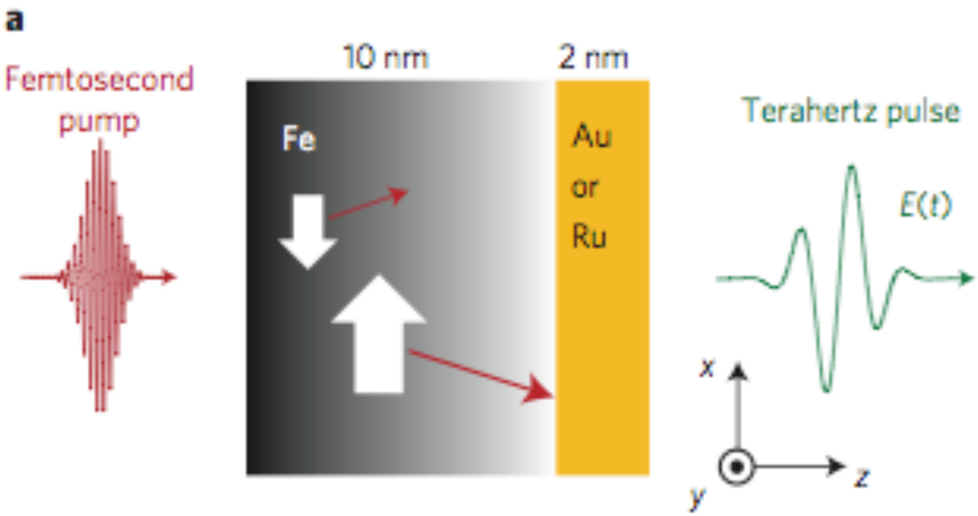
PHOTOCURRENTS IN MULTILAYERS FOR ULTRAFAST SPINORBITRONICS

25.10.2018 I FRANK FREIMUTH

JÜLICH
Forschungszentrum

MOTIVATION



Superdiffusive spin-current

↓

Inverse spin-Hall effect

↓

Interfacial charge current

T. Kampfrath et al., Nature Nanotechnology **8**, 256 (2013)

Are there additional different mechanisms for photocurrent generation at magnetic bilayer interfaces?

LASER-INDUCED CURRENTS IN THE MAGNETIC RASHBA MODEL

Magnetic Rashba model: $$H = \frac{-\hbar^2}{2m_e}\Delta - i\alpha(\boldsymbol{\nabla}\times\hat{\boldsymbol{e}}_z)\cdot\boldsymbol{\sigma} + \frac{\Delta V}{2}\boldsymbol{\sigma}\cdot\hat{\boldsymbol{n}}_{\mathrm{c}}(\boldsymbol{r})$$

No superdiffusive spin-current in the magnetic Rashba model!

Laser-induced charge current: $$J_\alpha = \frac{a_0^2 eI}{\hbar c}\left(\frac{\mathcal{E}_{\mathrm{H}}}{\hbar\omega}\right)^2 \mathrm{Im}\sum_{\beta\gamma}\epsilon_\beta\epsilon_\gamma^*\varphi_{\alpha\beta\gamma}$$

General:
$$\begin{aligned}\chi^{\mathcal{O}}_{\beta\gamma} = &\frac{2}{a_0\mathcal{E}_{\mathrm{H}}}\int\frac{\mathrm{d}^2k}{(2\pi)^2}\int \mathrm{d}\mathcal{E}\,\mathrm{Tr}\Big[\\
&f(\mathcal{E})\mathcal{O}G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})v_\beta G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E}-\hbar\omega)v_\gamma G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})\\
&-f(\mathcal{E})\mathcal{O}G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})v_\beta G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E}-\hbar\omega)v_\gamma G^{\mathrm{A}}_{\boldsymbol{k}}(\mathcal{E})\\
&+f(\mathcal{E})\mathcal{O}G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})v_\gamma G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E}+\hbar\omega)v_\beta G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})\\
&-f(\mathcal{E})\mathcal{O}G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})v_\gamma G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E}+\hbar\omega)v_\beta G^{\mathrm{A}}_{\boldsymbol{k}}(\mathcal{E})\\
&+f(\mathcal{E}-\hbar\omega)\mathcal{O}G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})v_\beta G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E}-\hbar\omega)v_\gamma G^{\mathrm{A}}_{\boldsymbol{k}}(\mathcal{E})\\
&+f(\mathcal{E}+\hbar\omega)\mathcal{O}G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E})v_\gamma G^{\mathrm{R}}_{\boldsymbol{k}}(\mathcal{E}+\hbar\omega)v_\beta G^{\mathrm{A}}_{\boldsymbol{k}}(\mathcal{E})\Big]\end{aligned}$$

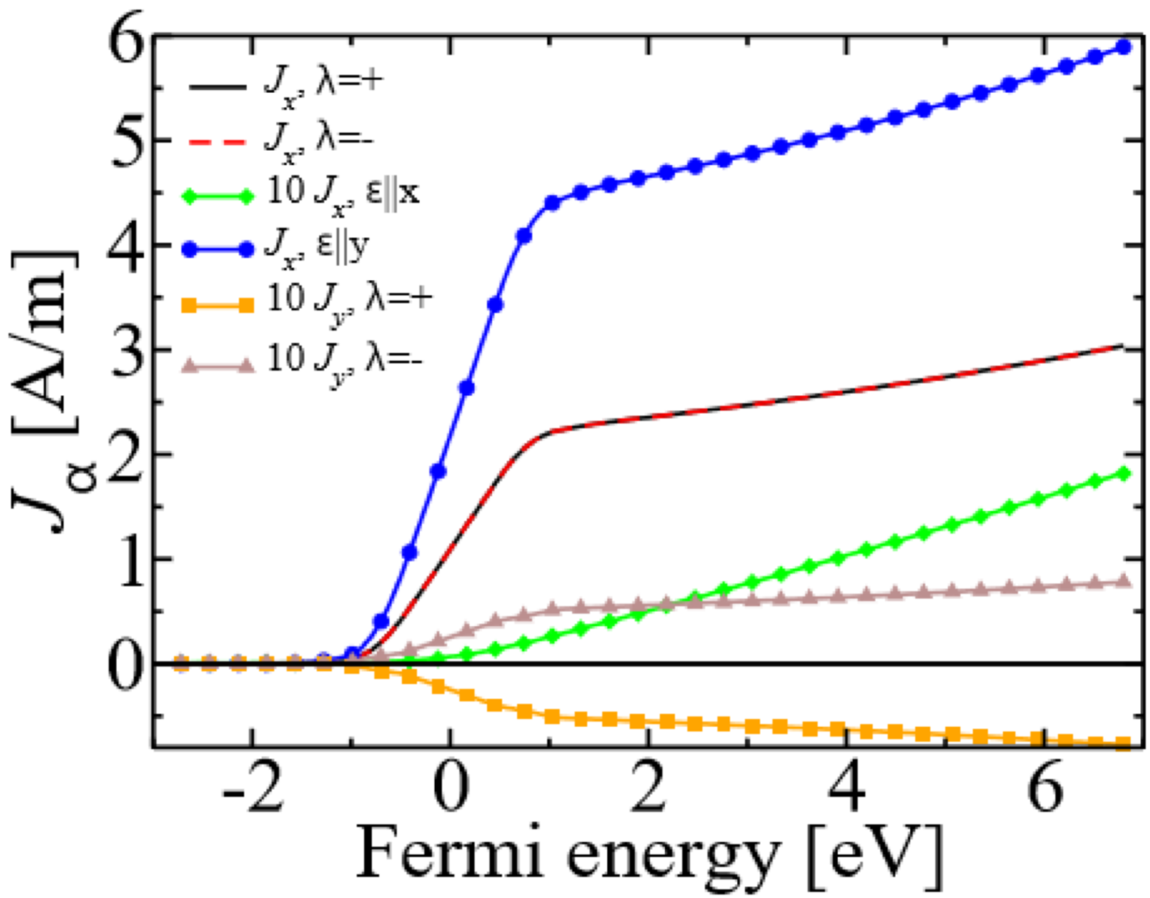
For charge current: $$\varphi_{\alpha\beta\gamma} = \chi^{v_\alpha}_{\beta\gamma}$$

F. Freimuth et al., arXiv:1710.10480

LASER-INDUCED CHARGE CURRENT

Small SOI, magnetization in y direction

$\alpha^{\mathrm{R}} = 0.1\mathrm{eV\AA}$ $\Delta V = 1\ \mathrm{eV}$ $\Gamma = 25\mathrm{meV}$



J_x: helicity-independent, polarization-dependent: Maximal for ε||y

J_y: helicity-dependent

J_x: helicity-even like superdiffusive+ISHE-mechanism

F. Freimuth et al., arXiv:1710.10480

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SYMMETRY ANALYSIS FOR BILAYER GEOMETRY

TABLE I: Symmetry properties of the magnetic photogalvanic effect in the ferromagnetic Rashba model with magnetization parallel to the y axis. $\emptyset$ means no effect. M_y means odd in magnetization, i.e., the effect changes sign when the magnetization is antiparallel to the y axis. λM_y means odd in the light helicity and odd in the magnetization. $|\lambda| M_y$ means even in the light helicity and odd in the magnetization.

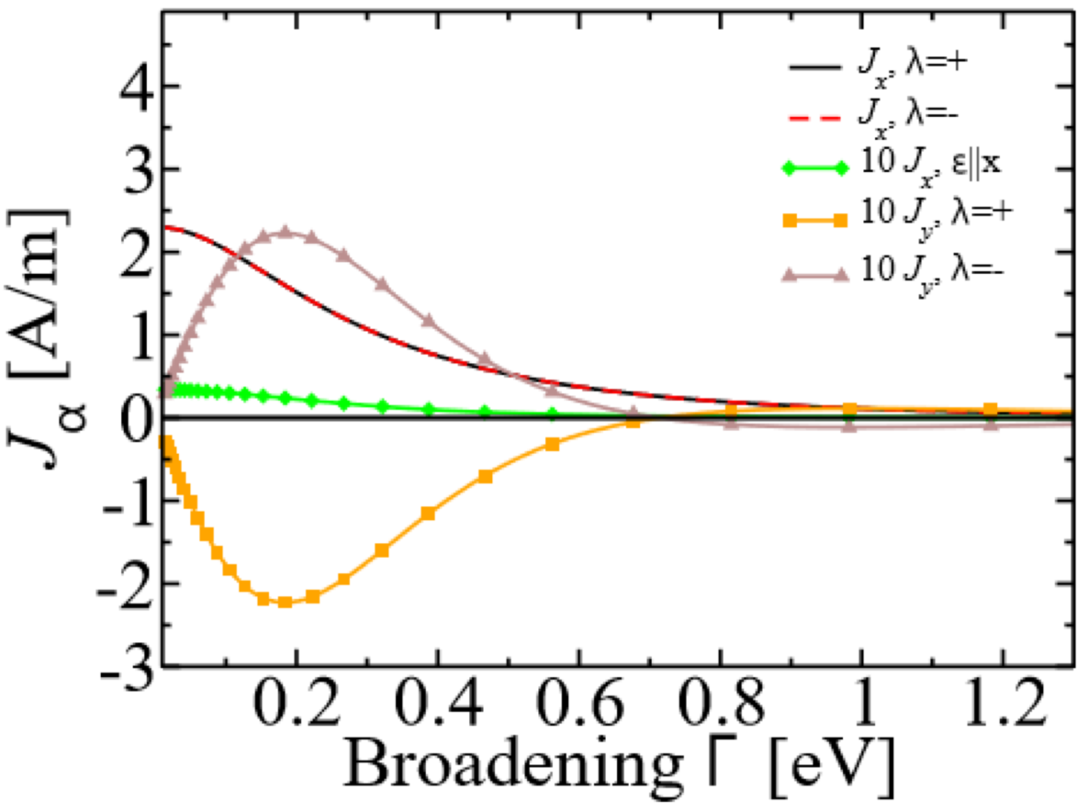
	circularly polarized	linearly polarized ($\epsilon \| x$ or $\epsilon \| y$)
J_x	$\|\lambda\| M_y$	M_y
J_y	λM_y	$\emptyset$

F. Freimuth et al., arXiv:1710.10480

LASER-INDUCED CHARGE CURRENT

Small SOI: Dependence on lifetime-broadening Γ

$$\alpha^{\mathrm{R}} = 0.1\mathrm{eV\AA} \qquad \Delta V = 1\ \mathrm{eV} \qquad \mathcal{E}_{\mathrm{F}} = 1.36\mathrm{eV}$$



J_x: Monotonic decrease

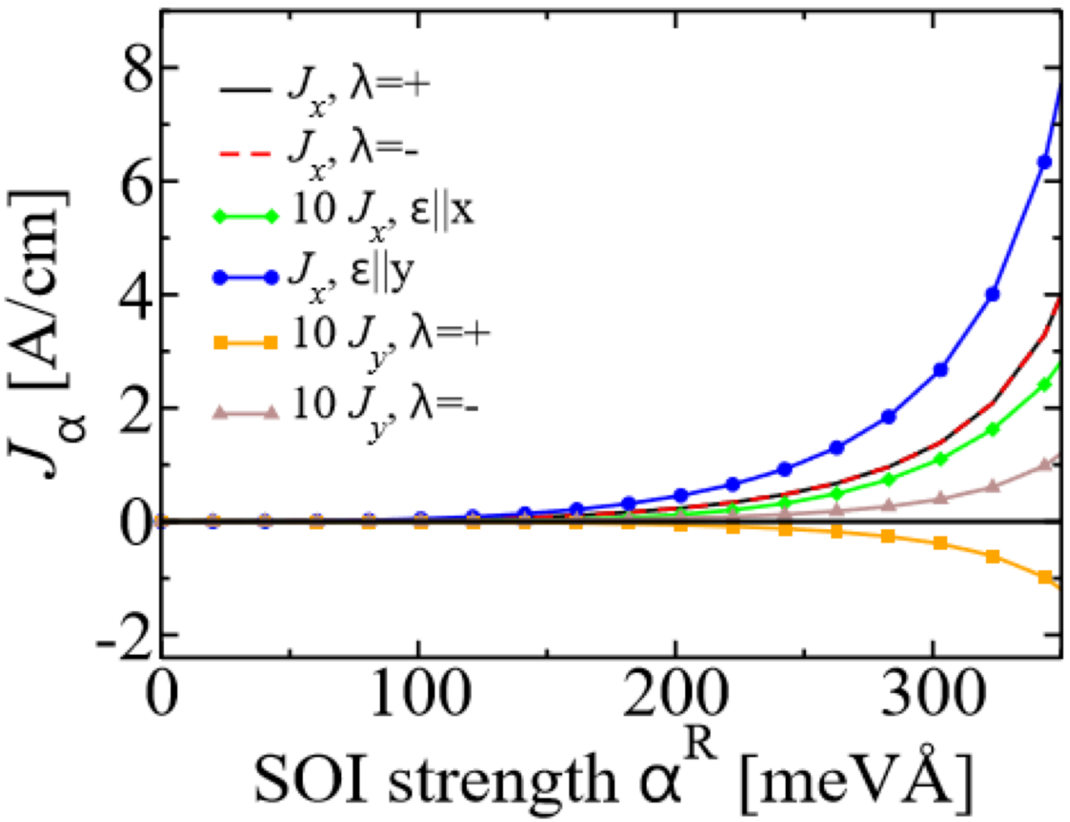
J_y: Maximum for optimal broadening when circularly polarized light is used
→ Can be optimized by disorder

F. Freimuth et al., arXiv:1710.10480

LASER-INDUCED CHARGE CURRENT

Dependence on SOI-Strength

$\Delta V = 1 \text{ eV}$ $\mathcal{E}_F = 1.36\text{eV}$ $\Gamma = 25 \text{ meV}$



Strong increase with SOI strength → look for it in giant Rashba systems, e.g., magnetically doped TI

F. Freimuth et al., arXiv:1710.10480

LASER-INDUCED SPIN CURRENTS

Symmetry analysis for nonmagnetic Rashba model

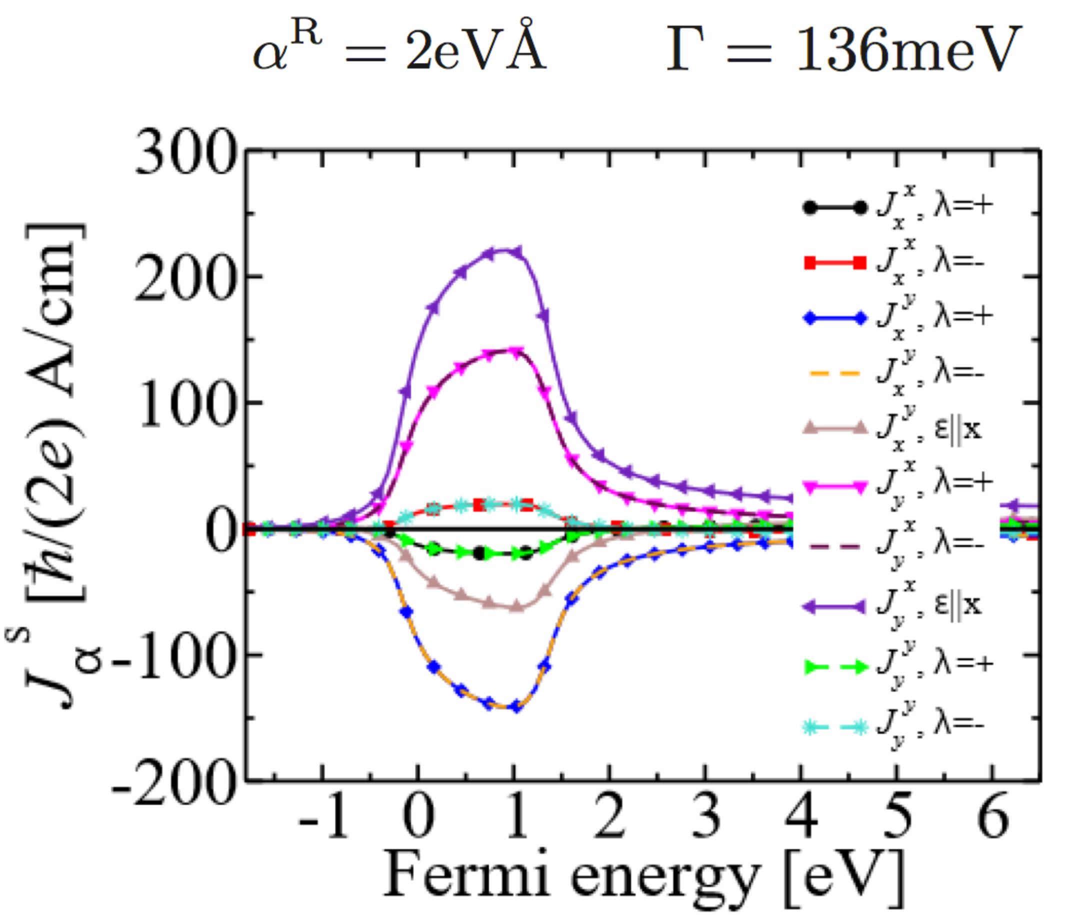
TABLE III: Symmetry properties of the laser-induced spin-current density in the nonmagnetic Rashba model. $\emptyset$ means there is no effect. ✓ means there is an effect. λ means the effect is odd in the helicity of light. $|\lambda|$ means the effect is even in the helicity of light.

	circularly polarized	linearly polarized
J_x^x	λ	$\emptyset$
J_x^y	$\|\lambda\|$	✓
J_x^z	$\emptyset$	$\emptyset$
J_y^x	$\|\lambda\|$	✓
J_y^y	λ	$\emptyset$
J_y^z	$\emptyset$	$\emptyset$

F. Freimuth et al., arXiv:1710.10480

LASER-INDUCED SPIN CURRENT

Symmetry analysis for nonmagnetic Rashba model



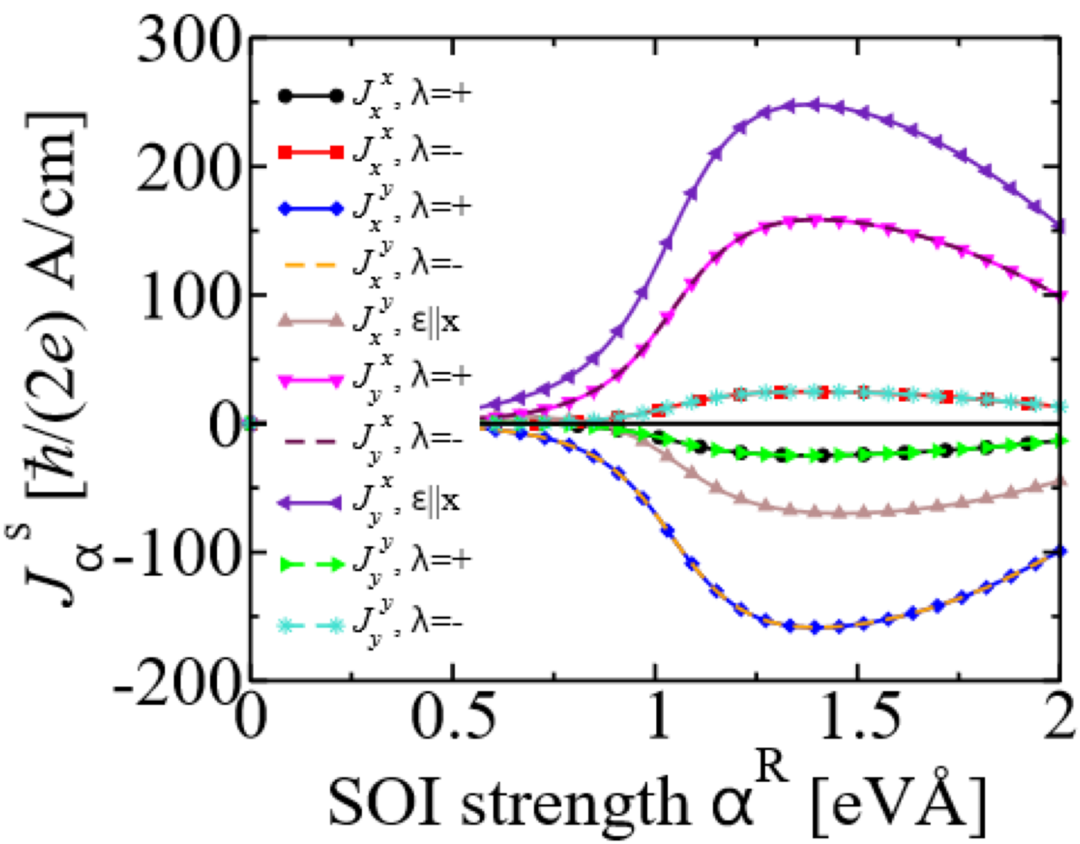
All symmetry-allowed components are present. Components with spin parallel to current (J_x^x, J_y^y) are smaller than the other components.

F. Freimuth et al., arXiv:1710.10480

LASER-INDUCED SPIN CURRENT

Nonmagnetic Rashba model: Dependence on SOI strength

$$\mathcal{E}_{\mathrm{F}} = 1.36\mathrm{eV} \qquad \Gamma = 136\mathrm{meV}$$



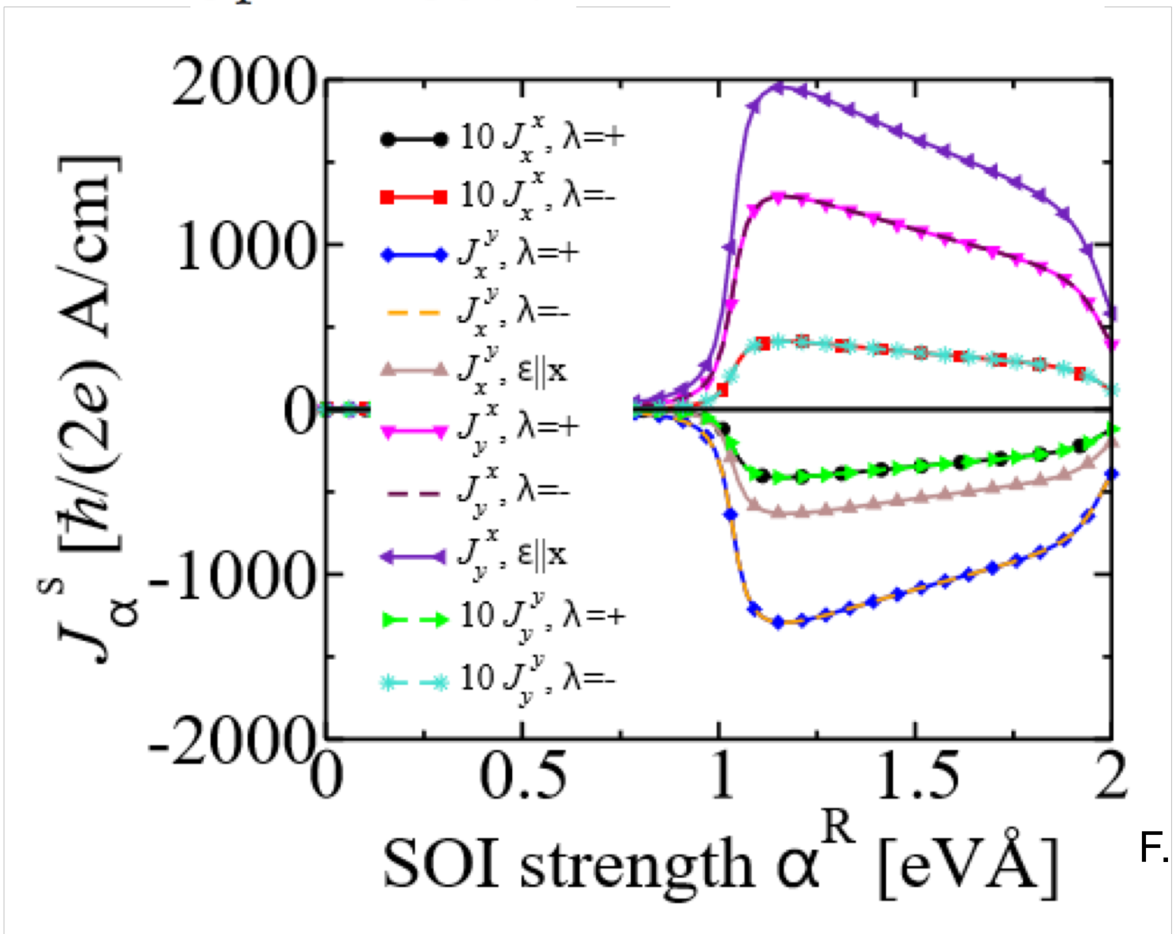
Strong increase with SOI strength → look for it in giant Rashba systems

F. Freimuth et al., arXiv:1710.10480

LASER-INDUCED SPIN CURRENT

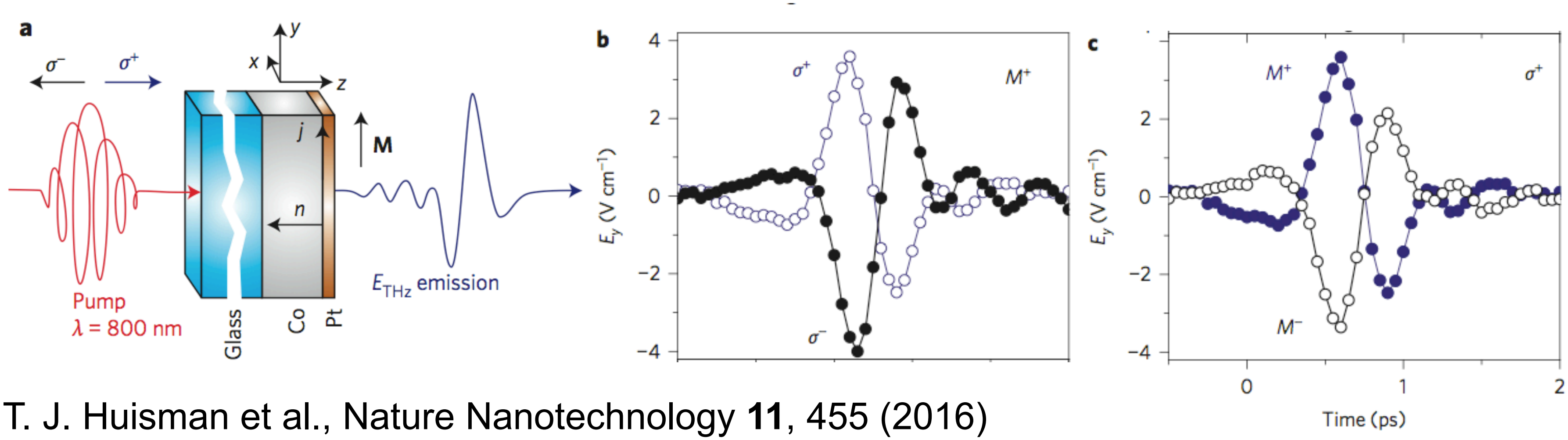
Even larger spin currents for small quasiparticle broadening

$\mathcal{E}_F = 1.36\text{eV} \quad \Gamma = 25 \text{ meV}$



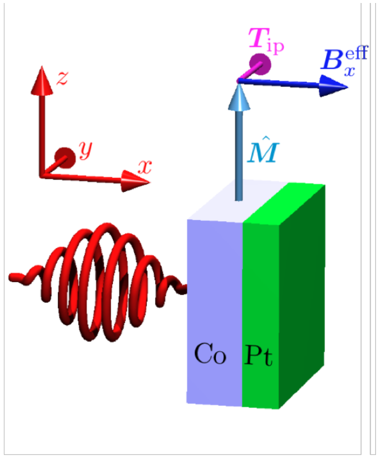
F. Freimuth et al., arXiv:1710.10480

PHOTOCURRENTS FROM LASER-INDUCED MAGNETIZATION DYNAMICS

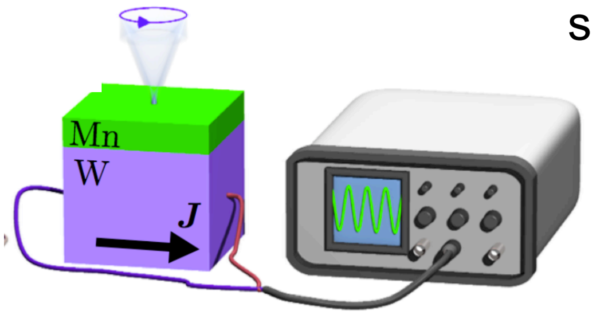


T. J. Huisman et al., Nature Nanotechnology **11**, 455 (2016)

Experimental observation can be explained in terms of IFE and ISOT:



Inverse Faraday effect (IFE)

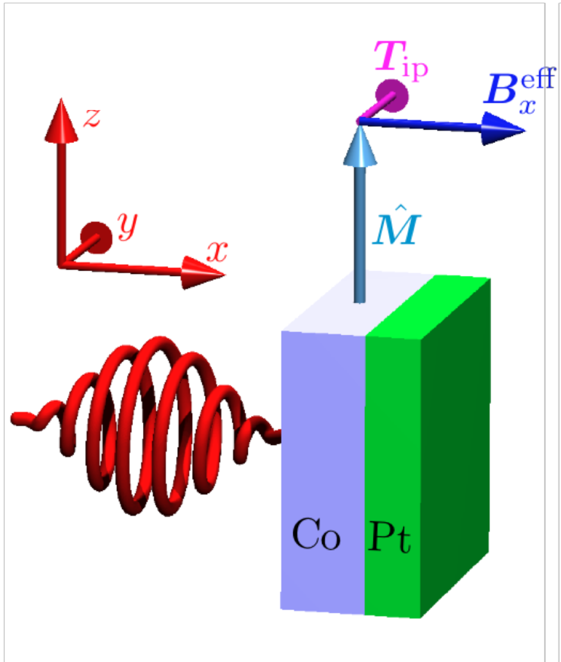


Inverse spin-orbit torque (ISOT)

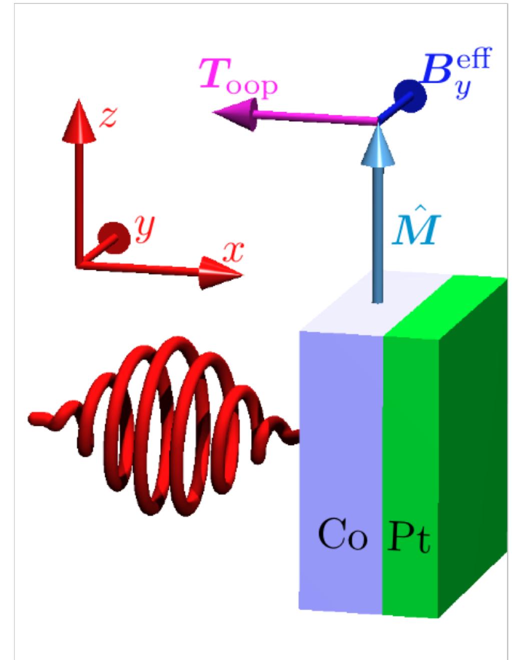
• Experiment: 50 fs pulse with 20GW/cm^2 (fluence 1mJ/cm^2) → IFE of 0.2 Tesla

Determine laser-induced torques from *ab-initio* (perpendicular IFE field)

LASER-INDUCED TORQUES FROM IFE & OSTT



Inverse Faraday effect (IFE)



Optical spin-transfer torque (OSTT)

$$H(\boldsymbol{r}) = H_0(\boldsymbol{r}) - \boldsymbol{m} \cdot \hat{\boldsymbol{M}}\Omega^{\mathrm{xc}}(\boldsymbol{r})$$

$$\boldsymbol{\mathcal{T}}(\boldsymbol{r}) = \boldsymbol{m} \times \hat{\boldsymbol{M}}\Omega^{\mathrm{xc}}(\boldsymbol{r})$$

$$\boldsymbol{T} = i\mathrm{Tr}\left[\boldsymbol{\mathcal{T}} G^{<}\right]$$

Parameters used in the calculation

- DFT plus Keldysh formalism
- Laser intensity is set to 10GW/cm^2
- Photon energy is 1.55 eV
- Assume continuous laser beam

in the calculation

F. Freimuth, S.Blügel and Y. Mokrousov, PRB **94**,144432 (2016)

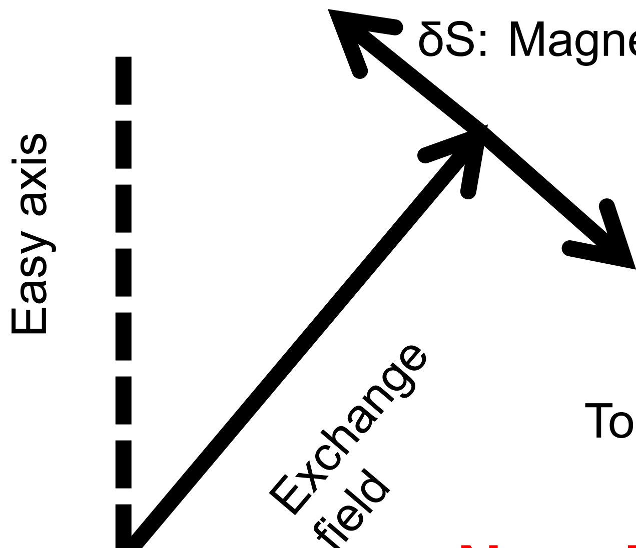
TORQUE OPERATOR

Magnetization direction

Exchange field

Kohn Sham Hamiltonian: $H(\boldsymbol{r}) = H_0(\boldsymbol{r}) - \boldsymbol{m} \cdot \hat{M}\Omega^{\mathrm{xc}}(\boldsymbol{r})$

Spin magnetization



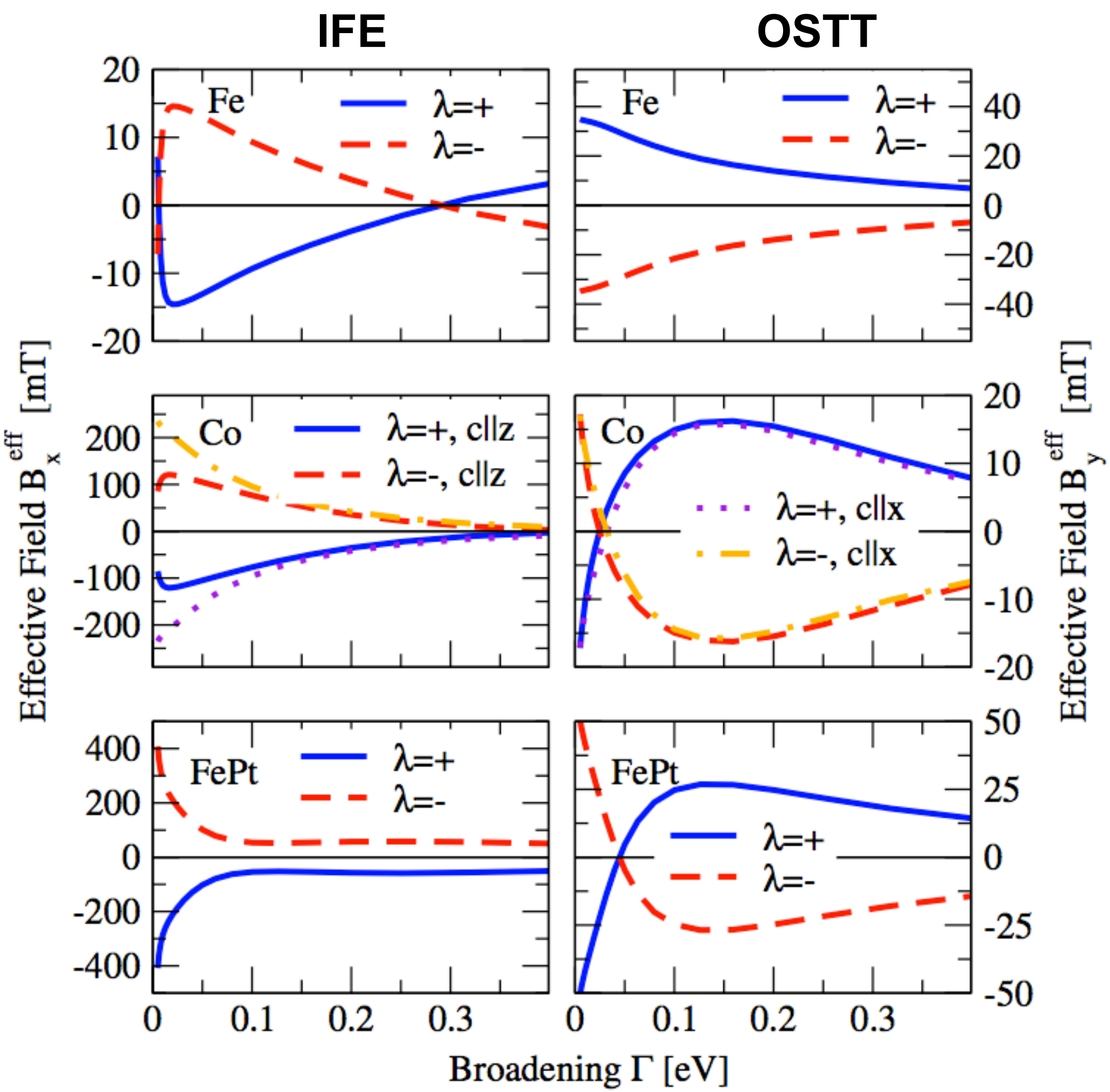
Torque: $\boldsymbol{T} = \frac{2\mu_{\mathrm{B}}}{\hbar} \int \mathrm{d}^3 r\, \Omega^{\mathrm{xc}}(\boldsymbol{r}) \delta \boldsymbol{s}(\boldsymbol{r}) \times \hat{\boldsymbol{M}}$

No orbital angular momentum in the torque

F. Freimuth, S. Blügel and Y. Mokrousov, PRB **90**, 174423 (2014)

F. Freimuth, S. Blügel and Y. Mokrousov, PRB **92**, 064415 (2015)

LASER-INDUCED EFF. MAGNETIC FIELDS



Effective magnetic field

$$\boldsymbol{B}^{\text{eff}} = \frac{\boldsymbol{T} \times \hat{\boldsymbol{M}}}{\mu}$$

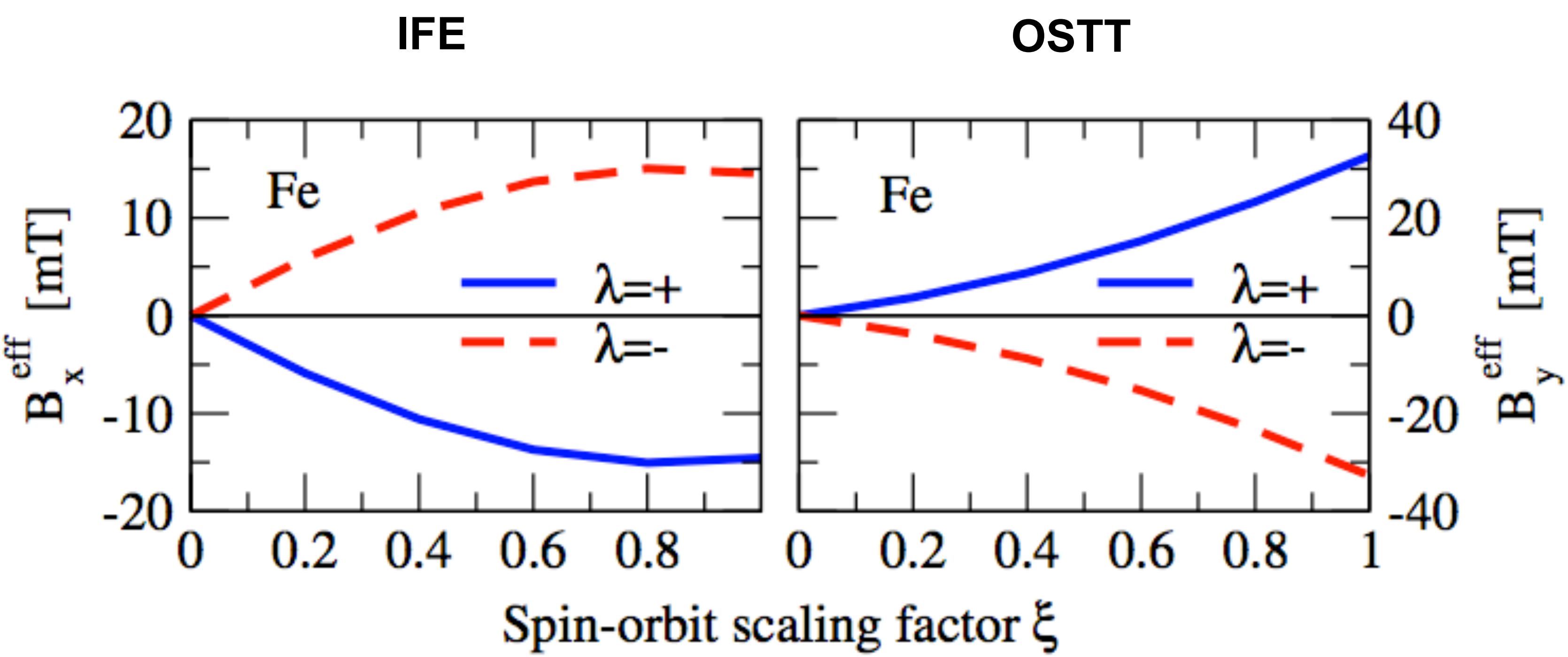
Magnetic moment

- odd in helicity λ
- OSTT dominates in Fe
- IFE dominates in FePt
- In Co IFE dominates for small and medium broadenings

Co: 2 x Experiment

F. Freimuth, S.Blügel and Y. Mokrousov, PRB **94**,144432 (2016)

DEPENDENCE ON SPIN-ORBIT STRENGTH

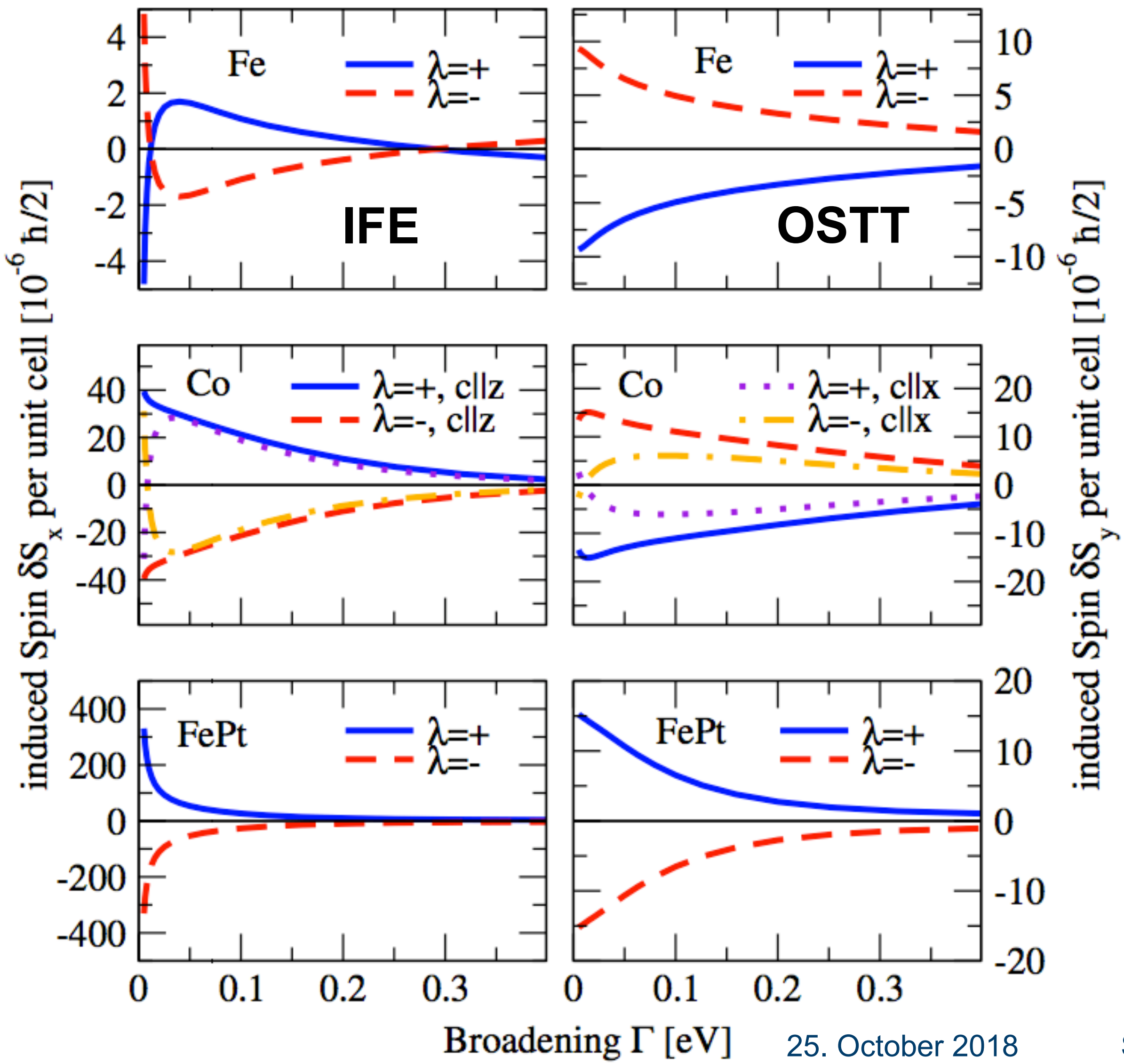


No laser-induced torque without spin-orbit interaction (in collinear ferromagnets)

F. Freimuth, S.Blügel and Y. Mokrousov, PRB **94**,144432 (2016)

LASER-INDUCED PERPENDICULAR SPIN

$$\delta \boldsymbol{S} = \int \mathrm{d}^3 r \, \delta \boldsymbol{s}(\boldsymbol{r}) = \frac{\hbar}{2i} \mathrm{Tr} \left[\boldsymbol{\sigma} G^{<} \right]$$



- odd in helicity λ
- no perfect correlation with B^{eff}

F. Freimuth et al., PRB **94**,144432 (2016)

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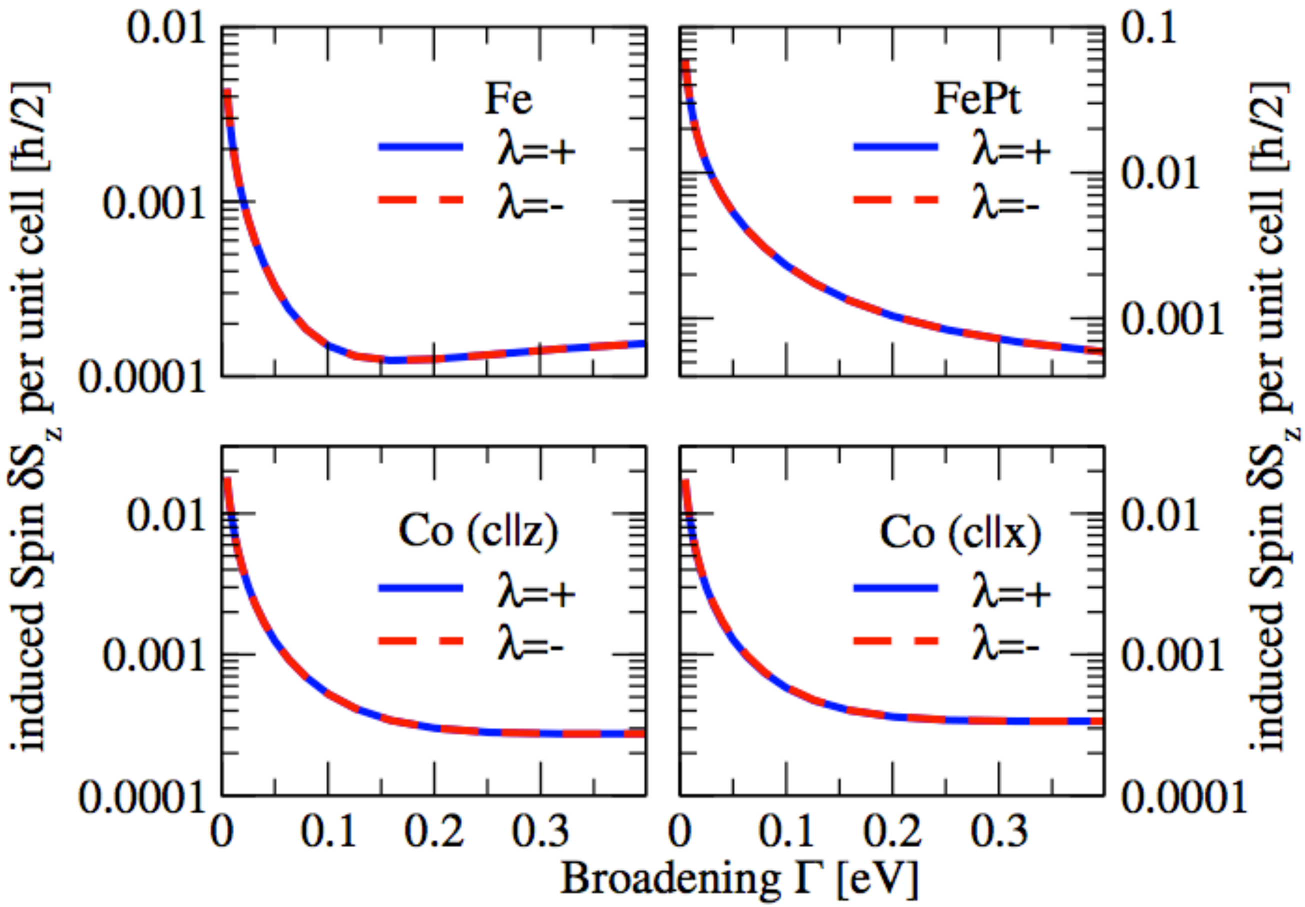
LASER-INDUCED PARALLEL SPIN



- even in helicity λ
- much larger than δS_x and δS_y

F. Freimuth, S.Blügel and Y. Mokrousov, PRB **94**,144432 (2016)

TRANSIENTS IMPORTANT?

- Experiment: 50 fs laser pulse leads to electric current with rise time 330 fs

→ Limitation of detector bandwidth might matter

→ Theory of time-dependent IFE might be necessary (transient response)

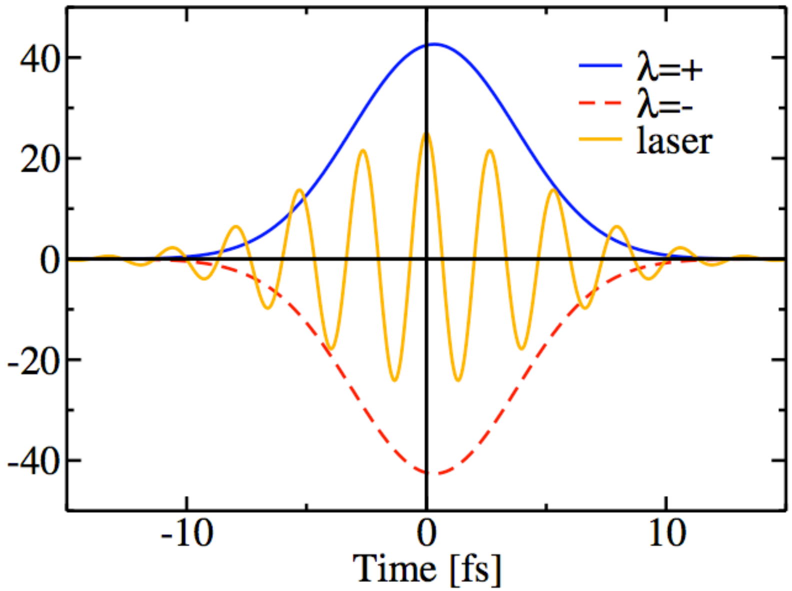
SPIN INDUCED BY LASER PULSE

$$\boldsymbol{E}(t) = \mathrm{Re}\left[E_0 \boldsymbol{\epsilon} \frac{\exp\left[\frac{-t^2}{2\Delta^2}\right]}{\Delta\sqrt{2\pi}} e^{-i\omega_0 t}\right]$$

Full width at half maximum (FWHM): $2\sqrt{2\ln 2}\Delta$

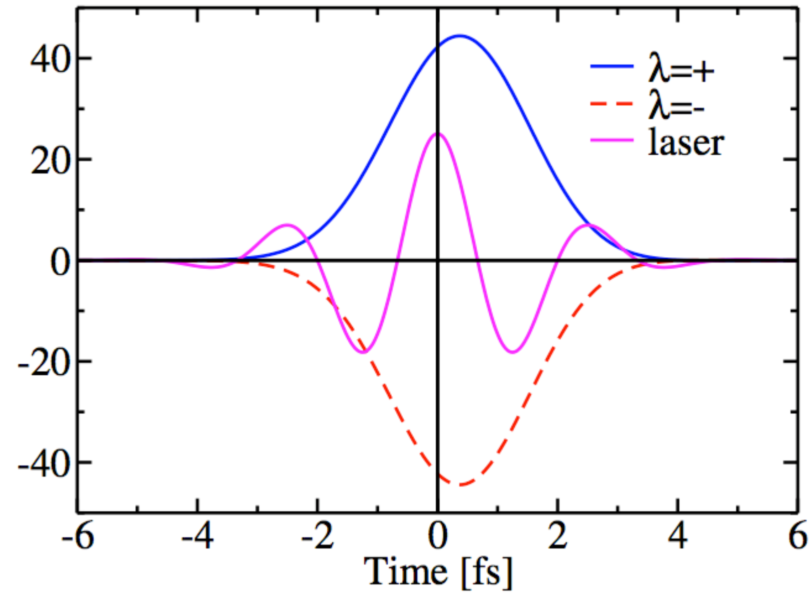
Nonmagnetic Rashba model: $H = \frac{-\hbar^2}{2m_e}\Delta - i\alpha(\boldsymbol{\nabla} \times \hat{e}_z) \cdot \boldsymbol{\sigma}$

Pulse: 11.4 fs (FWHM)



Response: 8 fs (FWHM)

Pulse: 3.8 fs (FWHM)



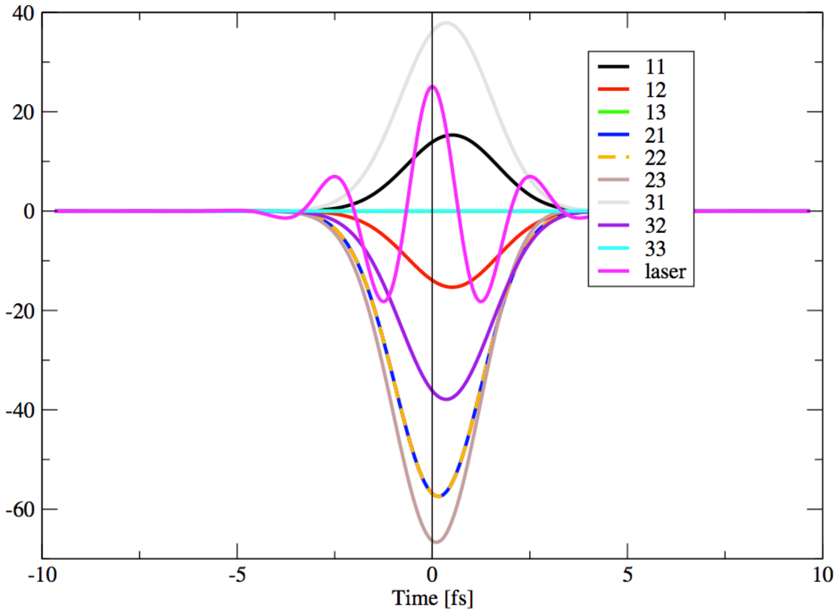
Response is retarded

SPIN INDUCED BY LASER PULSES

Magnetic Rashba model: $H = \frac{-\hbar^2}{2m_e}\Delta - i\alpha(\nabla \times \hat{e}_z)\cdot\boldsymbol{\sigma} + \frac{\Delta V}{2}\boldsymbol{\sigma}\cdot\hat{\boldsymbol{n}}_c(\boldsymbol{r})$

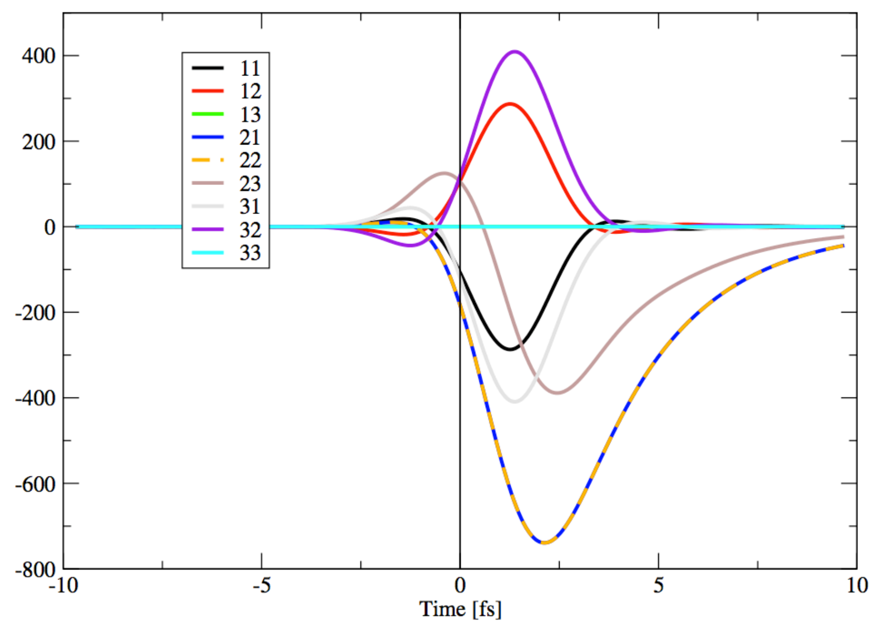
Magnetization in y direction

Pulse: 3.8 fs (FWHM)



Large lifetime broadening Γ=1.36 eV

Pulse: 3.8 fs (FWHM)



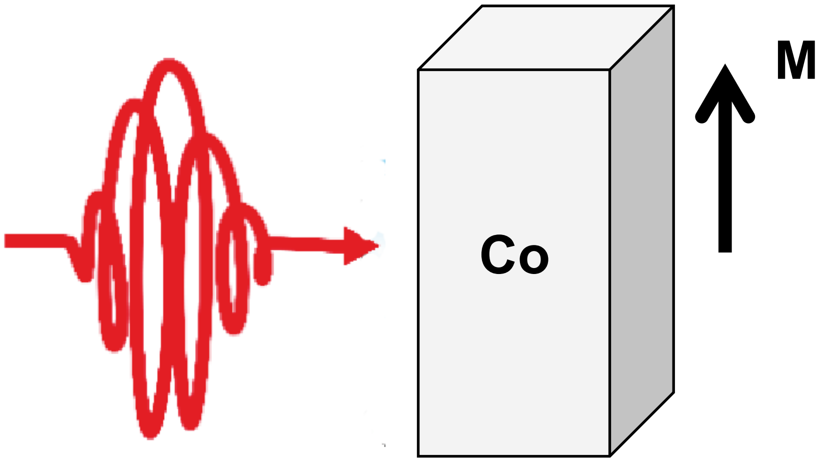
Small lifetime broadening Γ=13.6 meV

Strong prolongation of spin induced in y-direction.

≠ Experimental observation: Prolongation of IFE in z-direction

But: Only model without d-states → Need for *ab-initio*

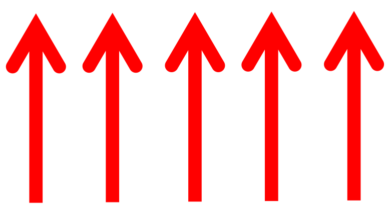
LASER-INDUCED ULTRAFAST DEMAGNETIZATION



What happens with the exchange splitting?

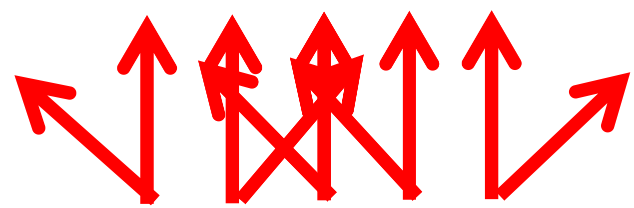
What happens with the local atomic exchange field?

Scenario 1



Reduction of local atomic magnetic moments
→ Collapse of local exchange field

Scenario 2



Transverse fluctuations

Does ultrafast demagnetization induce electric currents?

RESPONSE OF ELECTRONS TO COLLAPSING LOCAL EXCHANGE FIELD

$$H(\mathbf{r},t) = H_0(\mathbf{r}) - \mathbf{m}\cdot\hat{\mathbf{M}}(t)\Omega^{\mathrm{xc}}(\mathbf{r})$$

ISOT: Exchange field $\Omega^{\mathrm{xc}}(\mathbf{r})$ constant, magnetization direction precesses

ISOT current: $$j_\alpha = \sum_\beta \frac{e}{V}\lim_{\omega\to 0}\frac{\mathrm{Im}G^{\mathrm{R}}_{v_\alpha,\mathcal{T}_\beta}(\hbar\omega,\hat{\mathbf{M}})}{\hbar\omega}\left(\hat{\mathbf{M}}\times\frac{d\hat{\mathbf{M}}}{dt}\right)_\beta$$

Now: Magnetization direction constant, but exchange field collapses

$$H(\mathbf{r},t) = H_0(\mathbf{r}) - \mathbf{m}\cdot\hat{\mathbf{M}}\Omega^{\mathrm{xc}}(\mathbf{r},t)$$

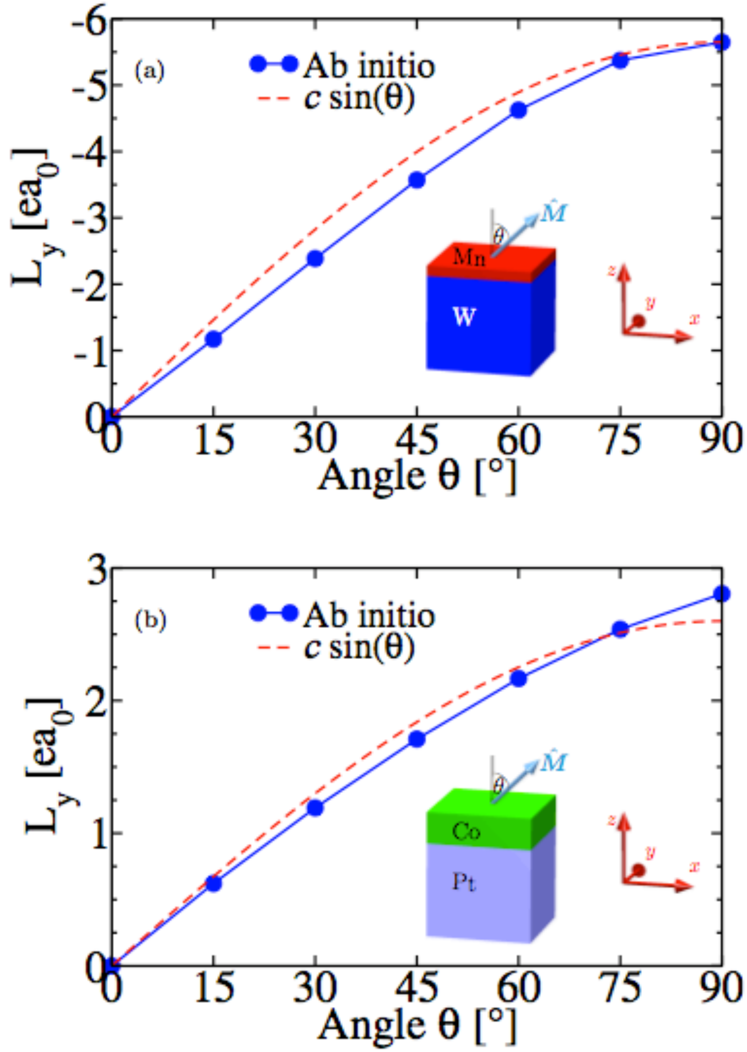
Ansatz: $$\Omega^{\mathrm{xc}}(\mathbf{r},t) = \Omega^{\mathrm{xc}}(\mathbf{r})\eta(t)$$

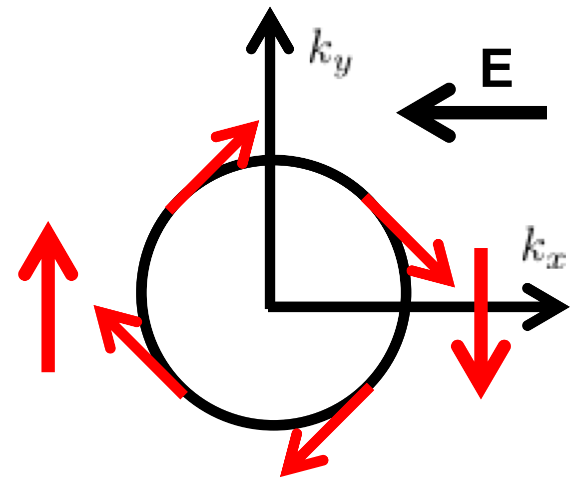
Demagnetization-induced current: $$j_\alpha = -\frac{e}{V}\lim_{\omega\to 0}\frac{\mathrm{Im}G^{\mathrm{R}}_{v_\alpha,\Omega^{\mathrm{xc}}m_\parallel}(\hbar\omega,\hat{\mathbf{M}})}{\hbar\omega}\frac{d\eta}{dt}$$

F. Freimuth, S. Blügel and Y. Mokrousov, PRB **95**, 094434 (2017)

Mn/W(001) AND Co/Pt(111)

Current pulse induced by collapsing exchange field





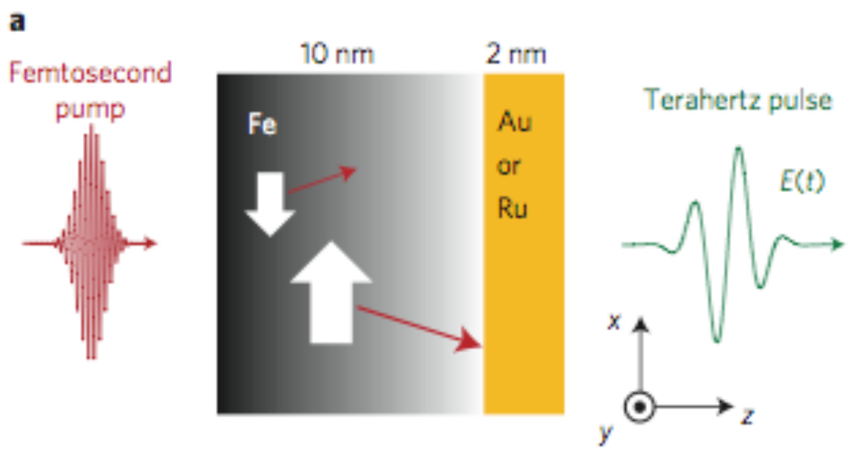
Spin-orbit field is in-plane
→ Effect is maximized for in-plane M
→Magnetic version of the
inverse Edelstein effect

F. Freimuth, S. Blügel and Y. Mokrousov, PRB **95**, 094434 (2017)

COLLAPSING EXCHANGE FIELD

Estimates of THz pulses induced by collapsing exchange field

Assume 10% demagnetization in 500fs and no transverse spin fluctuations.
→Theoretically expected demagnetization-induced THz pulse is of the same order of magnitude as the contribution from superdiffusive spin-current



Absent in Co/Pt und Fe/Au, where It clearly correlates with SHE.

T. Kampfrath et al.,
Nature Nanotechnology **8**, 256 (2013)

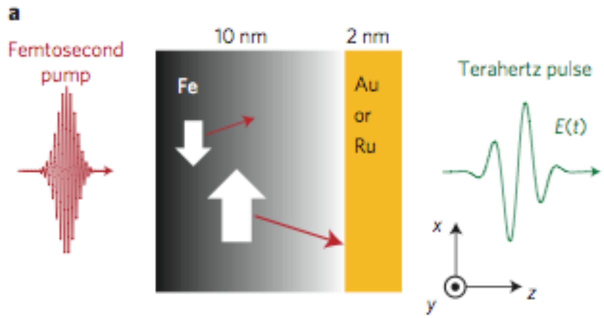
Not yet identified experimentally → Transverse spin fluctuations
→ Search in materials with clear evidence for exchange field collapse.
→ Search in materials with negligible superdiffusive spin-current (bulk compounds, e.g. Half Heuslers)

F. Freimuth, S. Blügel and Y. Mokrousov, PRB **95**, 094434 (2017)

SUMMARY

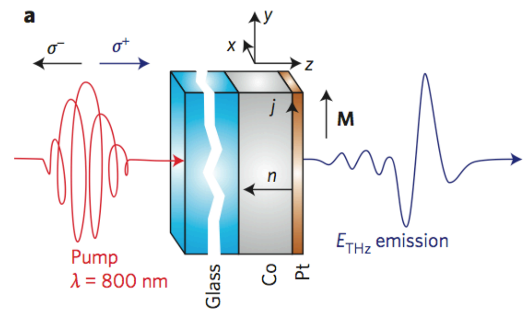
4 mechanisms for generation of in-plane photocurrents in magnetic bilayers

1.) Superdiffusive current + ISHE



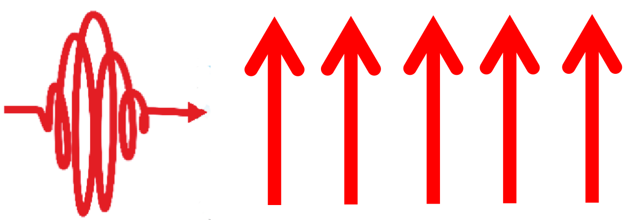
Nature Nanotechnology **8**, 256 (2013)

2.) Magnetization tilt (from IFE) + ISOT



Nature Nanotechnology **11**, 455 (2016)

3.) Driven by ultrafast demagnetization



PRB **95**, 094434 (2017)

4.) Direct conversion of light into electric currents and spin currents

arXiv:1710.10480

Importance of transients: Not so important in the Rashba model

Thank you for your attention