Towards THz spintronics: generation and transport of sub-picosecond spin current pulses The non-trivial interplay of transport, thermalization and electromagnetism



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from the 1st of November at

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Moore speed?



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Page 1/32

Ultrafast demagnetization



E. Beaurepaire et al., Phys. Rev. Lett. 76, 4250 (1996).





Ultrafast demagnetization



Few hundreds of fs!



E. Beaurepaire et al., Phys. Rev. Lett. 76, 4250 (1996).



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Page 3/32

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Page 3/32

The puzzle of the ultrafast demagnetization







The puzzle of the ultrafast demagnetization

Electron-phonon coupling

Spin to orbital momentum conversion



The puzzle of the ultrafast demagnetization

Electron-phonon coupling Spin to orbital momentum conversion Superdiffusive spin transport

 $\hat{\Phi}S^{eff} =$





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$$\frac{\partial n\left(\sigma, E, z, t\right)}{\partial t} + \frac{n\left(\sigma, E, z, t\right)}{\tau\left(\sigma, E, z\right)} = \left(-\frac{\partial}{\partial z}\hat{\Phi} + \hat{I}\right)S^{\text{eff}}\left(\sigma, E, z, t\right)$$

$$\widetilde{[\Delta t]}\left(z, z_{0}\right) = \int_{z_{0}}^{z} \frac{dz'}{v\left(z'\right)} \quad ; \qquad \widetilde{[\Delta t/\tau]}\left(z, z_{0}\right) = \int_{z_{0}}^{z} \frac{dz'}{v\left(z'\right)\tau\left(z'\right)}$$

$$\int_{-\infty}^{+\infty} dz_{0} \int_{-\infty}^{t} dt_{0}S^{\text{eff}}\left(\sigma, E, z_{0}, t_{0}\right) \frac{\widetilde{[\Delta t]}}{2\left(t - t_{0}\right)^{2}} e^{-\left(t - t_{0}\right)\frac{\widetilde{[\Delta t/\tau]}}{[\Delta t]}} \Theta\left[\left(t - t_{0}\right) - \left|\widetilde{[\Delta t]}\right|$$

Battiato, Carva, Oppeneer, Phys. Rev. Lett. 105, 027203 (2010)



















 $v \approx 1 nm/fs$



•







 $v \approx 1 nm/fs$

 $\tau \approx tens of fs$





04/04/2017 IOP Magnetism 2017 York Page 5/32



- $v \approx 1 nm/fs$
- $\tau \approx tens of fs$
- $\lambda \approx$ tens of nm









 $v \approx 1$ nm/fs

 $\tau \approx tens of fs$

 $\lambda \approx$ tens of nm















V. Zhukov, et al, Phys. Rev. B 73, 125105 (2006)





V. Zhukov, et al, Phys. Rev. B 73, 125105 (2006)





V. Zhukov, et al, Phys. Rev. B 73, 125105 (2006)













Boltzmann equation

$$\frac{\partial g}{\partial t} + \frac{1}{\hbar} \nabla_{\mathbf{r}} \cdot \left(g \nabla_{\mathbf{k}} \mathcal{E} \right) - \frac{e}{\hbar} \nabla_{\mathbf{k}} \cdot \left(g \left[\mathbf{E} + \frac{1}{e} \nabla_{\mathbf{r}} \mathcal{E} \right] \right) = \left(\frac{dg}{dt} \right)_{col} + \left(\frac{dg}{dt} \right)_{source} \left(\frac{dg}{dt} \right)_{source}$$

- Small perturbation of equilibrium
- Perfect screening (perfect metal)
- Spherically symmetric band structure
- Breaking of momentum conservation

Superdiffusive spin transport model

$$\frac{\partial n\left(\sigma, E, z, t\right)}{\partial t} + \frac{n\left(\sigma, E, z, t\right)}{\tau\left(\sigma, E, z\right)} = \left(-\frac{\partial}{\partial z}\hat{\phi} + \hat{I}\right)S^{\text{eff}}\left(\sigma, E, z, t\right)$$
$$\widetilde{[\Delta t]}\left(z, z_{0}\right) = \int_{z_{0}}^{z} \frac{dz'}{\tau\left(z'\right)} \quad ; \qquad \widetilde{[\Delta t/\tau]}\left(z, z_{0}\right) = \int_{z_{0}}^{z} \frac{dz'}{\tau\left(z'\right)\tau\left(z'\right)}$$
$$\hat{\phi}S^{\text{eff}} = \int_{-\infty}^{+\infty} dz_{0}\int_{-\infty}^{t} dt_{0}S^{\text{eff}}\left(\sigma, E, z_{0}, t_{0}\right) \frac{\widetilde{[\Delta t]}}{2\left(t - t_{0}\right)^{2}} e^{-\left(t - t_{0}\right)\left[\frac{\widetilde{[\Delta t]}}{[\Delta t]}\Theta\left[\left(t - t_{0}\right) - \left|\widetilde{[\Delta t]}\right|\right]}$$

Battiato, Carva, Oppeneer, Phys. Rev. Lett. 105, 027203 (2010)





Battiato, Carva, Oppeneer, Phys. Rev. Lett. 105, 027203 (2010)





Battiato, Carva, Oppeneer, Phys. Rev. Lett. 105, 027203 (2010)





Battiato, Carva, Oppeneer, Phys. Rev. Lett. 105, 027203 (2010)





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Melnikov et al. Phys. Rev, Lett. 107, 076601 (2011)

Rudolf, Battiato et al. Nature Comm. 3, 1038 (2012)

Eschenlohr, Battiato et al. Nature Mater. 12, 332 (2013)

Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)

Schellekens et al. Phys. Rev. B 90, 104429 (2014)





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Page 10/32
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Demagnetisation without direct laser excitation



Eschenlor, Battiato et al, Nature Mater 12, 332 (2013).





Demagnetisation without direct laser excitation





Eschenlor, Battiato et al, Nature Mater 12, 332 (2013).





Demagnetisation without direct laser excitation





Eschenlor, Battiato et al, Nature Mater 12, 332 (2013).



Ultrafast increase of magnetization



Rudolf, Battiato et al. Nature Comm. 3, 1038 (2012)





Ultrafast incre









Rudolf, Battiato et al. Nature Comm. 3, 1038 (2012)





Ultrafast increa



0.4



Ultrafast increase of magnetization

- Strong confirmation of ultrafast spin diffusion
- First observation of the ultrafast magnetization increase



Can we control these ultrafast spin currents?

Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)







Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)









Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)



Detection ultrashort spin currents

Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)



Detection ultrashort spin currents

Conversion spin current to charge current



Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)





Detection spin currents

- Conversion spin current to charge current
- Conversion charge current to THz radiation



Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)







Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)





- Control of ultrafast spin currents shape
- Identification mechanism of THz emission in UD
- Ultrafast spin Ampere-meter
- Broadband THz emitter (covering gap 5-10 THz)

Kampfrath, Battiato et al. Nature Nanotech. 8, 256 (2013)

Ultrafast spintronics

Wire

Diode



Wires: ultrafast spin injection in semiconductors

Ultrashort spin pulses diffuse in metals only up to few hundreds of nm





short mean free path



Wires: ultrafast spin injection in semiconductors



Battiato, Held, Phys. Rev. Lett. 116, 196601 (2016)



Wires: ultrafast spin injection in semiconductors



Battiato, Held, Phys. Rev. Lett. 116, 196601 (2016)



Ultrafast spin injection in semiconductors



Ultrafast spin injection in semiconductors



Battiato, Held, Phys. Rev. Lett. 116, 196601 (2016)

Ultrafast spin injection in semiconductors



Battiato, Held, Phys. Rev. Lett. 116, 196601 (2016)

Boltzmann + Maxwell system



Boltzmann + Maxwell system

Electronic population Electromagnetic field

 $f[t, \mathbf{x}, \mathbf{k}]$ $\mathbf{E}[t, \mathbf{x}] \quad \mathbf{B}[t, \mathbf{x}]$

$$\frac{\partial f}{\partial t} + \frac{1}{\hbar} \nabla_{\mathbf{k}} \mathcal{E} \cdot \nabla_{\mathbf{k}} f + \frac{e}{\hbar} \mathbf{E} \cdot \nabla_{\mathbf{k}} f - \left(\frac{\partial f}{\partial t}\right)_{\mathrm{sc}} - \left(\frac{\partial f}{\partial t}\right)_{\mathrm{ex}} = 0$$

$$\nabla \cdot \epsilon \mathbf{E} = \rho$$
$$\nabla \cdot \mathbf{B} = 0$$
$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$
$$\nabla \times \frac{\mathbf{B}}{\mu} = \mathbf{j} + \frac{\partial \epsilon \mathbf{E}}{\partial t}$$

$$\begin{split} \rho[t,\mathbf{x}] &= -\frac{e}{(2\pi)^3} \int f[t,\mathbf{x},\mathbf{k}] d\mathbf{k} \\ \mathbf{j}[t,\mathbf{x}] &= -\frac{e}{(2\pi)^3 \hbar} \int \frac{\partial \mathcal{E}}{\partial \mathbf{k}} f[t,\mathbf{x},\mathbf{k}] d\mathbf{k} \end{split}$$

Boltzmann equation

$$\frac{\partial f}{\partial t} + \left(\frac{1}{\hbar} \nabla_{\mathbf{k}} \mathcal{E} \cdot \nabla_{\mathbf{x}} f + \frac{e}{\hbar} \mathbf{E} \cdot \nabla_{\mathbf{k}} f\right) - \left(\frac{\partial f}{\partial t}\right)_{\mathrm{SC}} - \left(\frac{\partial f}{\partial t}\right)_{\mathrm{ex}} = 0$$

1+1+3 dimensional problem: not cheap, but can be handled

Runge Kutta Discontinuous Galerkin

- Unstructured meshes
- Arbitrarily high order of convergence (depending on max polynomial power)
- Mass, energy and momentum conserving
- High stability in presence of shocks



Boltzmann equation



$$\begin{split} S_{ijklm} &= \sum_{\mathbf{G}} \int d\mathbf{k} d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \\ & \left(w(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \cdot \\ & \delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3 + \mathbf{G}) \ \delta(\mathcal{E}(\mathbf{k}) + \mathcal{E}(\mathbf{k}_1) - \mathcal{E}(\mathbf{k}_2) - \mathcal{E}(\mathbf{k}_3)) \cdot \\ & b_i(\mathbf{k}) b_j(\mathbf{k}) b_k(\mathbf{k}_1) b_l(\mathbf{k}_2) b_m(\mathbf{k}_3) \right) \end{split}$$

$$\begin{split} \mathcal{S}_{ijklm} &= \sum_{\mathbf{G}} \int d\mathbf{k} d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \\ & \left(w(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \cdot \\ & \delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3 + \mathbf{G}) \ \delta(\mathcal{E}(\mathbf{k}) + \mathcal{E}(\mathbf{k}_1) - \mathcal{E}(\mathbf{k}_2) - \mathcal{E}(\mathbf{k}_3)) \cdot \\ & b_i(\mathbf{k}) b_j(\mathbf{k}) b_k(\mathbf{k}_1) b_l(\mathbf{k}_2) b_m(\mathbf{k}_3) \right) \end{split}$$

5 dimensional tensor: number of entries is N⁵
 (400 basis functions -> 10¹³ integrals to calculate and 10TB to store)

$$S_{ijklm} = \sum_{\mathbf{G}} \int d\mathbf{k} d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3$$
$$\begin{pmatrix} w(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \\ \delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3 + \mathbf{G}) \ \delta(\mathcal{E}(\mathbf{k}) + \mathcal{E}(\mathbf{k}_1) - \mathcal{E}(\mathbf{k}_2) - \mathcal{E}(\mathbf{k}_3)) \\ b_i(\mathbf{k}) b_j(\mathbf{k}) b_k(\mathbf{k}_1) b_l(\mathbf{k}_2) b_m(\mathbf{k}_3) \end{pmatrix}$$

- 5 dimensional tensor: number of entries is N⁵
 (400 basis functions -> 10¹³ integrals to calculate and 10TB to store)
- 12 dimensional integral

$$\begin{split} S_{ijklm} &= \sum_{\mathbf{G}} \int d\mathbf{k} d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \\ & \begin{pmatrix} w(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \cdot \\ \delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3 + \mathbf{G}) \ \delta(\mathcal{E}(\mathbf{k}) + \mathcal{E}(\mathbf{k}_1) - \mathcal{E}(\mathbf{k}_2) - \mathcal{E}(\mathbf{k}_3)) \\ b_i(\mathbf{k}) b_j(\mathbf{k}) b_k(\mathbf{k}_1) b_l(\mathbf{k}_2) b_m(\mathbf{k}_3) \end{pmatrix} \end{split}$$

- 5 dimensional tensor: number of entries is N⁵
 (400 basis functions -> 10¹³ integrals to calculate and 10TB to store)
- 12 dimensional integral
- Dirac deltas inside the integral

$$\begin{split} S_{ijklm} &= \sum_{\mathbf{G}} \int d\mathbf{k} d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \\ & \left(w(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \cdot \\ & \delta(\mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}_3 + \mathbf{G}) \ \delta(\mathcal{E}(\mathbf{k}) + \mathcal{E}(\mathbf{k}_1) - \mathcal{E}(\mathbf{k}_2) - \mathcal{E}(\mathbf{k}_3)) \cdot \\ & b_i(\mathbf{k}) b_j(\mathbf{k}) b_k(\mathbf{k}_1) b_l(\mathbf{k}_2) b_m(\mathbf{k}_3) \right) \end{split}$$

- 5 dimensional tensor: number of entries is N⁵
 (400 basis functions -> 10¹³ integrals to calculate and 10TB to store)
- 12 dimensional integral
- Dirac deltas inside the integral
- Failure to integrate to extreme precision leads to breaking of particle, energy and momentum conservation

Thermalization in real band structures

$$\begin{split} \left(\frac{\partial f}{\partial t}\right)_{col}(t, \mathbf{x}, \mathbf{k}) &= \\ \int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \ W(\mathbf{x}, \mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \Big[\left(1 - f(t, \mathbf{x}, \mathbf{k})\right) \left(1 - f(t, \mathbf{x}, \mathbf{k}_1)\right) f(t, \mathbf{x}, \mathbf{k}_2) f(t, \mathbf{x}, \mathbf{k}_3) + \\ &- f(t, \mathbf{x}, \mathbf{k}) f(t, \mathbf{x}, \mathbf{k}_1) \left(1 - f(t, \mathbf{x}, \mathbf{k}_2)\right) \left(1 - f(t, \mathbf{x}, \mathbf{k}_3)\right) \Big] \end{split}$$



Michael Wais

Check his poster out!!!



Thermalization in real band structures

$$\begin{pmatrix} \frac{\partial f}{\partial t} \end{pmatrix}_{col} (t, \mathbf{x}, \mathbf{k}) = \\ \int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{k}_3 \ W(\mathbf{x}, \mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \Big[(1 - f(t, \mathbf{x}, \mathbf{k})) (1 - f(t, \mathbf{x}, \mathbf{k}_1)) f(t, \mathbf{x}, \mathbf{k}_2) f(t, \mathbf{x}, \mathbf{k}_3) + \\ - f(t, \mathbf{x}, \mathbf{k}) f(t, \mathbf{x}, \mathbf{k}_1) (1 - f(t, \mathbf{x}, \mathbf{k}_2)) (1 - f(t, \mathbf{x}, \mathbf{k}_3)) \Big]$$





Future plans / Areas of interest













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THANK YOU

Financial support from: FWF Lise Meitner grant M1925-N28

Der Wissenschaftsfonds.

UIF

K. Holldack

R. Mitzner

A. Föhlisch

S. Mathias

P. Grychtol

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Multiple PhD and PostDoc positions

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NAP Award (1M S\$ + PhD's)







QS World ranking

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 11th World Ranking
 3rd Engineering and Technology
 7th Materials Science



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- Physics (theory)
- Mathematics
- Electronics



- Ultrafast dynamics
- Ab initio methods
- Boltzmann equation
- Discontinuous Galerkin