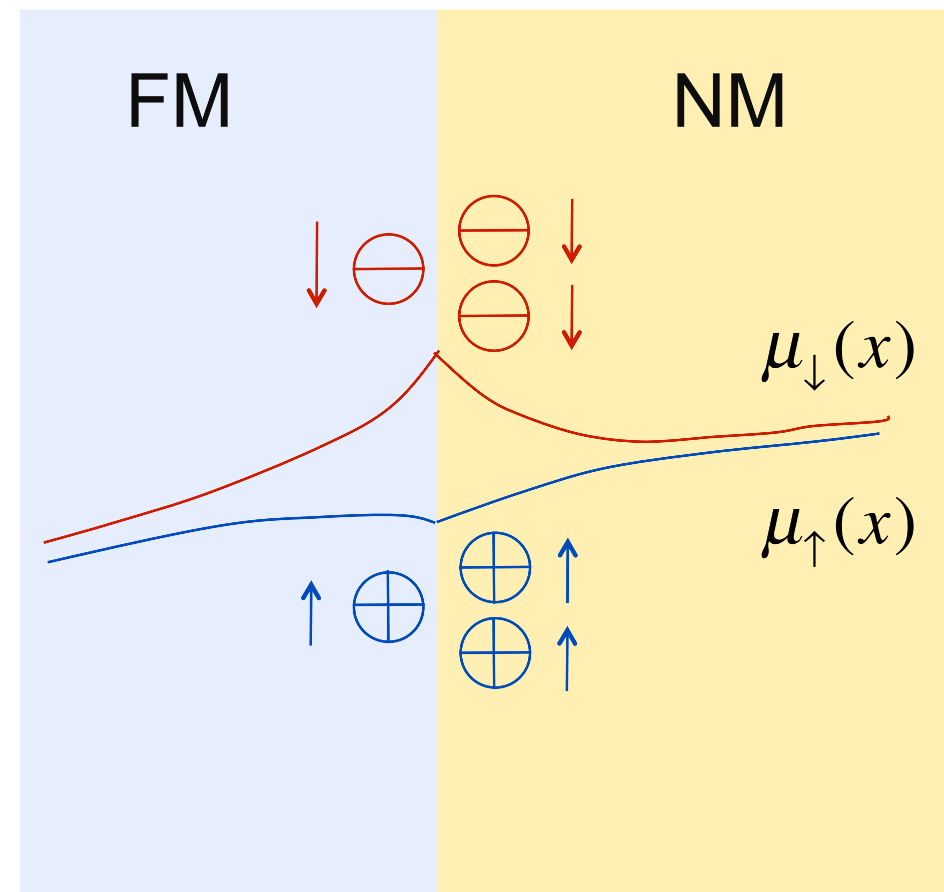
N. F. Mott, Proc. Roy. Soc. A **153**, 699 (1936)

Basics (2): Spin Injection with Current (DC)

- FM-NM junction: collinear magnetization



Electro-chemical potential



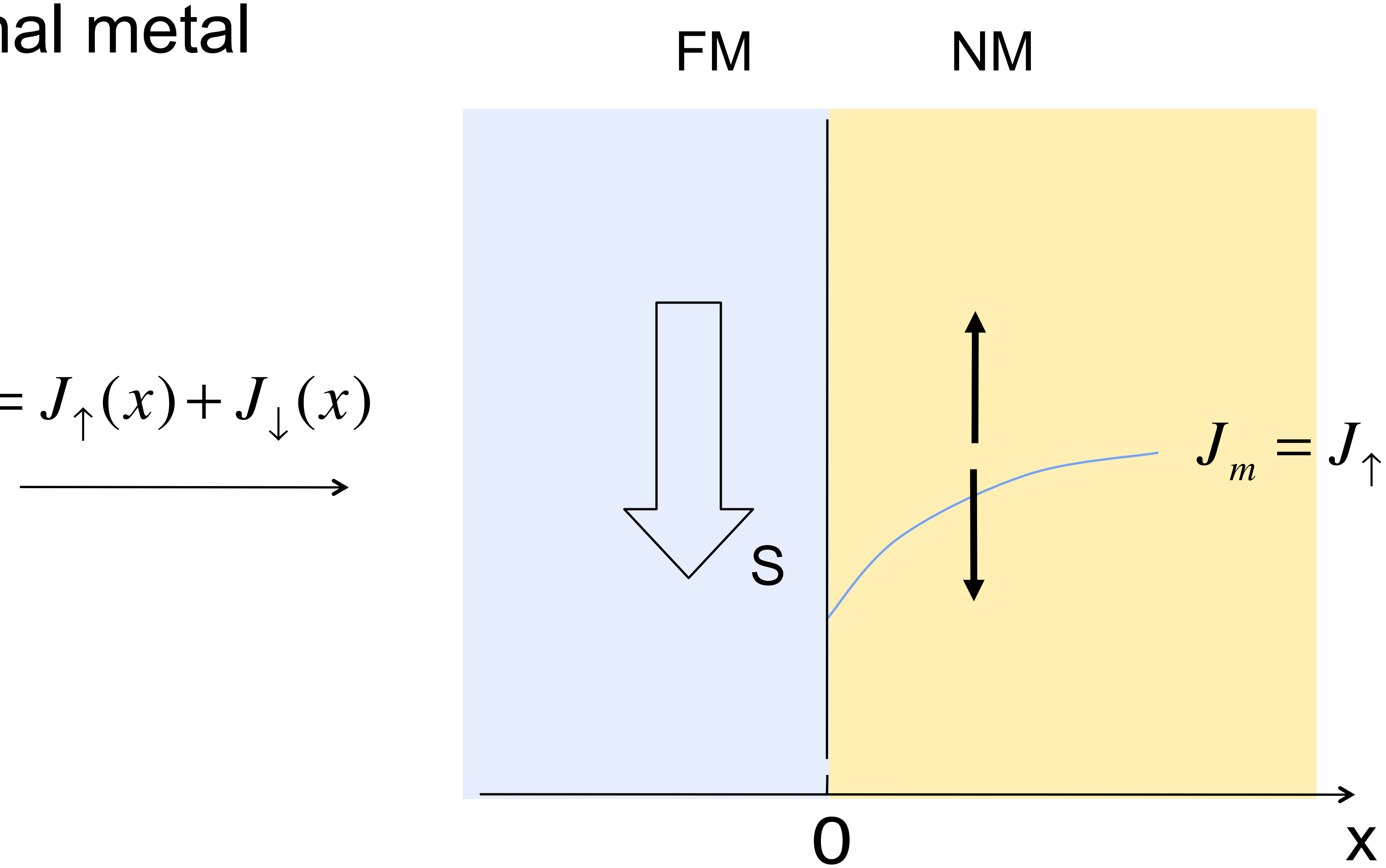
On the length scale of λ_{sf} :
Spin current injected into NM

Spin accumulation (local charge neutrality: metals)

Spin Signal-Propagation

- Spin signal transmitted into normal metal

$$J = J_{\uparrow}(x) + J_{\downarrow}(x)$$



$$J_m = J_{\uparrow}(x) - J_{\downarrow}(x)$$

Spin Signal-Propagation

- Spin signal transmitted into normal metal

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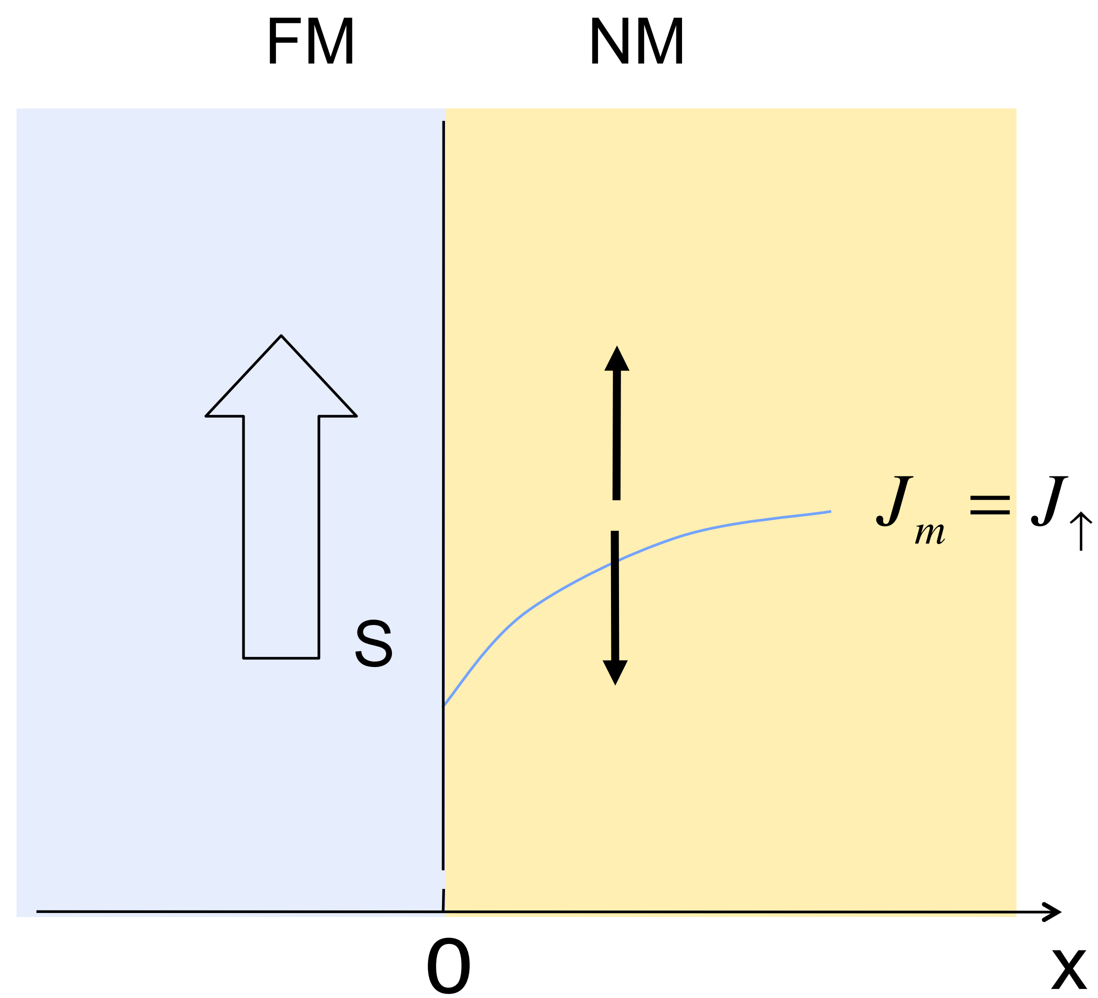

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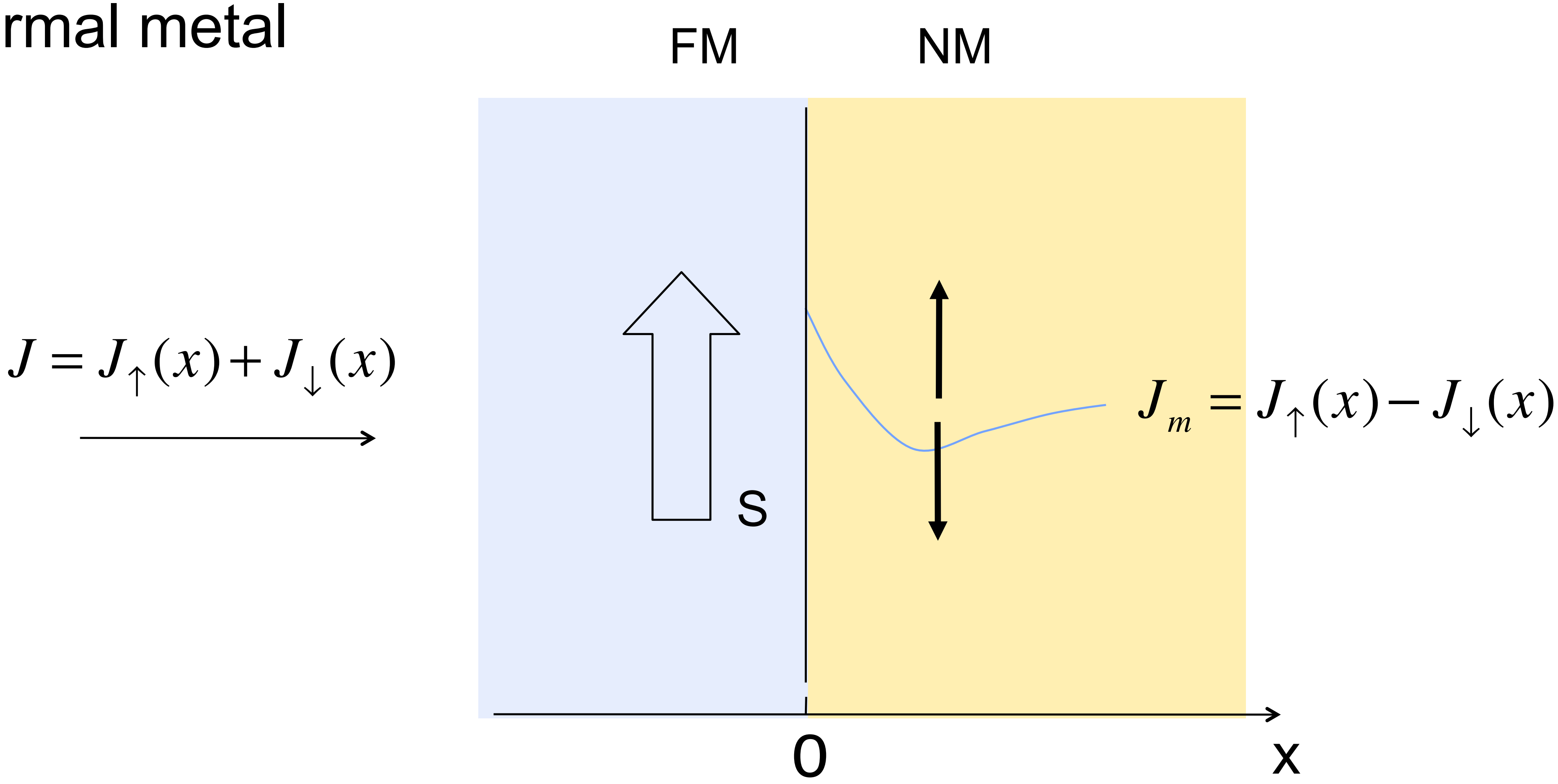

Spin Signal-Propagation

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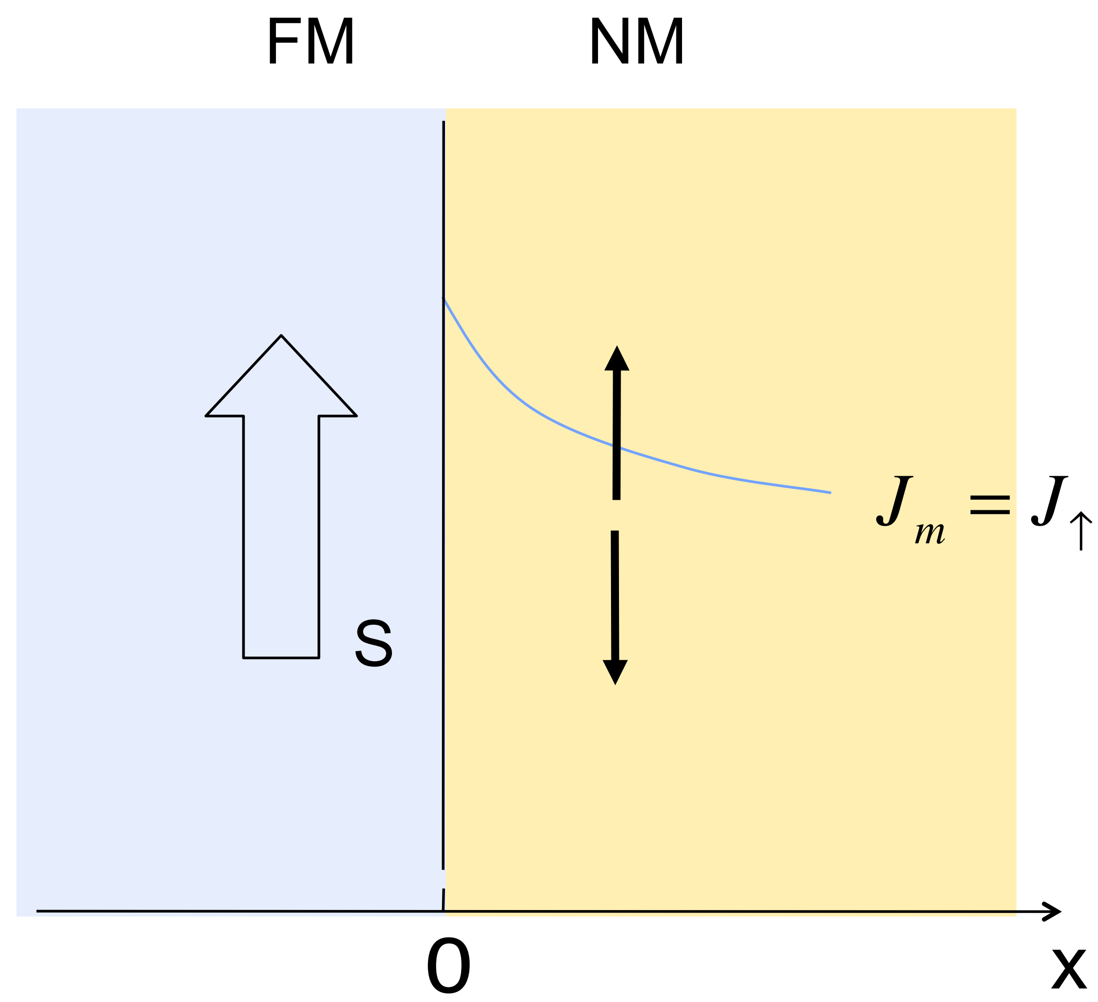

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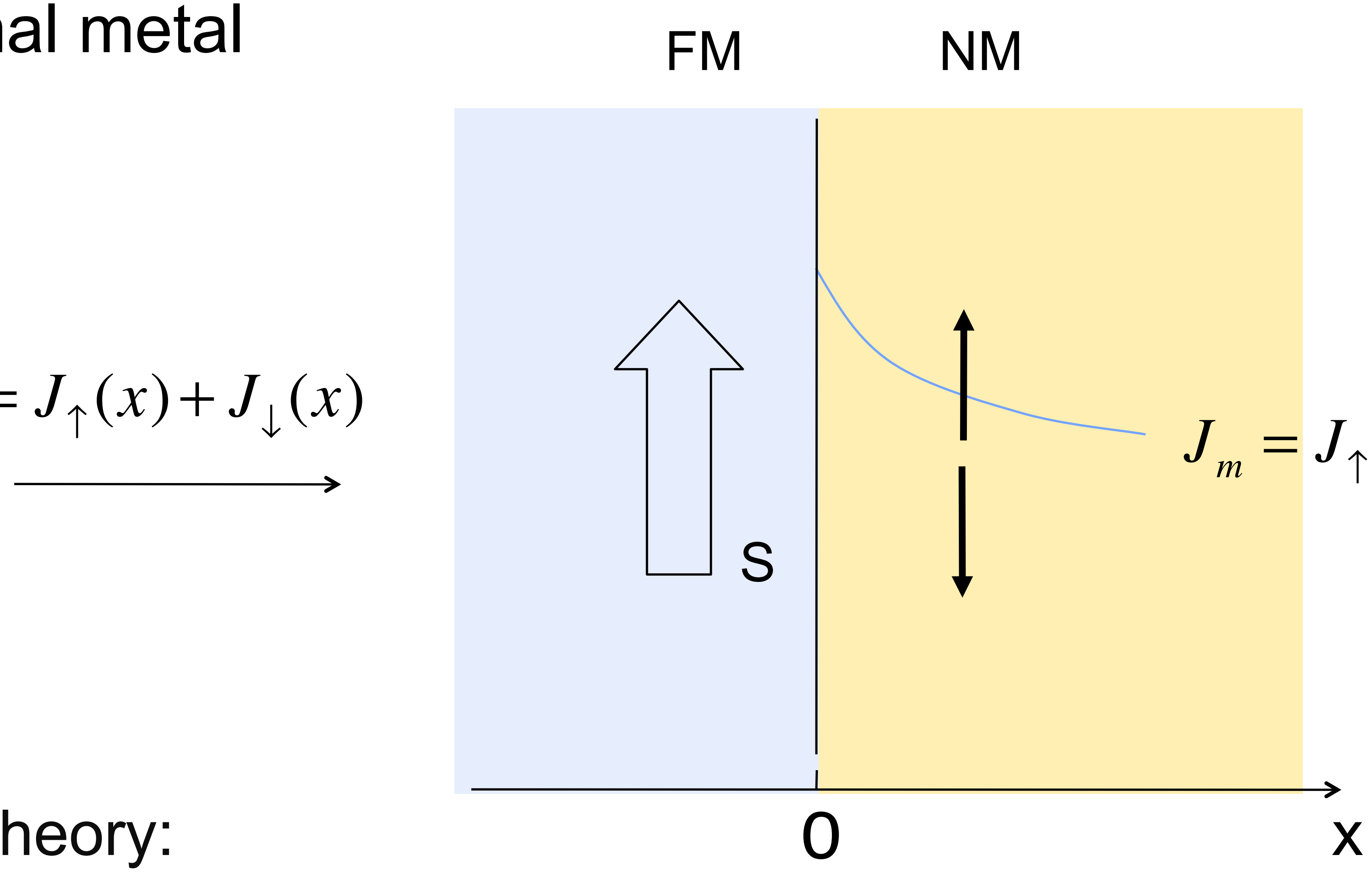


$$J_m = J_\uparrow(x) - J_\downarrow(x)$$

Diffusion theory:

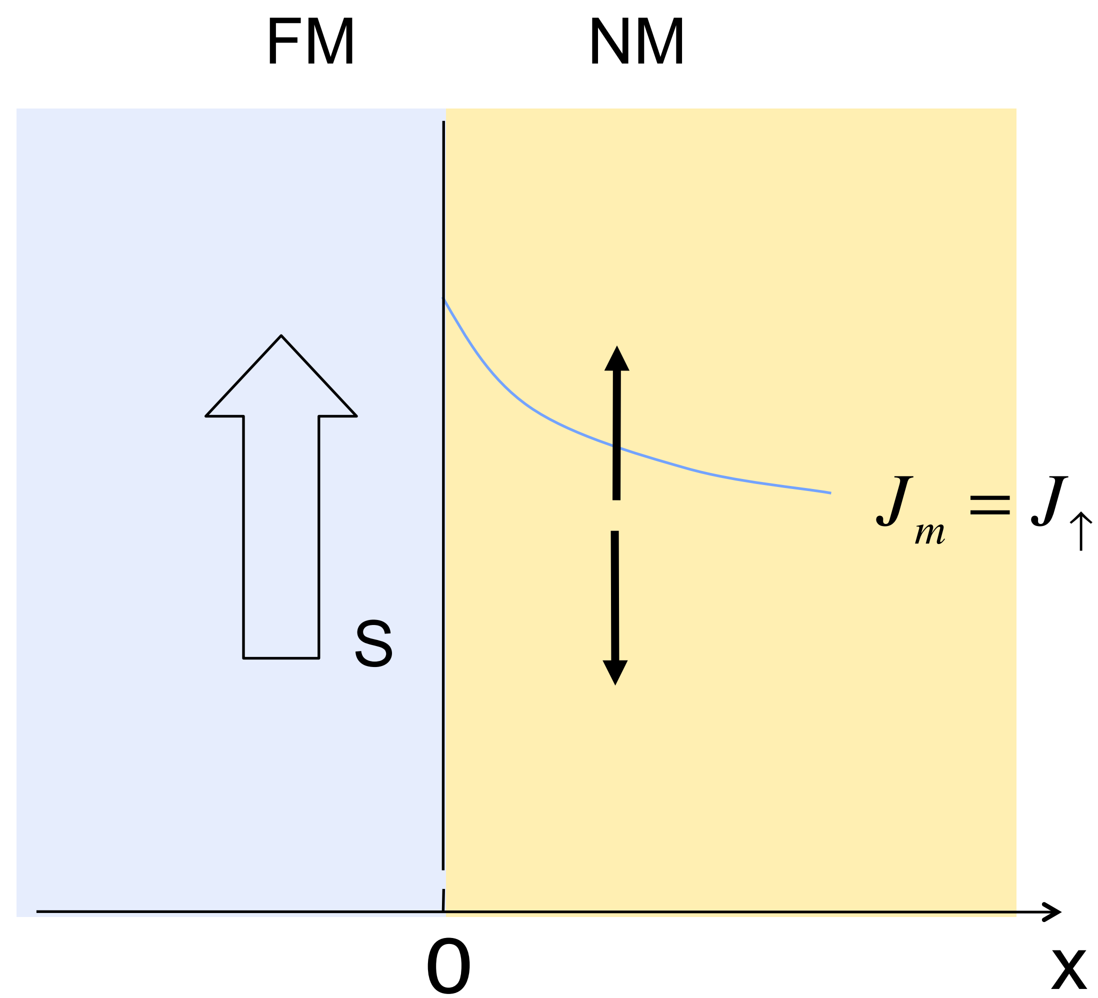
$$\mu_m(z,t) \propto \int_0^t \frac{dt'}{\sqrt{\pi \bar{D} t'}} \exp\left[-\frac{z^2}{4\bar{D}t'} - \frac{t'}{T_1}\right]$$

$$J_m(z,t) \propto \frac{\partial \mu_m(z,t)}{\partial z}$$

Spin Signal-Propagation

- Spin signal transmitted into normal metal

$$J = J_{\uparrow}(x) + J_{\downarrow}(x)$$



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$$J_m(z,t) \propto \frac{\partial \mu_m(z,t)}{\partial z}$$

Infinite signal velocity

Boltzmann Equation (noncollinear)

- Single-particle density matrix

$$\hat{\rho}_{\sigma,\sigma'}(\vec{r}_1,\vec{r}_2) = \left\langle \psi^\dagger_\sigma(\vec{r}_1)\psi_{\sigma'}(\vec{r}_2)\right\rangle \rightarrow \hat{\rho}_{\sigma,\sigma'}(\vec{k},\vec{r})$$

- Matrix in "spin space": $\hat{\rho} = \begin{pmatrix} \rho_{\uparrow\uparrow} & \rho_{\uparrow\downarrow} \\ \rho_{\downarrow\uparrow} & \rho_{\downarrow\downarrow} \end{pmatrix}$
- Boltzmann equation

$$\left.\frac{\partial \hat{\rho}}{\partial t}\right|_{\text{relax}}$$

$$\frac{\partial \hat{\rho}}{\partial t} + v_x \frac{\partial \hat{\rho}}{\partial x} - \frac{eE}{m^*}\frac{\partial \hat{\rho}}{\partial v_x} + \frac{1}{2}\gamma(\vec{u}\times\vec{B}_s)\cdot\boldsymbol{\sigma} = -\frac{\hat{\rho} - \langle\hat{\rho}\rangle_a}{\tau} - \frac{\langle\hat{\rho}\rangle_a - (\hat{I}/2)\mathrm{Tr}\langle\hat{\rho}\rangle_a}{T_1}$$

- Bloch vector: $\vec{u} = \mathrm{Tr}\left[\vec{\sigma}\,\hat{\rho}\right]$ spin direction

Y.-H. Zhu, B. Hillebrands, and H. C. Schneider, Phys. Rev. B **79**, 214412 (2009), see also Y. Qi and S. Zhang, Phys. Rev. B **67**, 052407 (2003).

Boltzmann Equation (noncollinear)

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- Boltzmann equation

$$\left.\frac{\partial \hat{\rho}}{\partial t}\right|_{\text{relax}}$$

$$\frac{\partial \hat{\rho}}{\partial t} + v_x \frac{\partial \hat{\rho}}{\partial x} - \frac{eE}{m^*}\frac{\partial \hat{\rho}}{\partial v_x} + \frac{1}{2}\gamma(\vec{u}\times\vec{B}_s)\cdot\boldsymbol{\sigma} = -\frac{\hat{\rho}-\langle\hat{\rho}\rangle_a}{\tau} - \frac{\langle\hat{\rho}\rangle_a - (\hat{I}/2)\mathrm{Tr}\langle\hat{\rho}\rangle_a}{T_1}$$

- Bloch vector: $\vec{u} = \mathrm{Tr}\left[\vec{\sigma}\,\hat{\rho}\right]$ spin direction

- Macroscopic equations: summation over k (or v); only space dependence in x

$$\vec{S}(\vec{r},t) = \frac{\hbar}{2V}\sum_{\vec{k}}\vec{u}(\vec{k},\vec{r},t)$$

$$Q_{\alpha,\beta}(\vec{r},t) = \frac{\hbar}{2V}\sum_{\vec{k}} v_\alpha u_\beta(\vec{k},\vec{r},t)$$

spin density vector: $\vec{S}(x,t)$

spin current density tensor: $J_m^\alpha(x,t) = Q_{\alpha,x}(x,t)$

Y.-H. Zhu, B. Hillebrands, and H. C. Schneider, Phys. Rev. B **79**, 214412 (2009),
see also Y. Qi and S. Zhang, Phys. Rev. B **67**, 052407 (2003).

General Macroscopic Dynamical Equations

- General structure

$$\frac{\partial \vec{S}(x,t)}{\partial t} = -\gamma \vec{S} \times \vec{B} - \frac{\partial \vec{J}_m}{\partial x} - \left.\frac{\partial \vec{S}(x,t)}{\partial t}\right|_{\text{relax}}$$

$$c_{\text{sig}} = \frac{v_{\text{F}}}{\sqrt{3}}$$

$$\frac{\partial \vec{J}_m(x,t)}{\partial t} = -c_{sig}^2 \frac{\partial S}{\partial x} - \frac{e}{m^*} E(x,t)\vec{S} - \gamma \vec{J}_m \times \vec{B} - \left.\frac{\partial \vec{J}_m}{\partial t}\right|_{\text{relax}}$$

- Including simple relaxation terms: spin flip ($T_2 = T_1 = \frac{1}{2}\tau_{\text{sf}}$) and momentum relaxation times (τ)

$$\frac{\partial \vec{S}(x,t)}{\partial t} = -\gamma \vec{S} \times \vec{B} - \frac{\vec{S}}{T_1} - \frac{\partial \vec{J}_m}{\partial x}$$

$$\frac{\partial \vec{J}_m(x,t)}{\partial t} = -c_{sig}^2 \frac{\partial S}{\partial x} - \frac{e}{m^*} E(x,t)\vec{S} - \gamma \vec{J}_m \times \vec{B} - \frac{1}{\tau}\vec{J}_m$$

General Macroscopic Dynamical Equations

- General structure

$$\frac{\partial \vec{S}(x,t)}{\partial t} = -\gamma \vec{S} \times \vec{B} - \frac{\partial \vec{J}_m}{\partial x} - \left.\frac{\partial \vec{S}(x,t)}{\partial t}\right|_{\text{relax}}$$

$$c_{\text{sig}} = \frac{v_{\text{F}}}{\sqrt{3}}$$

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General Macroscopic Dynamical Equations

- General structure

$$\frac{\partial \vec{S}(x,t)}{\partial t} = -\gamma \vec{S} \times \vec{B} - \frac{\partial \vec{J}_m}{\partial x} - \left.\frac{\partial \vec{S}(x,t)}{\partial t}\right|_{\text{relax}}$$

$$c_{\text{sig}} = \frac{v_{\text{F}}}{\sqrt{3}}$$

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- Including simple relaxation terms: spin flip ($T_2 = T_1 = \frac{1}{2}\tau_{\text{sf}}$) and momentum relaxation times (τ)

$$\frac{\partial \vec{S}(x,t)}{\partial t} = -\gamma \vec{S} \times \vec{B} - \frac{\vec{S}}{T_1} - \frac{\partial \vec{J}_m}{\partial x}$$

$$\vec{J}_m(x,t) = -D \frac{\partial S}{\partial x} - \mu E(x,t) \vec{S} - \tau \gamma \vec{J}_m \times \vec{B} - \tau \frac{\partial \vec{J}_m}{\partial t}$$

Collinear Macroscopic Equations

Averaging time-dependent Boltzmann around Fermi energy

→ wave diffusion equations

$$\frac{\partial n_s(x,t)}{\partial t} + \frac{\partial J_s(x,t)}{\partial x} = -\frac{n_s(x,t) - n_{-s}(x,t)}{\tau_{\mathrm{sf}}} \quad \text{where} \quad T_1 = \frac{1}{2}\tau_{\mathrm{sf}}$$

$$\frac{J_s(x,t)}{\tau_s} = -c_{\mathrm{sig}}^2 \frac{\partial n_s(x,t)}{\partial x} - \boxed{\frac{\partial J_s(x,t)}{\partial t}}$$

not included in diffusion theory

Finite signal propagation velocity $\boxed{c_{\mathrm{sig}} = \frac{v_{\mathrm{F}}}{\sqrt{3}}}$

Y.-H. Zhu, B. Hillebrands, and H. C. Schneider, PRB **78**, 054429 (2008)

Analytical Solution

wave-diffusion equation

$$\frac{\partial^2 n_m}{\partial t^2} + \left(\frac{1}{\tau} + \frac{1}{T_1}\right)\frac{\partial n_m}{\partial t} + \frac{n_m}{\tau T_1} = c_{\mathrm{sig}}^2 \frac{\partial^2 n_m}{\partial x^2}$$

with $n_m = n_+ - n_-$

plane-wave ansatz $n_m(x,t) \propto \exp\left[i(kx - \omega t)\right]$

dispersion relation $\omega(k) = \omega_R(k) + \omega_I(k)$

$$-\omega^2 - i\left(\frac{1}{\tau} + \frac{1}{T_1}\right)\omega + \frac{1}{\tau T_1} = -c_{\mathrm{sig}}^2 k^2$$

calcuate mean-square displacement for intial condition $n_m(x,t=0) = \delta(x)$

$$\Delta_x^2 = \int dx x^2 n_m(x,t)$$

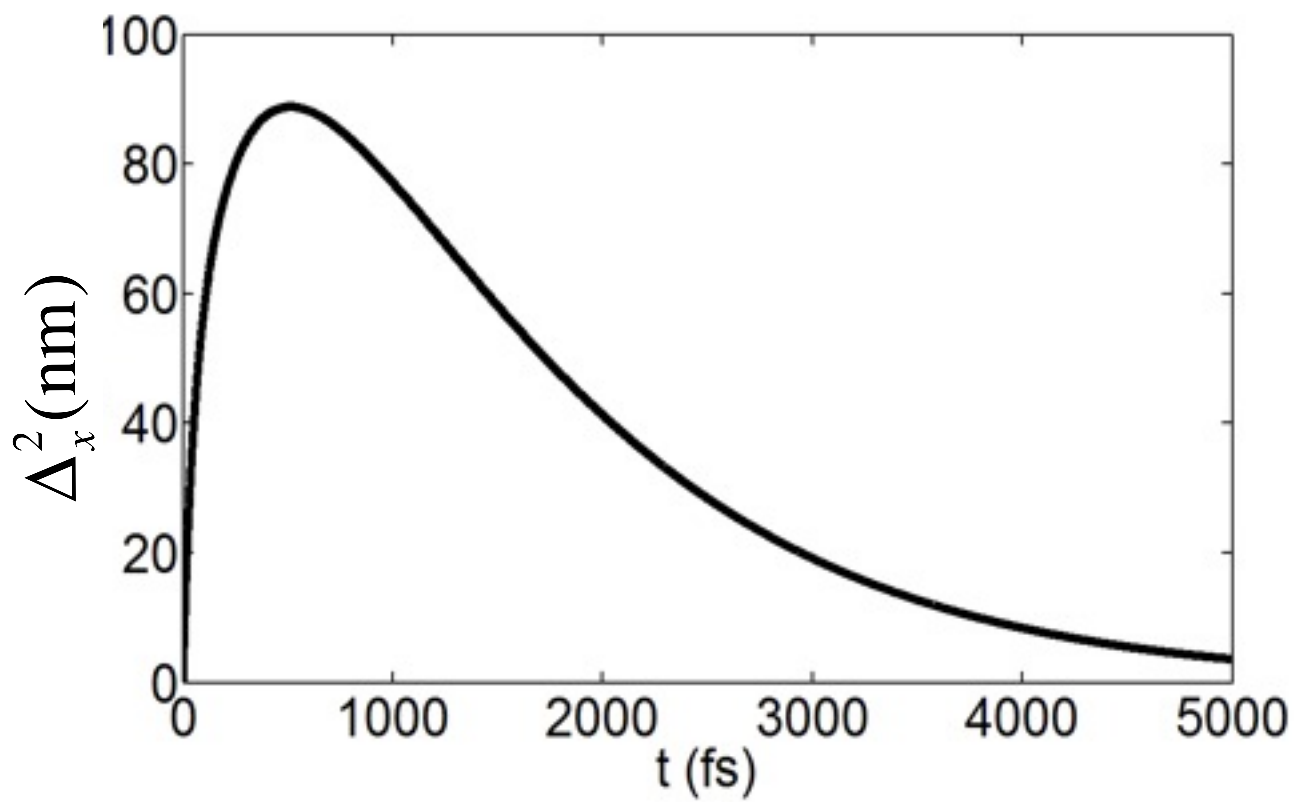
Steffen Kaltenborn, Yao-Hui Zhu, and Hans Christian Schneider, Phys. Rev. B **85**, 235101

Spin Wave-Diffusion Equation: Analytical Solutions (2)

- Calculate mean-square displacement for wave solution

$$\Delta_x^2 = \int dx\, x^2 n_m(x,t) \propto c_{\text{sig}}^2 t \left(\frac{1}{\tau} + \frac{1}{T_1} \right) \left(e^{-t/\tau} - e^{-t/T_1} \right)$$

- If $\Delta_x^2 \propto c_{sig}^2 t^2$ transport is <u>ballistic</u>
- If $\Delta_x^2 \propto t$ transport is <u>diffusive</u>



parameters for Cu

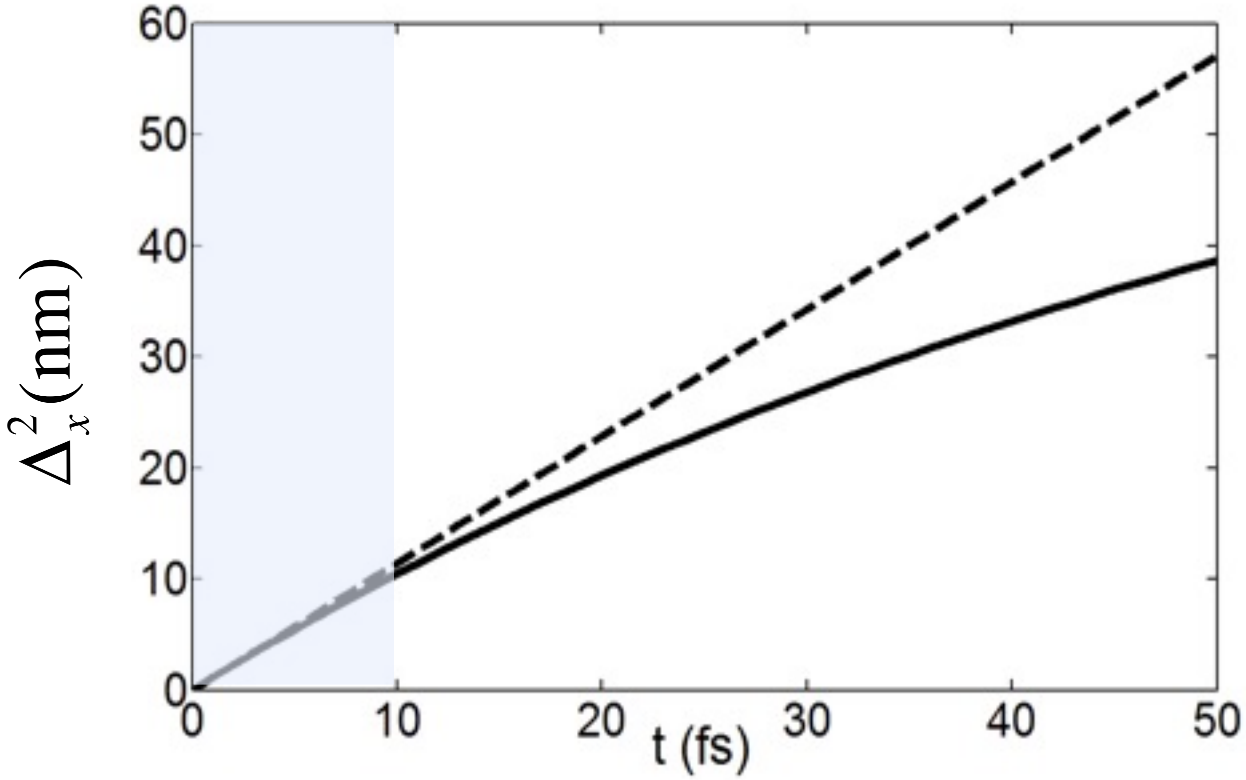
$$v_{\text{F}} = 1.4 \text{ nm/fs}$$

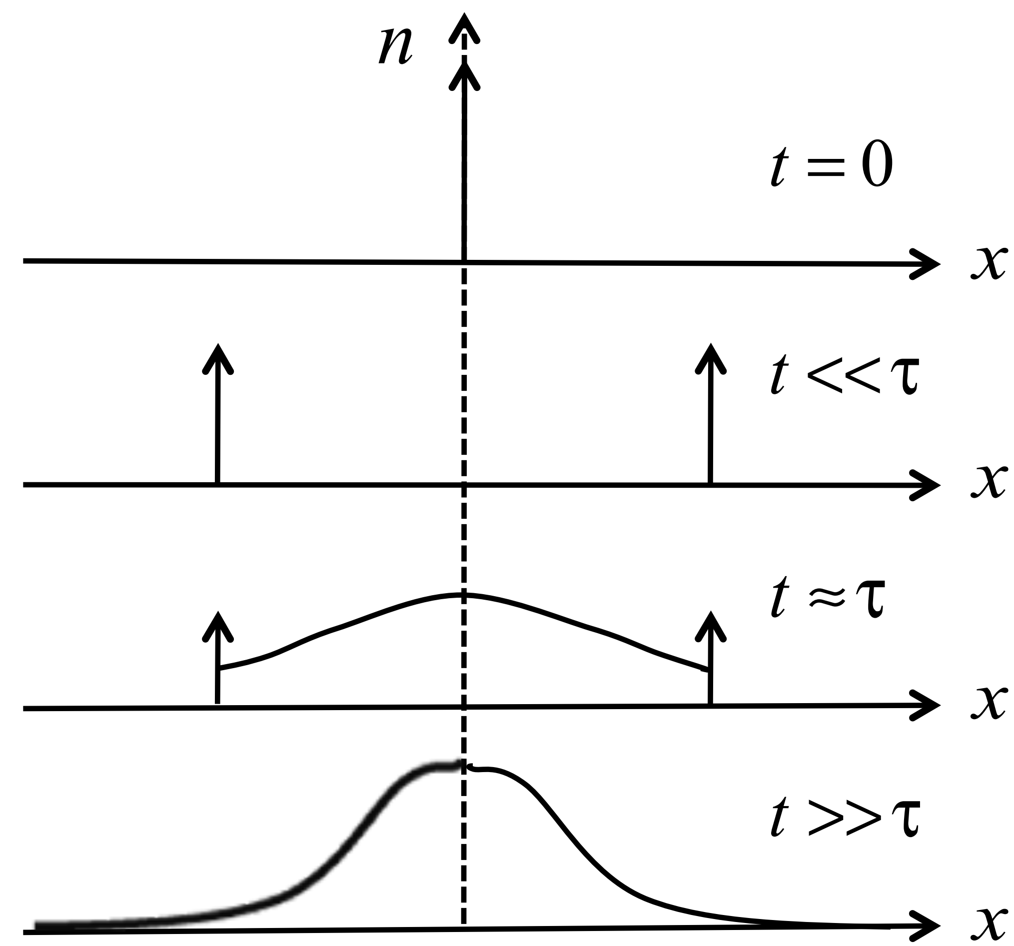
$$\tau = 30 \text{ fs}$$

$$T_1 = 515 \text{ fs}$$

Ballistic and Diffusive Transport

- Ballistic and diffusive behavior from wave-diffusion equation
- Transition between two regimes also covered

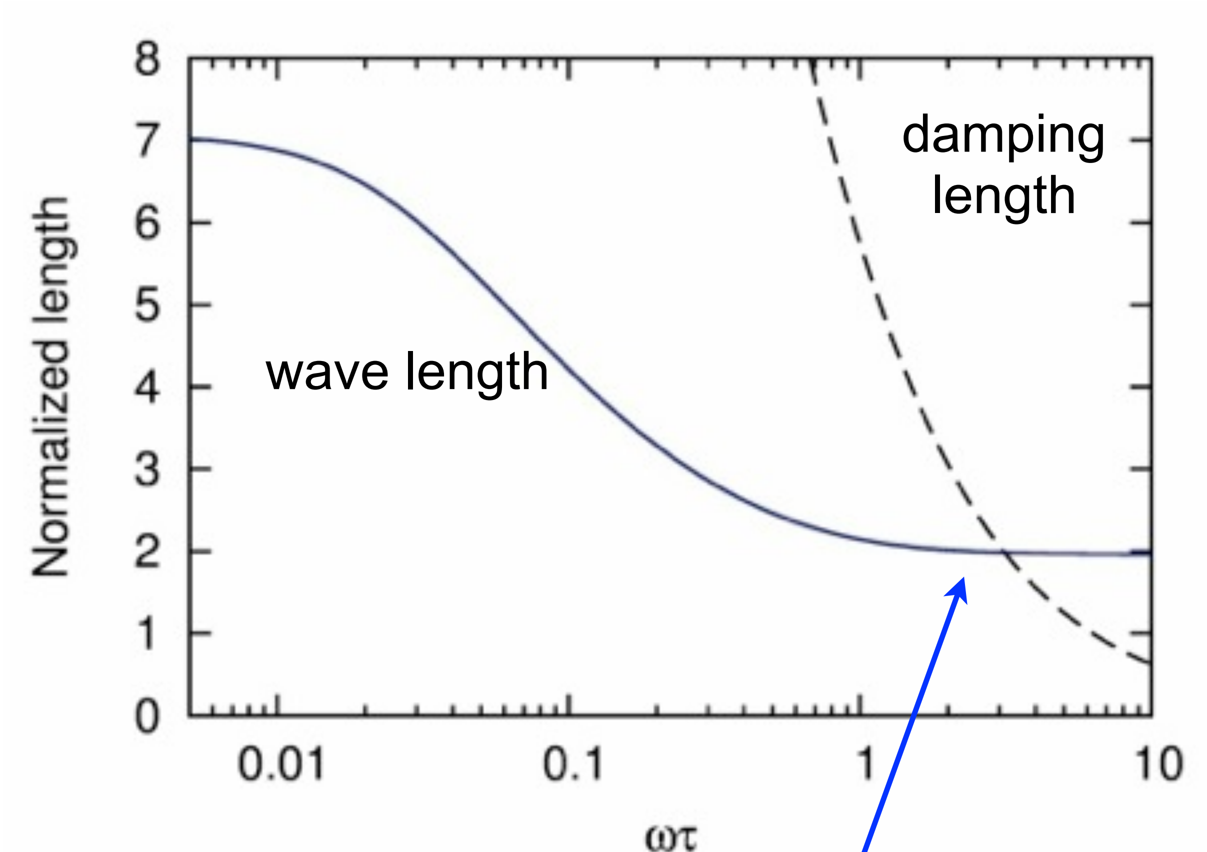




Steffen Kaltenborn, Yao-Hui Zhu, and Hans Christian Schneider, Phys. Rev. B **85**, 235101

Spin Wave-Diffusion Equation: Analytical Solutions (3)

- Plane wave ansatz for steady state: $J_m(x,t) \propto \exp\left[i(kx-\omega t)\right]$
- Complex wave vector $k(\omega)=k_R(\omega)+k_I(\omega)$
- Frequency-dependent wavelength and damping length



$\nu_c = 7.1$ THz for Cu

Wave character significant above the critical frequency

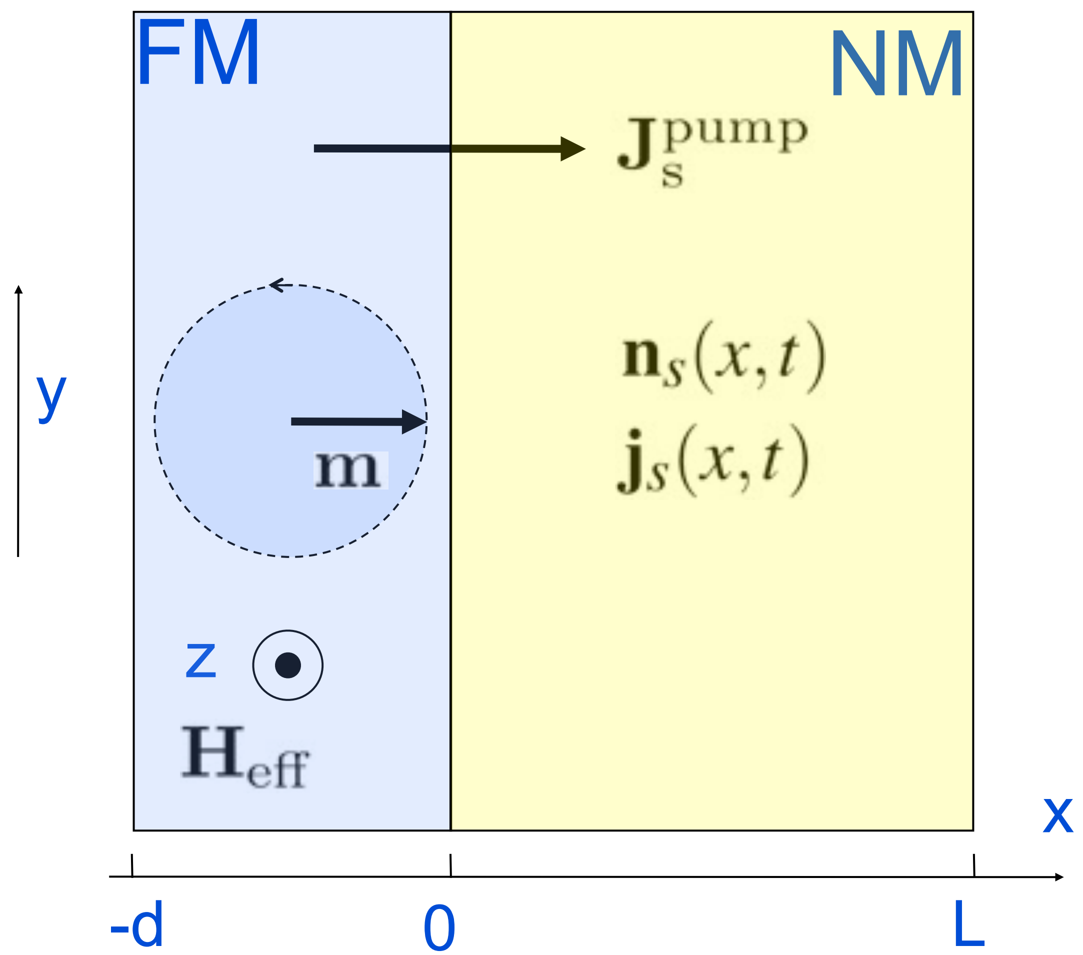
Y.-H. Zhu, B. Hillebrands, and H. C. Schneider, PRB **78**, 054429 (2008),

Y.-H. Zhu and H. C. Schneider, IEEE Trans. Magn. **45**, 4395 (2009)

Outline

1. Ultrafast demagnetization in ferromagnets
2. Elliott-Yafet demagnetization due to electron-phonon scattering and optical excitation dynamics from ab-initio calculations
3. Limits to scattering in a fixed bandstructure
4. Extensions of the Elliott-Yafet approach
5. Wave-diffusion theory of spin and charge transport in metals
6. Application to noncollinear spin currents
7. Application to optically excited spin-polarized currents

FM precessing around magnetic field



Spin-mixing conductance

- Spin current pumped into NM: $$\vec{j}_s^{\,\text{pump}} = \frac{1}{2\pi}\frac{g^{\uparrow\downarrow}}{S}\left[\vec{m}\times\frac{d\vec{m}}{dt}\right]$$

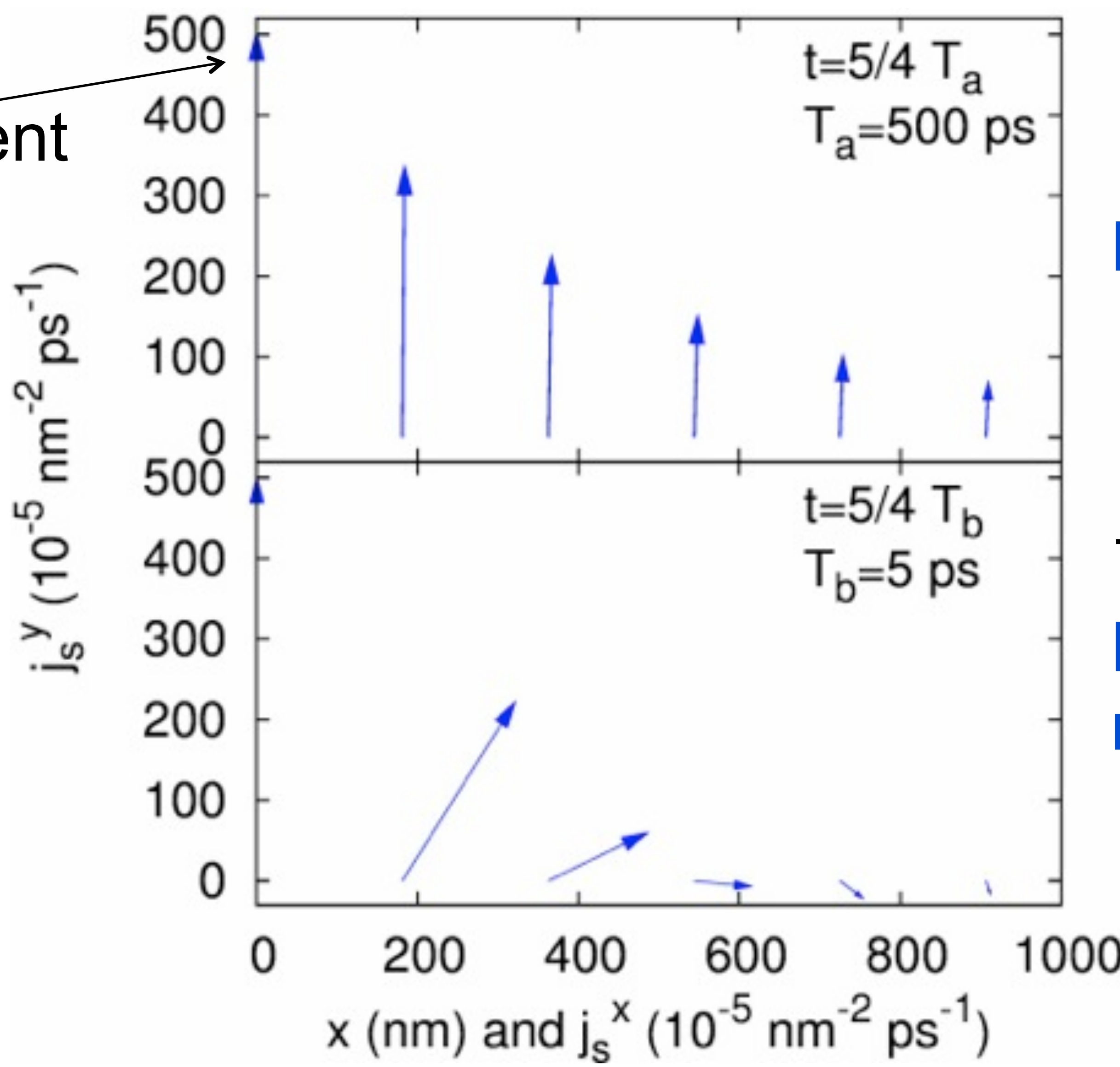
Tserkovnyak et al, Phys. Rev. Lett. **88**, 117601 (2002);
Phys. Rev. B **66**, 224403 (2002)

Diffusion-Dominated Region

- Below the critical frequency: 7.11 THz

Snapshots of transverse components

Pumped current



f_a=2 GHz

Diffusion equation applicable

f_b=200 GHz

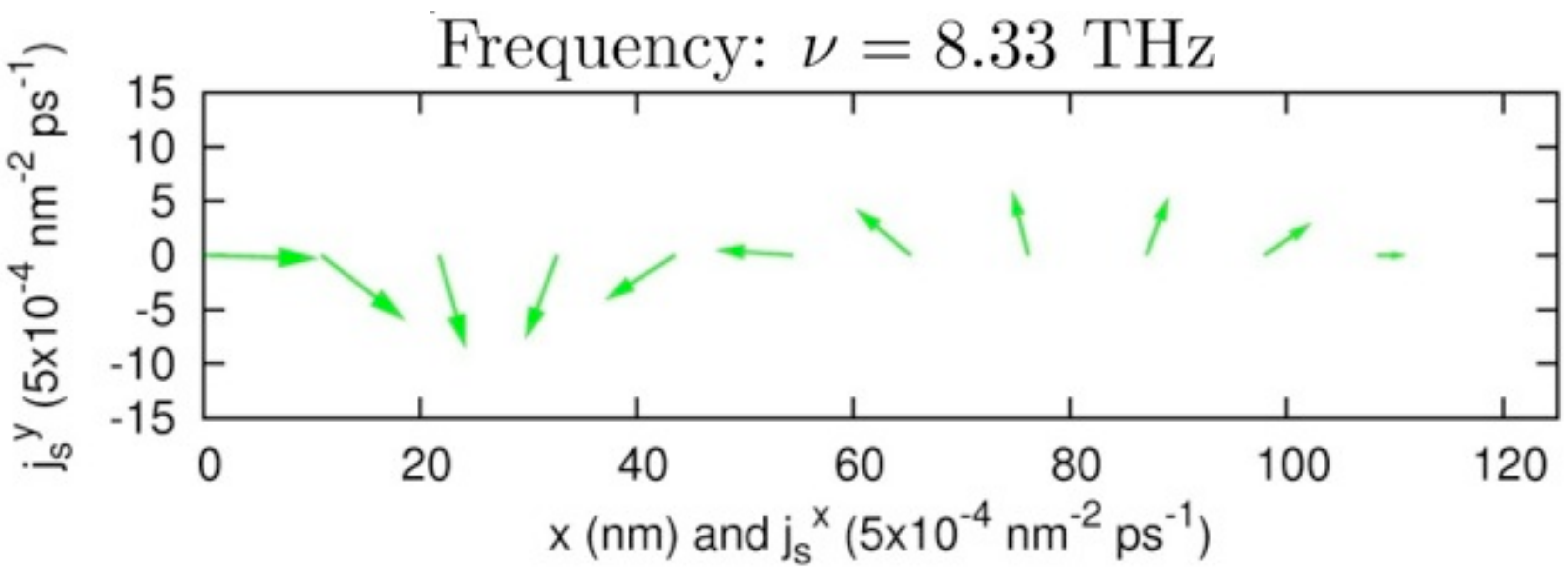
Diffusion equation yields wrong result

- ‘Skin’ effect: decrease of damping length with frequency

Y.-H. Zhu, B. Hillebrands, and H. C. Schneider, Phys. Rev. B **79**, 214412 (2009)

Wave-Dominated Region

- Above the critical frequency



Wave-front velocity estimated: $\approx c_s = \frac{v_F}{\sqrt{3}}$

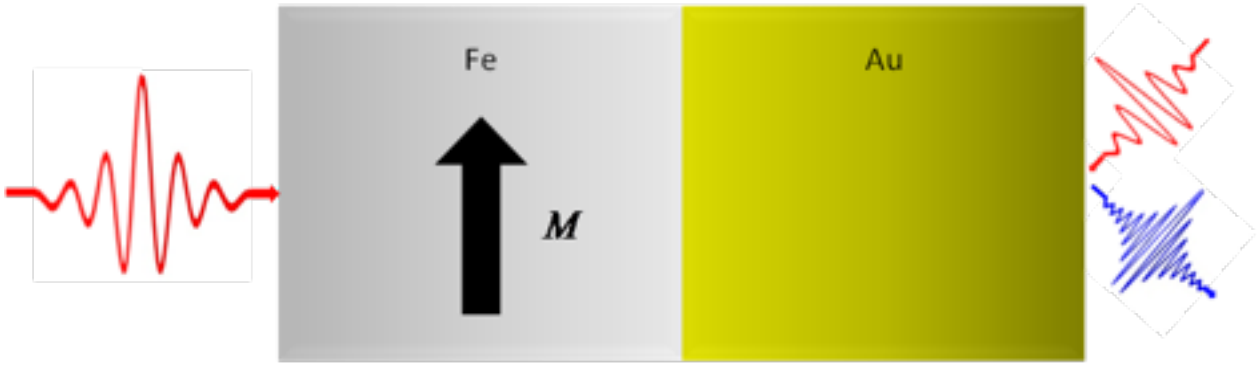
- Completely different from diffusion theory!

Y.-H. Zhu, B. Hillebrands, and H. C. Schneider, Phys. Rev. B **79**, 214412 (2009)

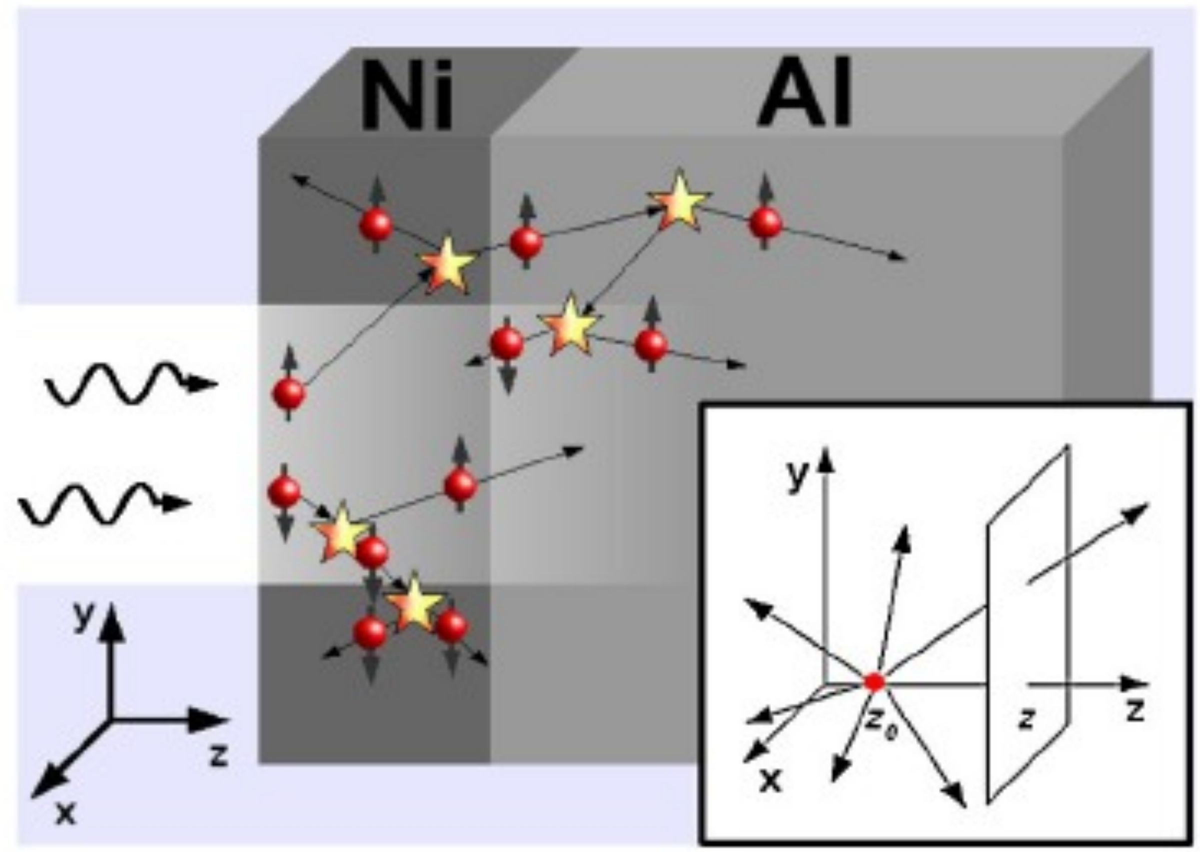
Optical Excitation, Scattering Transport in Metals/ Ferromagnets

- Spin-dependent transport on ultrashort timescales
- Ballistic transport important
- Influence of “hot” electrons?
- No Boltzmann transport and scattering calculation available

- Here: Use our simpler, macroscopic approach



A. Melnikov et al., PRL **107**, 0766011 (2011)



M. Battiato et al., PRL **105**, 027203 (2010): superdiffusive transport (includes scattering)

Outline

1. Ultrafast demagnetization in ferromagnets
2. Elliott-Yafet demagnetization due to electron-phonon scattering and optical excitation dynamics from ab-initio calculations
3. Limits to scattering in a fixed bandstructure
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7. Application to optically excited spin-polarized currents

Optically Excited Dynamics in Gold

Spin

$$\frac{\partial n_s(x,t)}{\partial t} + \frac{\partial J_s(x,t)}{\partial x} = -\frac{n_s(x,t) - n_{-s}(x,t)}{\tau_{\mathrm{sf}}}$$

$$\frac{J_s(x,t)}{\tau_s} = -c_{\mathrm{sig}}^2 \frac{\partial n_s(x,t)}{\partial x} - \frac{\partial J_s(x,t)}{\partial t}$$

Charge

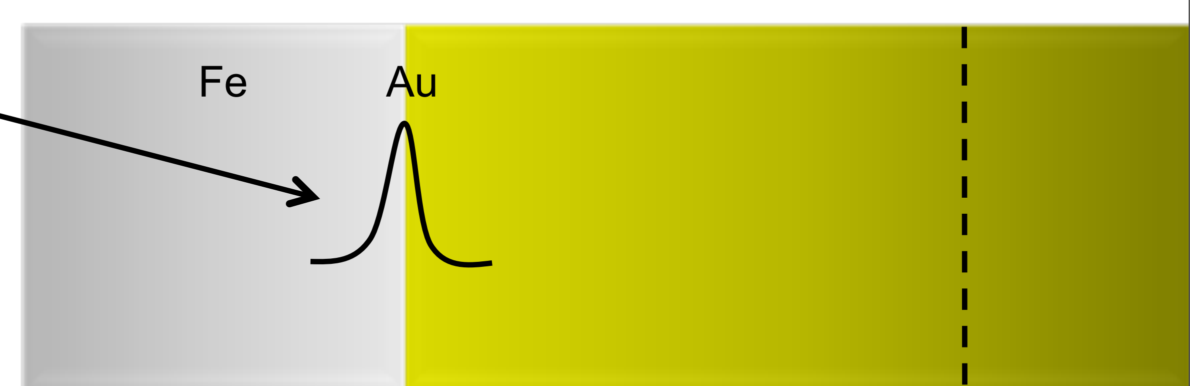
$$\frac{\partial n(x,t)}{\partial t} + \frac{\partial J(x,t)}{\partial x} = 0$$

$$\frac{J(x,t)}{\tau_s} = -c_{\mathrm{sig}}^2 \frac{\partial n(x,t)}{\partial x} - \frac{\partial J(x,t)}{\partial t}$$

left boundary condition: Gaussian pulse

$$J_s(x=0,t) = J^{(0)} \exp\left(-\frac{(t-t_0)^2}{2\sigma^2}\right)$$

pulse duration: 35 fs

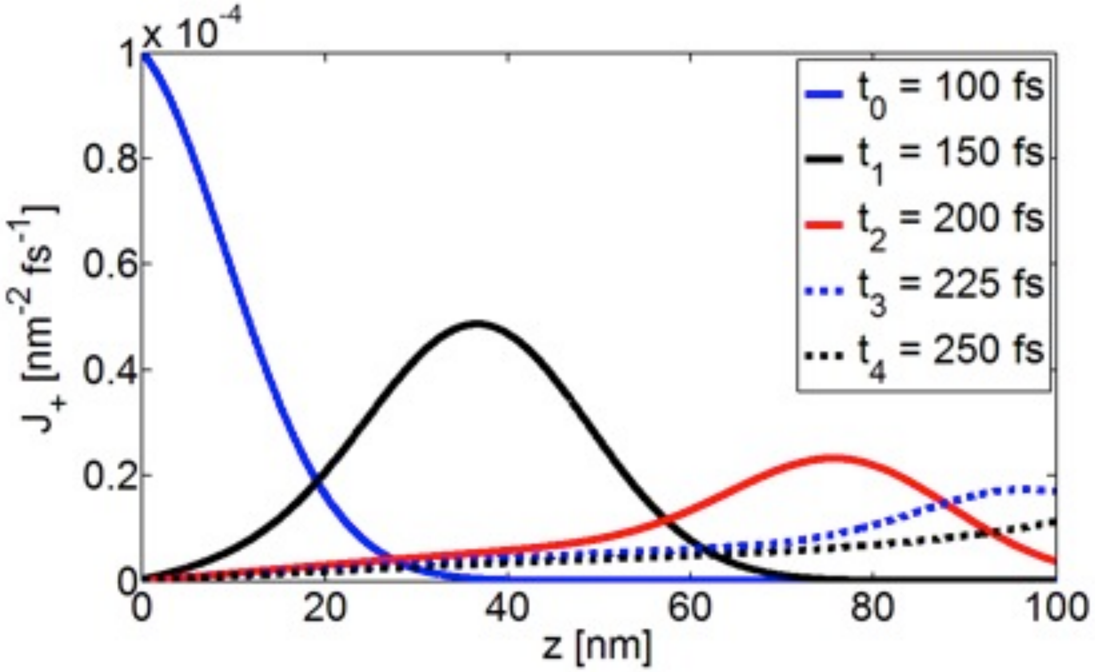


different right boundary conditions: with and without reflection

<u>model spin dependence by</u> $\tau_{\uparrow} = 30\,\mathrm{fs} \neq \tau_{\uparrow} = 31.5\,\mathrm{fs}$

Current Dynamics

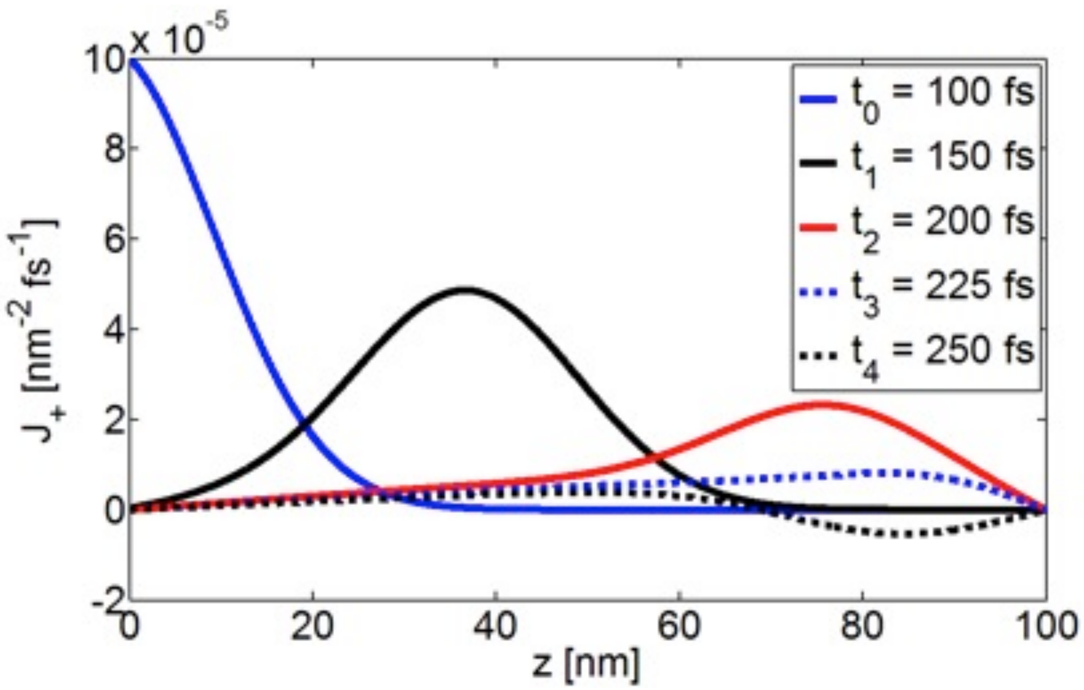
Without Reflection



Gaussian pulse propagates

Broadening shows ballistic and diffusive contributions

With Reflection



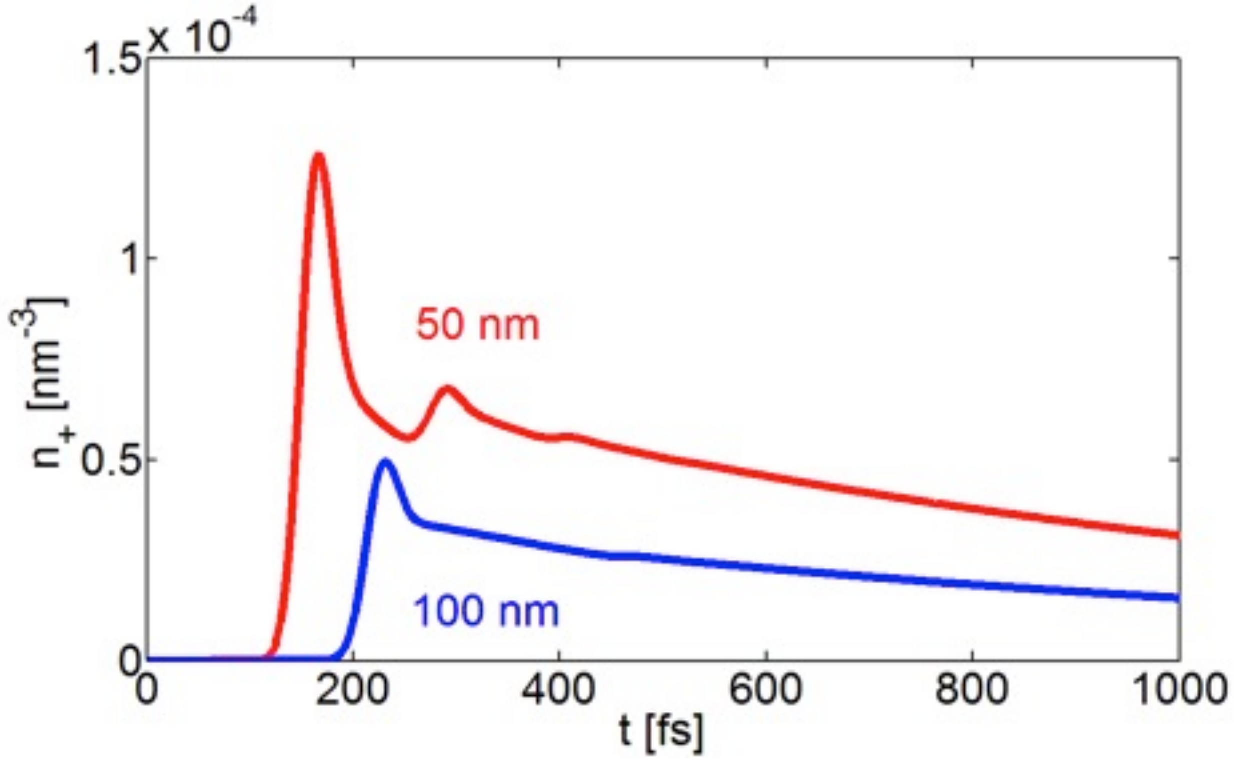
Finite slab thickness/reflection yields a negative spin-current density

Steffen Kaltenborn, Yao-Hui Zhu, and Hans Christian Schneider, Phys. Rev. B **85**, 235101

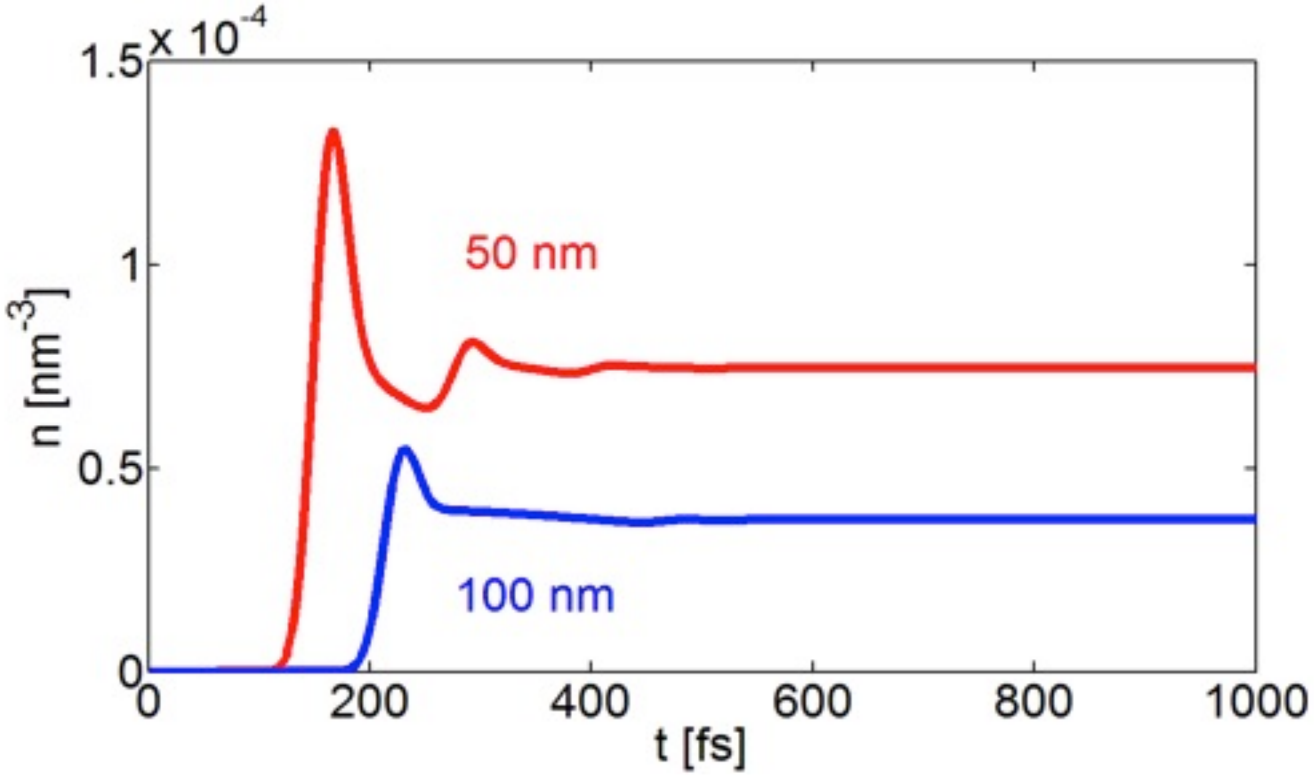
Density Dynamics

- Aha

Majority Electrons



Charge



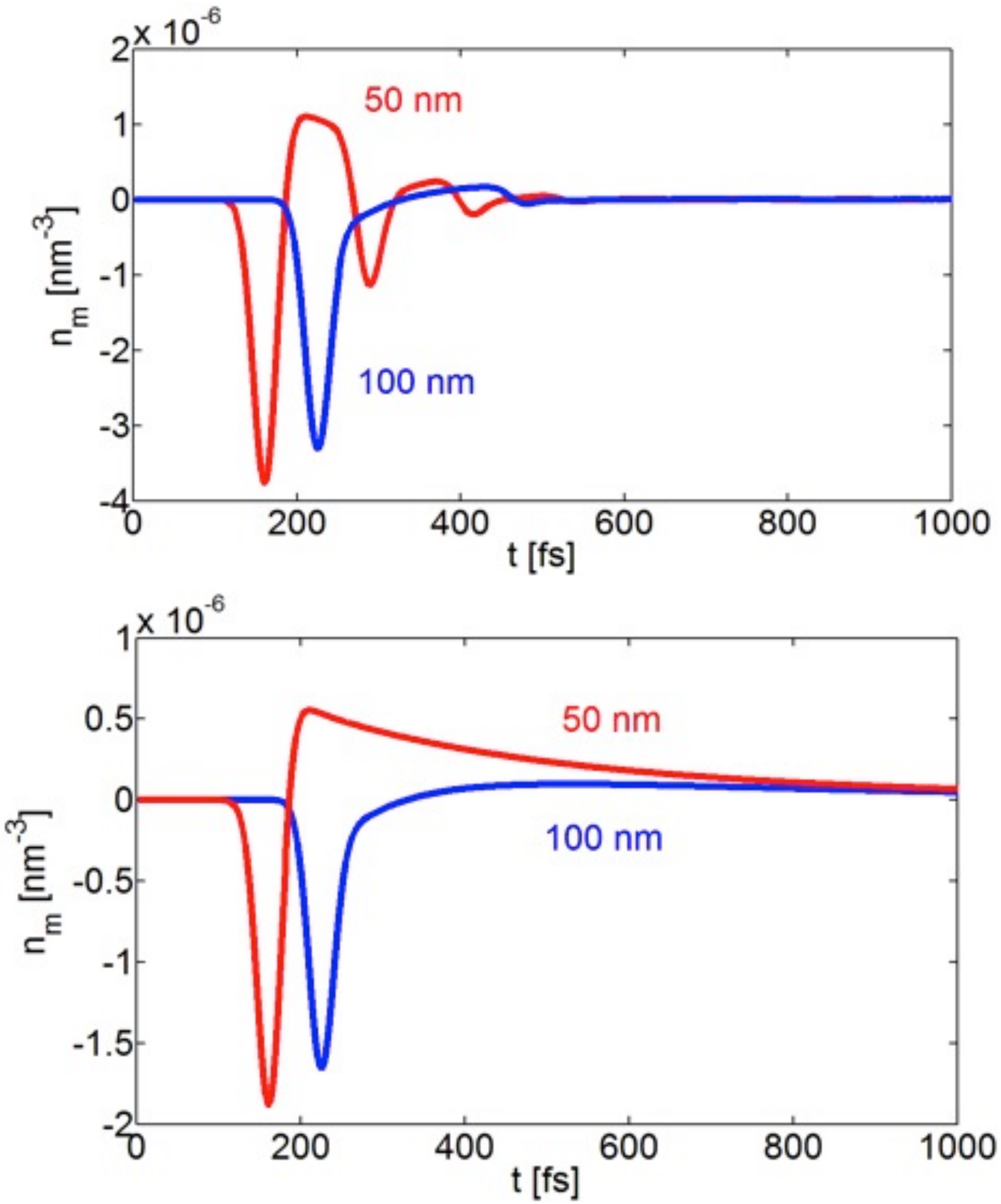
finite width of the peak

→ ballistic and diffusive contributions

multiple reflections

Dynamics at Right Boundary

Spin Polarization



Spin dynamics change for spin-dependent excitation

$$\tau_+ = 30 \text{ fs} \neq \tau_- = 31.5 \text{ fs}$$

typical characteristic:

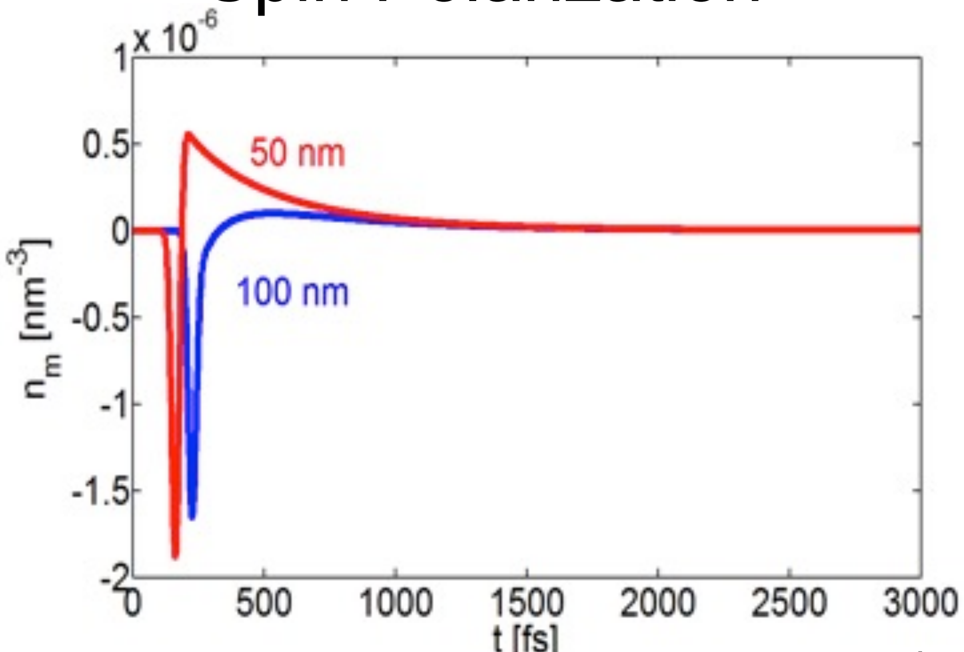
short negative spike

long positive tail

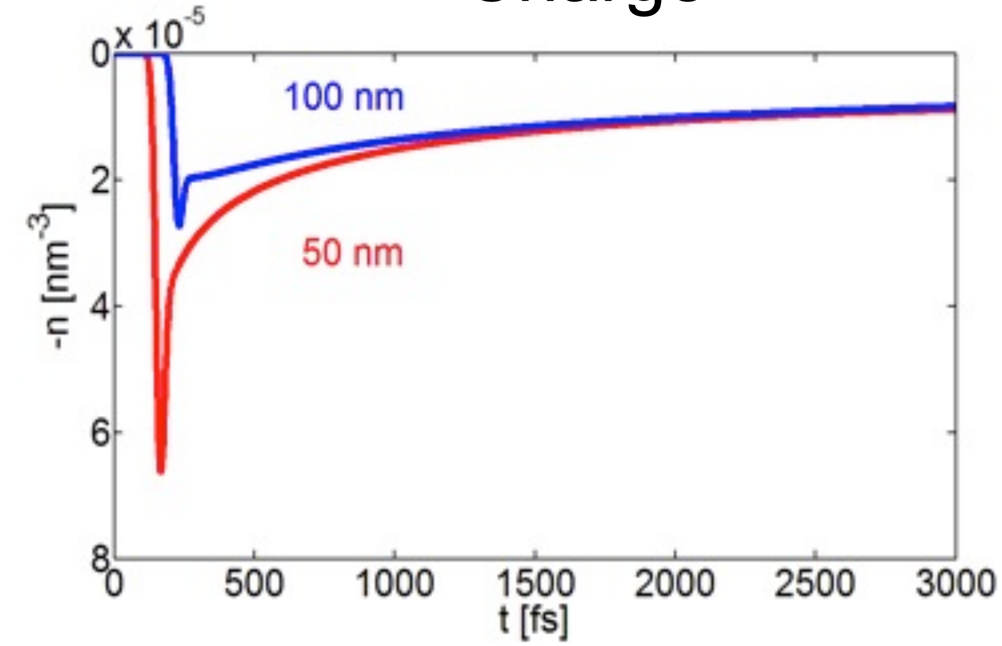
Steffen Kaltenborn, Yao-Hui Zhu, and Hans Christian Schneider, Phys. Rev. B **85**, 235101

Spin and Charge Dynamics: Expt. vs. Theory

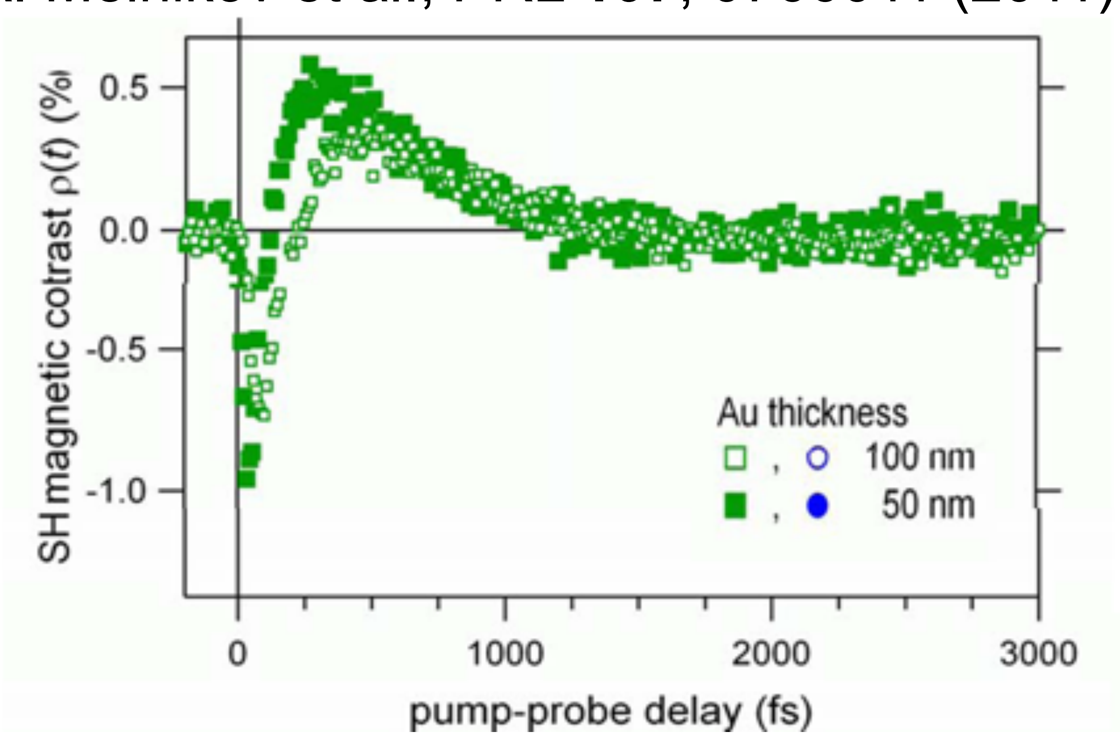
Spin Polarization

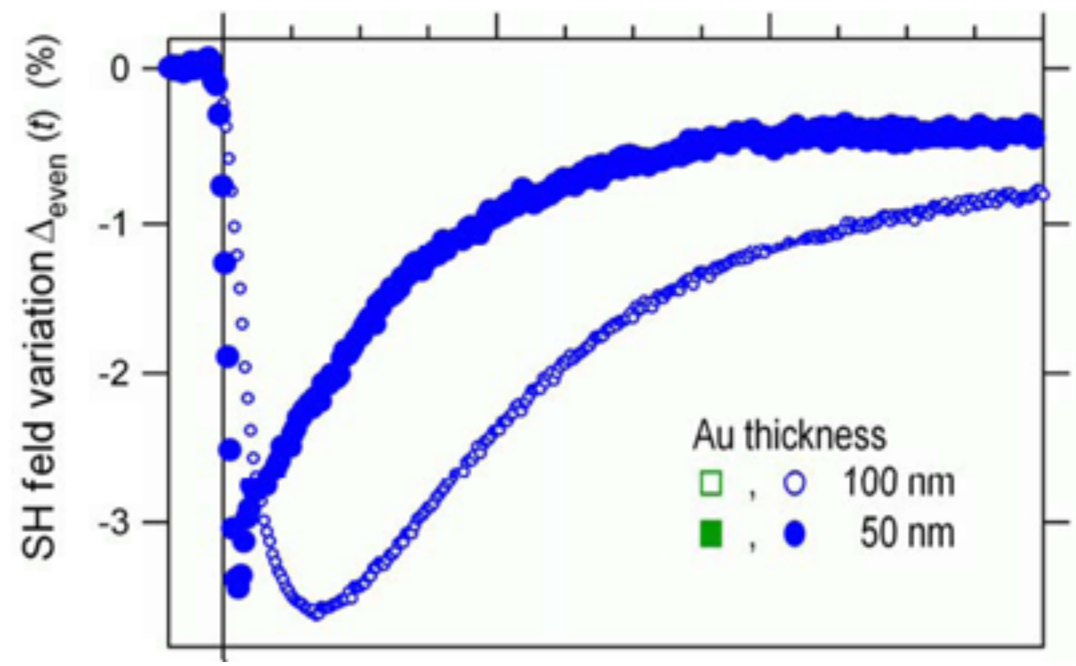


Charge



A. Melnikov et al., PRL **107**, 0766011 (2011):





<u>Qualitative agreement</u>

Conclusions (2)

- Macroscopic equation system for unified description of ballistic and diffusive spin and charge transport

- Model based on well established transport parameters

$$\tau, v_F \text{ and } T_1$$

- Qualitative explanation of key features of spin and charge dynamics after ultrashort pulse excitation
- No superdiffusive transport

- Quantitative studies and comparisons needed