

# Analytic and *ab initio* theory of magnetization dynamics

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# Outline

- The Landau-Lifshitz-Gilbert equation: a derivation from fundamental principles
- Relativistic field-derivative torque, inertia, and optical spin-orbit torque

$$T^{FDT} \propto M \times \frac{\partial H}{\partial t} \quad T^{inert} = M \times \left[ I \cdot \frac{\partial^2 M}{\partial t^2} \right] \quad T^{OSOT} = -\frac{e^2}{2m^2 c^2 \epsilon_0} M \times j_s$$

- *Ab initio* theory of SOTs & spin & orbital accumulation in Pt/3d-metal bilayers

$$\mathcal{T}_o = +2\mu_B |\mathbf{B}_{XC}| |\mathbf{E}| \chi_{yx}^S \mathbf{u}_x \quad \mathcal{T}_e = -2\mu_B |\mathbf{B}_{XC}| |\mathbf{E}| \chi_{xx}^S \mathbf{u}_y$$

(FL SOT) (DL SOT)

- *Ab initio* theory of field-induced spin & orbital Rashba-Edelstein effects in noncentrosymmetric CuMnAs and Mn<sub>2</sub>Au antiferromagnets

# Thanks to



Ritwik Mondal



Marco Berritta



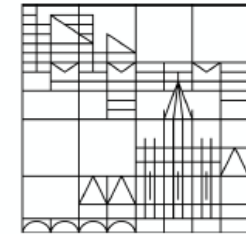
Ashis Nandy



Leandro Salemi

Ulrike Ritzmann, Andreas Donges, Ulrich Nowak

Universität  
Konstanz



*Knut and Alice  
Wallenberg  
Foundation*

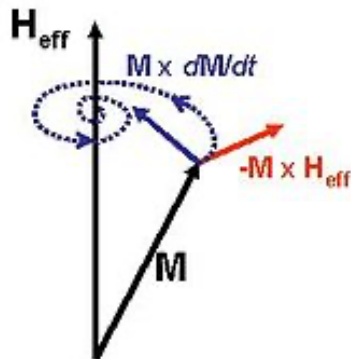


S-NEBULA

# Motivation: How to derive relativistic torques ?!

Landau-Lifshitz-Gilbert equation:

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} + \alpha \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}$$



Semi-empirical, dissipative,  
microscopic level, continuous  
description, long timescale  $\sim$ ns

Torques added *ad hoc*

Dirac-Kohn-Sham theory:

$$\mathcal{H} = c \boldsymbol{\alpha} \cdot (\mathbf{p} - e\mathbf{A}) + (\beta - \mathcal{I}) mc^2 + V$$

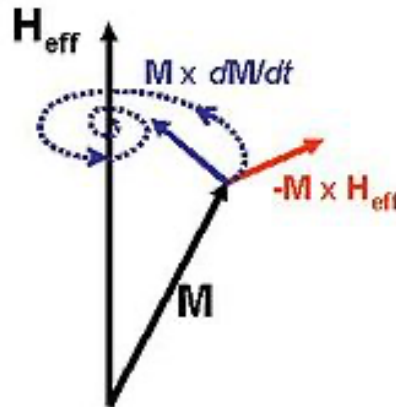
Fundamental, Hermitian  
(non-dissipative), electronic  
level, quantized,  
short timescale  $\sim$ atto-femto sec.

- Any relation between the equations?
- Expressions for relativistic torques & Gilbert damping?



# Relativistic theory of Gilbert damping

Origin of Gilbert damping:



$$\propto \mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t}$$

*Spin-orbit coupling related*

Theories of damping

Kamberský, Can. J. Phys. **48**, 2906 (1970)

Kamberský, Phys. Rev B **76**, 134416 (2007)

Gilmore et al, PRL **99**, 027204 (2007)

Brataas et al, PRL **101**, 037207 (2008)

Ebert et al, PRL **107**, 066603 (2011)

Fähnle & Illg, JPCM **23**, 493201 (2011)

Hickey, Moodera, PRL **102**, 137601 (2009)

Breathing Fermi surface model

Torque-torque correlation model

Scattering theory formulation

Linear resp. theory, CPA

Effective field theory

Derivation from Pauli equation

➡ Start from general Dirac-Kohn-Sham Hamiltonian

# Most general relativistic Hamiltonian

Dirac KS hamiltonian:  $H = c\boldsymbol{\alpha} \cdot (\mathbf{p} - e\mathbf{A}) + (\beta - \mathbb{1})mc^2 + V - \mu_B\beta \boldsymbol{\Sigma} \cdot \mathbf{B}^{\text{xc}}$

external E-M field

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ \boldsymbol{\sigma} & 0 \end{pmatrix} \quad \beta = \begin{pmatrix} \mathbb{1} & 0 \\ 0 & -\mathbb{1} \end{pmatrix}$$

$$\boldsymbol{\Sigma} = \mathbb{1} \otimes \boldsymbol{\sigma}$$

Foldy-Wouthuysen transformation with  $\mathbf{B}^{\text{xc}}$ :

$$H_{\text{FW}} = \frac{(\mathbf{p} - e\mathbf{A})^2}{2m} + V - \mu_B \boldsymbol{\sigma} \cdot \mathbf{B}^{\text{xc}} - \mu_B \boldsymbol{\sigma} \cdot \mathbf{B} - \frac{(\mathbf{p} - e\mathbf{A})^4}{8m^3c^2} - \frac{1}{8m^2c^2}(p^2V) - \frac{e\hbar^2}{8m^2c^2} \nabla \cdot \mathbf{E}$$

$$+ \frac{i}{4m^2c^2} \boldsymbol{\sigma} \cdot (\mathbf{p}V) \times (\mathbf{p} - e\mathbf{A}) - \frac{e\hbar}{8m^2c^2} \boldsymbol{\sigma} \cdot \{ \mathbf{E} \times (\mathbf{p} - e\mathbf{A}) - (\mathbf{p} - e\mathbf{A}) \times \mathbf{E} \}$$

$$+ \frac{\mu_B}{8m^2c^2} \{ [p^2(\boldsymbol{\sigma} \cdot \mathbf{B}^{\text{xc}})] + 2\boldsymbol{\sigma} \cdot (\mathbf{p}\mathbf{B}^{\text{xc}}) \cdot (\mathbf{p} - e\mathbf{A}) + 2(\mathbf{p} \cdot \mathbf{B}^{\text{xc}})\boldsymbol{\sigma} \cdot (\mathbf{p} - e\mathbf{A}) + 4[\mathbf{B}^{\text{xc}} \cdot (\mathbf{p} - e\mathbf{A})]\boldsymbol{\sigma} \cdot (\mathbf{p} - e\mathbf{A}) \}$$

$$+ \frac{i\mu_B}{4m^2c^2} [(\mathbf{p} \times \mathbf{B}^{\text{xc}}) \cdot (\mathbf{p} - e\mathbf{A})].$$

- Relativistic correction to exchange field  $\mathbf{B}_{\text{eff}}^{\text{xc}} \equiv \mathbf{B}^{\text{xc}} + \mathbf{B}_{\text{corr}}^{\text{xc}}$
- Full expression for SO with external E-M fields (new terms)
- Gauge invariant and Hermitian Hamiltonian

## New terms in SO Hamiltonian

Relativistic Hamiltonian couples *angular momentum of light*  $j$  with electron spin  $\sigma$

$$\mathcal{H}_{\text{SOC}} = \frac{i}{4m^2c^2} \boldsymbol{\sigma} \cdot (\mathbf{p}V) \times (\mathbf{p} - e\mathbf{A}) - \frac{e\hbar}{8m^2c^2} \boldsymbol{\sigma} \cdot \{ \mathbf{E}_{\text{ext}} \times (\mathbf{p} - e\mathbf{A}) - (\mathbf{p} - e\mathbf{A}) \times \mathbf{E}_{\text{ext}} \}$$

Unusual coupling:  $\mathcal{H} = \frac{e^2\hbar}{4m^2c^2} \boldsymbol{\sigma} \cdot (\mathbf{E} \times \mathbf{A})$

$$\xi \boldsymbol{\sigma} \cdot \mathbf{j}_s$$

$$\mathbf{j}_s = -2\mathbf{E} \times \mathbf{A} \quad (\epsilon_0 = 1)$$

Spin-photon ang. moment coupling  $\mathcal{H}_{\text{light-spin}}^{\text{ext}} = \frac{e^2}{2m^2c^2\epsilon_0} \mathbf{S} \cdot \mathbf{j}_s$

*Optical spin-orbit torque:*

$$B_{\text{opt}} \propto \frac{e^2}{2m^2c^2\mu_B\omega} E_0^2$$

$$\left. \frac{\partial \mathbf{M}}{\partial t} \right|_{\text{light-spin}}^{\text{ext}} = -\frac{e^2}{2m^2c^2\epsilon_0} \mathbf{M} \times \mathbf{j}_s$$

“inverse Faraday effect”

$$B_{\text{opt}} \sim 0.3 - 4 \text{ mT}$$

$$\mathbf{T} = -\gamma \mathbf{M} \times \mathbf{B}_{\text{opt}}$$

cf. Tesarova et al., Nat. Phot. **7**, 492 (2013)

Mondal, Berritta, Paillard et al, PRB **92**, 100402R (2015)

Mondal, Berritta, PMO, JPCM **29**, 194002 (2017)

## Equation of motion for spin dynamics

Full *spin* Hamiltonian:  $\mathcal{H}^S(t) = \mathcal{H}^0 + \mathcal{H}_{\text{soc}}^{\text{int}} + \mathcal{H}_{\text{soc}}^{\text{ext}}$

Spin operator dynamics:  $\partial \vec{S} / \partial t = -i[\vec{S}, H^S] / \hbar$

Magnetization element  $M = \sum_j \frac{g\mu_B}{\mathcal{V}} \text{Tr}\{\rho S^j\}$

*Magnetization Dynamics*  $\frac{\partial M}{\partial t} = \frac{g\mu_B}{\mathcal{V}} \frac{1}{i\hbar} \sum_j \text{Tr}\{\rho [S^j, \mathcal{H}^S(t)]\}$

$$\frac{\partial M}{\partial t} = -\gamma M \times B_{\text{eff}} + M \times \left[ A \cdot \frac{\partial M}{\partial t} \right]$$

$\mathcal{H}_{\text{soc}}^{\text{ext}}$

(Without spin-photon term and currents!)

Mondal, Berritta, PMO, Phys. Rev. B **94**, 144419 (2016)

With *currents*: Mondal, Berritta, PMO, PRB **98**, 214429 (2018)

# Intrinsic spin damping contributions

Gilbert damping  $\mathbf{A}$  for *harmonic* fields:  $\left. \frac{\partial \mathbf{M}}{\partial t} \right|_{\text{soc}}^{\text{ext}} = \mathbf{M} \times \left[ \mathbf{A} \cdot \frac{\partial \mathbf{M}}{\partial t} \right]$

$$A_{ij} = -\frac{e\mu_0}{8m^2c^2} \sum_{n,k} \left[ \underbrace{\langle r_i p_k + p_k r_i \rangle - \langle r_n p_n + p_n r_n \rangle \delta_{ik}}_{\text{Electronic damping}} \right] \underbrace{(\mathbb{1} + \chi^{-1})_{kj}}_{\text{Spin-spin corr.*}}$$

$$\langle r_\alpha p_\beta \rangle = -\frac{i\hbar}{2m} \sum_{n,n',k} \frac{f(E_{nk}) - f(E_{n'k})}{E_{nk} - E_{n'k}} p_{nn'}^\alpha p_{n'n}^\beta$$

\*Garate & MacDonald, PRB **79**, 064403 (2009)

Damping for *general time-dependent* magn. fields:

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma_0 \mathbf{M} \times \mathbf{H}_{\text{eff}} + \mathbf{M} \times \left[ \bar{\mathbf{A}} \cdot \left( \frac{\partial \mathbf{M}}{\partial t} + \frac{\partial \mathbf{H}}{\partial t} \right) \right]$$

➤ New expression for spin dynamics in presence of time-dep. fields

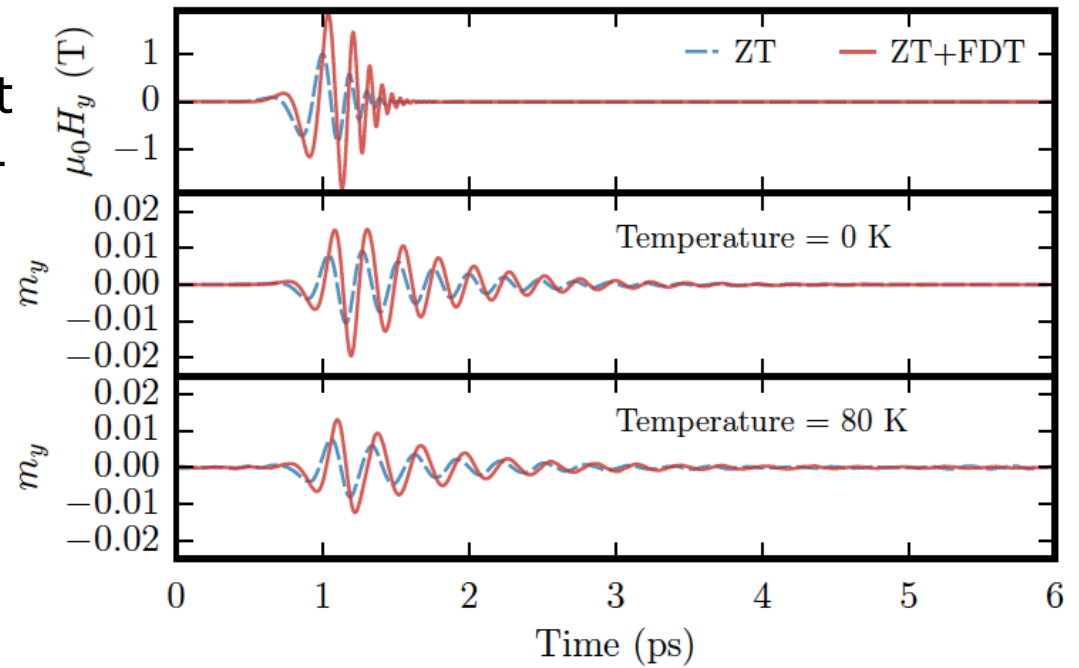
# Field-derivative torque

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma_0 \mathbf{M} \times \mathbf{H}_{\text{eff}} + \mathbf{M} \times \left[ \bar{\mathbf{A}} \cdot \left( \frac{\partial \mathbf{M}}{\partial t} + \frac{\partial \mathbf{H}}{\partial t} \right) \right] \quad (\text{LLG})$$

$$\frac{\partial \mathbf{m}_i(t)}{\partial t} = -\frac{\gamma}{(1 + \alpha^2)\mu_s} \mathbf{m}_i(t) \times \left[ \left( \mathbf{H}_i^{\text{eff}} - \frac{\alpha a^3}{\gamma} \frac{\partial \mathbf{H}}{\partial t} \right) + \alpha \mathbf{m}_i(t) \times \left( \mathbf{H}_i^{\text{eff}} - \frac{\alpha a^3}{\gamma} \frac{\partial \mathbf{H}}{\partial t} \right) \right] \quad (\text{LL})$$

Zeeman ZT
FDT

CoO resonant spin excitation at  $\alpha = 0.02$



- for THz pulses FDT could be important
- phase difference between ZT and FDT
- at high damping, FDT can be large
- No experimental observation so far !

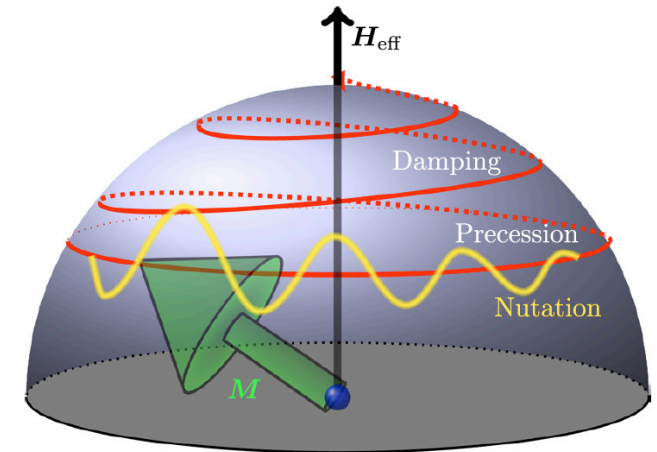
# Origin of **inertial** spin dynamics?

## Inertial dynamics:

$$\frac{\partial \mathbf{M}}{\partial t} = \mathbf{M} \times \left( -\gamma_0 \mathbf{H} + \Gamma \cdot \frac{\partial \mathbf{M}}{\partial t} + \underline{\mathcal{I}} \cdot \frac{\partial^2 \mathbf{M}}{\partial t^2} \right)$$

## Earlier work:

- Kimel et al, Nat. Phys. **5**, 727 (2009)
- Ciornei et al, PRB **83**, 020410R (2011)
- Fähnle et al, PRB **84**, 172403 (2011)
- Bhattacharjee et al, PRL **108**, 057204 (2012)
- Böttcher & Henk, PRB **86**, 020404 (2012)



## Can inertial dynamics (nutation) be a higher order relativistic effect?

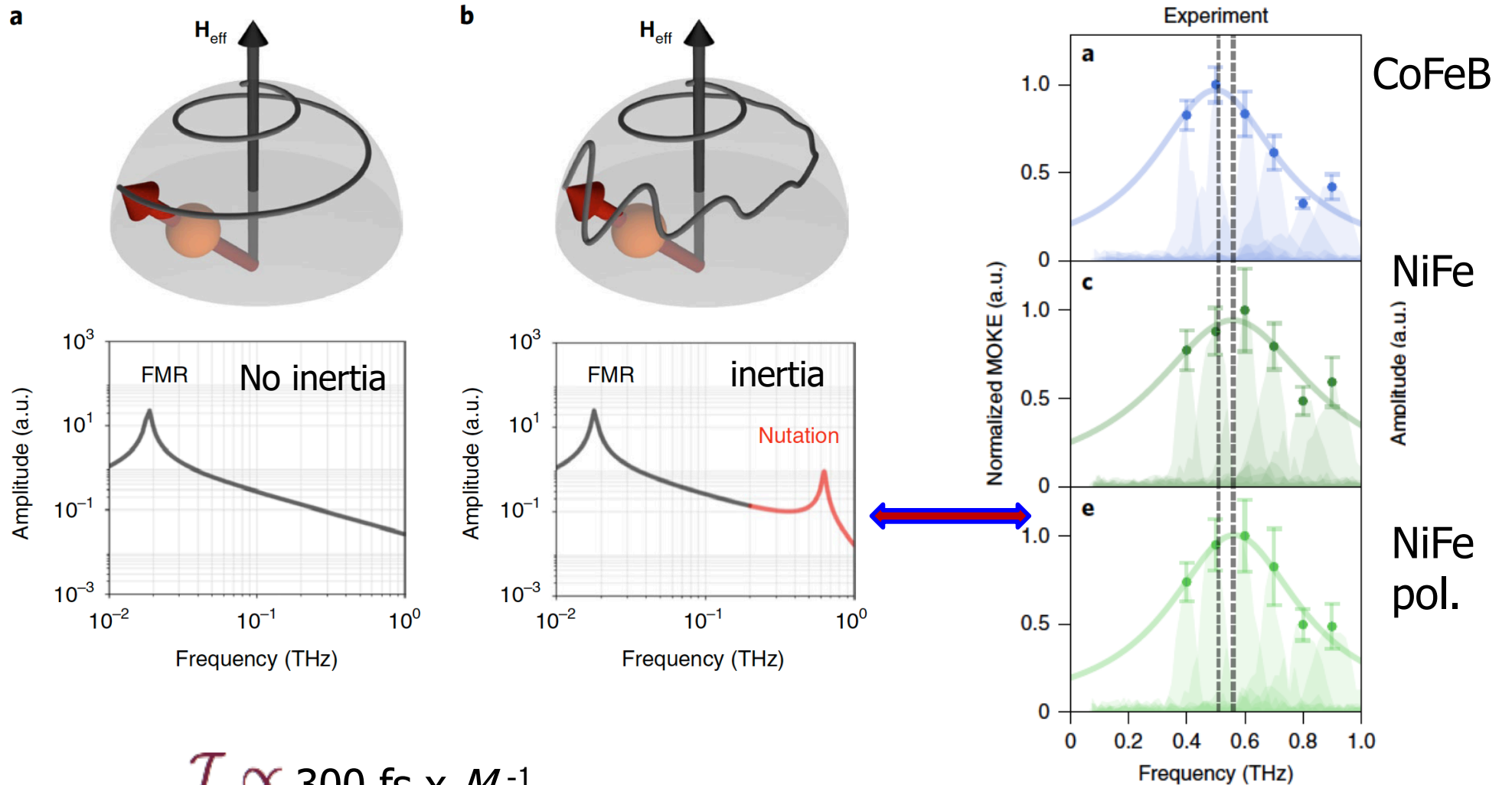
FW transformation for all terms up to order  $1/c^4$  (higher order terms in SO Ham.)

$$\begin{aligned} \mathcal{H} &= -\frac{i e \hbar^2}{16 m^3 c^4} \boldsymbol{\sigma} \cdot [\partial_t \mathbf{E}_{\text{tot}} \times (\mathbf{p} - e \mathbf{A}) + (\mathbf{p} - e \mathbf{A}) \times \partial_t \mathbf{E}_{\text{tot}}] \\ &= \frac{e \hbar^2}{8 m^3 c^4} \mathbf{S} \cdot \partial_{tt} \mathbf{B} \end{aligned} \quad \Rightarrow \quad \left. \frac{\partial \mathbf{M}}{\partial t} \right|_{\text{iner.}} = \mathbf{M} \times \left[ \mathcal{I} \cdot \frac{\partial^2 \mathbf{M}}{\partial t^2} \right]$$

Mondal, Berritta, Nandy, PMO, PRB **96**, 024425 (2017)

Mondal, Berritta, PMO, JPCM **30**, 265801 (2018)

# Recent observation of inertial dynamics



$$I \propto 300 \text{ fs} \times M^{-1}$$

Kerr amplitude of magn. dynamics

Neeraj et al, Nat. Phys. **17**, 245 (2021)



## Size of *intrinsic* inertia

For ac magn. field - **inertia tensor**:  $\mathcal{I}_{ij} = \frac{\mu_0 \gamma \hbar^2}{8m^2 c^4} [\mathbb{1} + \text{Re}(\chi_m^{-1})_{ij}]$

Gilbert damping tensor: Imaginary part of susceptibility, inertia tensor real part, *but smaller because of pre-factor*

$$\Gamma_{ij} = - \frac{\mu_0 \gamma \hbar}{4mc^2} \sum_{n,k} \left[ \frac{\langle r_i p_k + p_k r_i \rangle - \langle r_n p_n + p_n r_n \rangle \delta_{ik}}{i \hbar} \right] \text{Im}(\chi_m^{-1})_{kj}$$

$$\left. \begin{array}{l} \text{Gilbert damping tensor} \\ \text{inertia tensor real part} \end{array} \right\} \mathcal{I} \propto -\Gamma \bar{\tau}$$

- Inertial dynamics important at *short time scales* (1 fs – 1 ps)
- Offers options for THz spin dynamics

$$\bar{\tau} = 746 \pm 46 \text{ fs}$$

for 3 different Co films

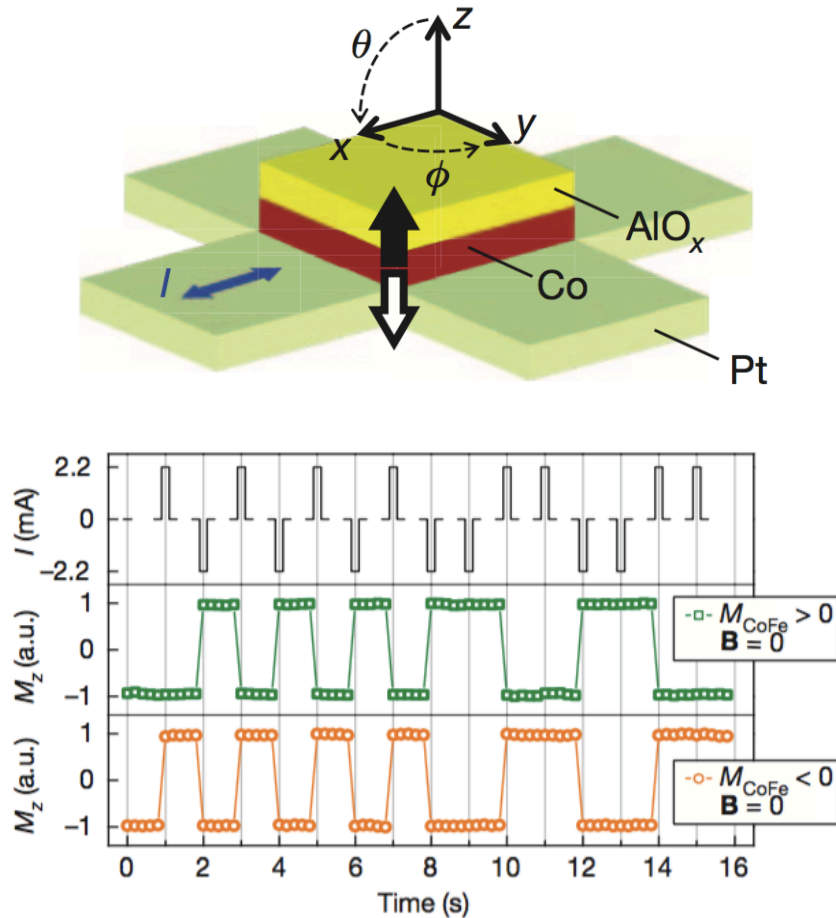
Unikandanunni et al,  
arXiv 2109.03076

Perhaps consistent with  
relativistic theory

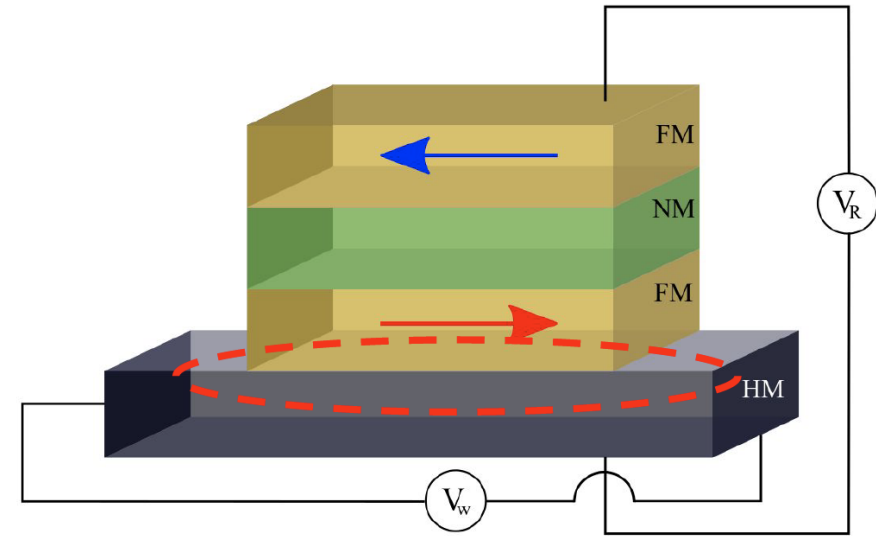
### Recent theories

Bajpai, Nikolic, PRB **99**, 134409 (2019)  
 Makhfudz, Olive, Nicolis, APL **117**, 132403 (2020)  
 Cherkasskii et al, PRB **102**, 184432 (2020)  
 Titov et al, PRB **103**, 144433 (2021)  
 Mondal et al, PRB **103**, 104404 (2021)  
 Thibaudeau, Nicolis, arXiv 2103.04787

## II. Spin-orbit torque - Magnetization switching with SHE



Miron et al, Nature **476**, 189 (2011)  
Liu et al, Science **336**, 555 (2012)



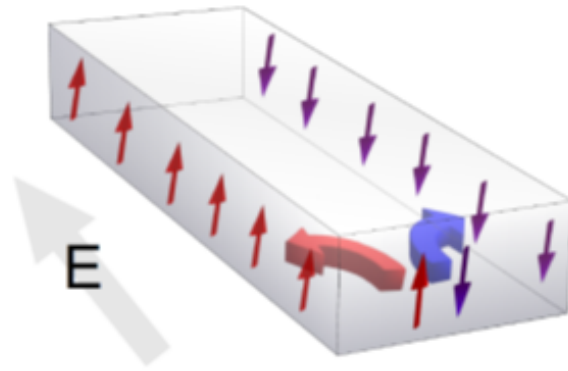
Switching due to large SHE of heavy metals (Pt, W, Ta) and interfacial effect

Sinova, Valenzuela, Wunderlich, Back, Jungwirth, Rev.Mod.Phys. **87**, 1213 (2015)

Manchon et al, Rev.Mod.Phys. **91**, 035004 (2019)

# Origin of SOT: charge-to-spin conversion

Spin Hall effect



*Transport*

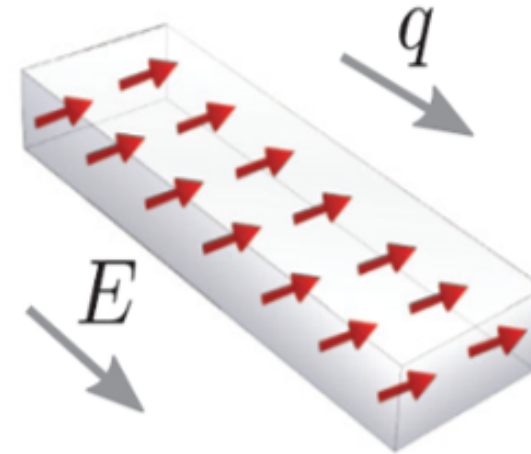
$$J_x^{S_z} = \sigma_{xy}^{S_z} \cdot E_y$$

Dyakonov & Perel, JETP Lett. **13**, 467 (1971)

Hirsch, PRL **83**, 1834 (1999)

Infinite bulk crystal

Inverse spin galvanic effect,  
Rashba-Edelstein effect



*Local*

$$\delta S_x^{ind} = \chi_{xy} \cdot E_y$$

Edelstein, Solid State  
Commun. **73**, 233 (1990)

(Rashba SOC + 2D)

# Ab initio calculations

## Rashba-Edelstein effect

$$M_i^{ind} = \chi_{ij} \cdot E_j \quad \text{magneto-electric effect, possible for inversion symm. breaking}$$

Linear-response theory formulation:  $\delta M = \mu_B \delta(\mathbf{L} + 2\mathbf{S}) \longrightarrow A$

$$\chi_{ij}^A = -\frac{ie}{m_e} \int_{\Omega} \frac{dk}{\Omega} \sum_{n \neq m} \frac{f_{nk} - f_{mk}}{\hbar\omega_{nmk}} \frac{A_{mnk}^i P_{nmk}^j}{-\omega_{nmk} + i\tau_{inter}^{-1}}$$

$$-\frac{ie}{m_e} \int_{\Omega} \frac{dk}{\Omega} \sum_n \frac{\partial f_{nk}}{\partial \epsilon} \frac{A_{nnk}^i P_{nnk}^j}{i\tau_{intra}^{-1}}$$

$$\delta S = \chi^S E$$

$$\delta L = \chi^L E$$

---

Spin Hall effect  $\mathbf{J}^{S_k} = \boldsymbol{\sigma}^{S_k} \mathbf{E} \quad A = \hat{\mathbf{J}}^{S_k} = \frac{\{\hat{S}_k, \hat{\mathbf{p}}\}}{2m_e V}$

Orbital Hall effect  $\mathbf{J}^{L_k} = \boldsymbol{\sigma}^{L_k} \mathbf{E}$  Spin/orbital current operator

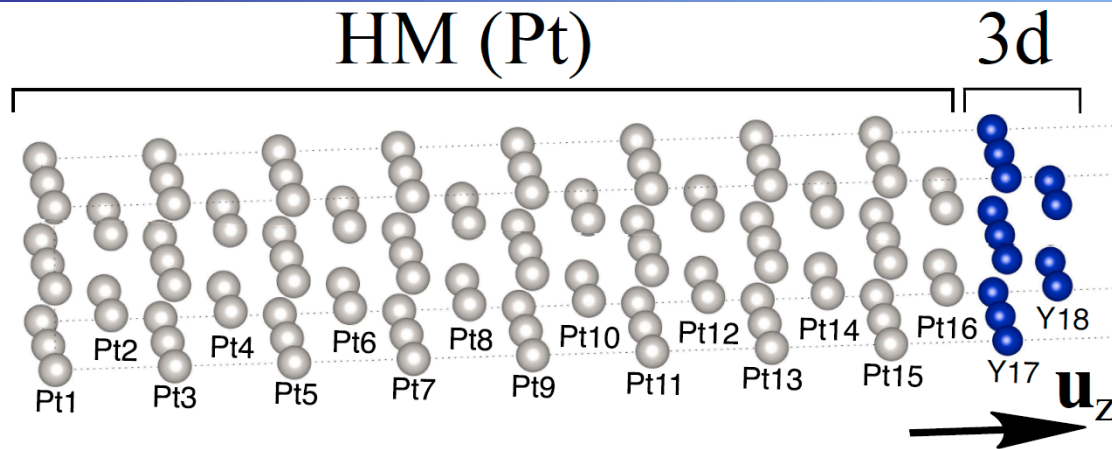
SHE: Guo, Yao, and Niu, PRL **94**, 226601 (2005)

OHE: Tanaka et al, Phys. Rev. B **77**, 165117 (2008)

Jo, Go, and Lee, PRB **98**, 214405 (2018)

(Relativistic WIEN2k)

# SOTs at symmetry broken interface Pt/3d FM



$n \text{ Pt} / 2 \text{ Y}$   
 $\text{Y} = \text{Ni, Co, Cu or Pt}$

$E$  along  $x$

$$\left[ \begin{array}{l} \delta S_x = \chi_{xx}^S E_x \\ \delta S_y = \chi_{yx}^S E_x \end{array} \right.$$

Effective torques

$$\mathcal{T} = \mathbf{M} \times \delta \mathbf{B} \quad \delta \mathbf{B} \approx |\mathbf{B}_{\text{XC}}| \frac{\delta \mathbf{S}}{|\mathbf{S}|}$$

$$\mathcal{T}_o = + 2\mu_B |\mathbf{B}_{\text{XC}}| |\mathbf{E}| \chi_{yx}^S \mathbf{u}_x$$

$$\mathcal{T}_e = - 2\mu_B |\mathbf{B}_{\text{XC}}| |\mathbf{E}| \chi_{xx}^S \mathbf{u}_y$$

Time-rev. odd - FL

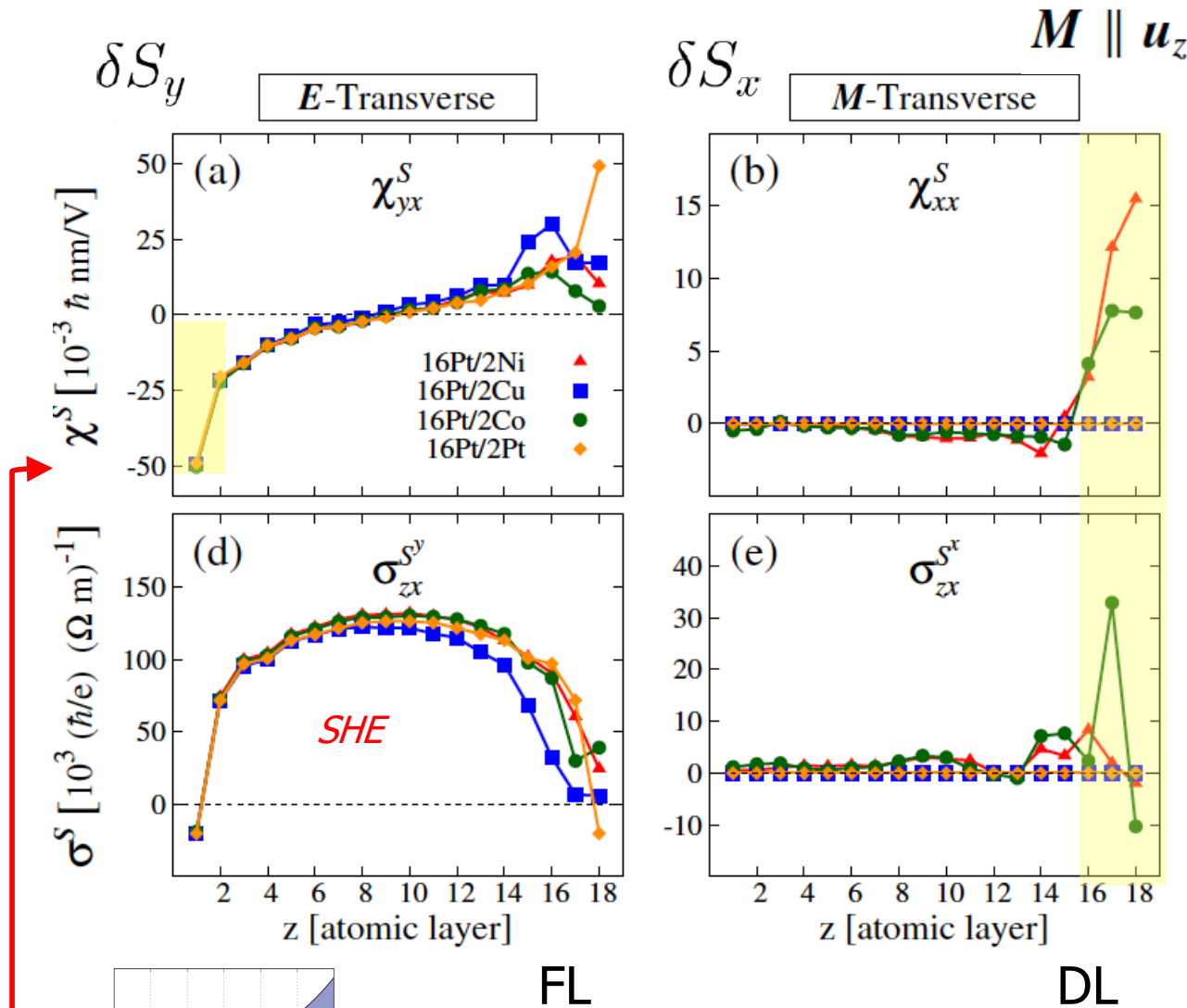
Time-rev. even - DL

Early work:

Haney, Lee, Lee, Manchon, Stiles, PRB **88**, 214417 (2013)

Freimuth, Blügel, Mokrousov, PRB **90**, 174423 (2014)

# Results Pt/3d-bilayers – induced spin polarization

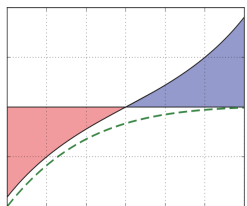


- ❖ Typical transverse spin accumulation (stationary state)
- ❖ Modified at the interface
- ❖ M, t-even effect

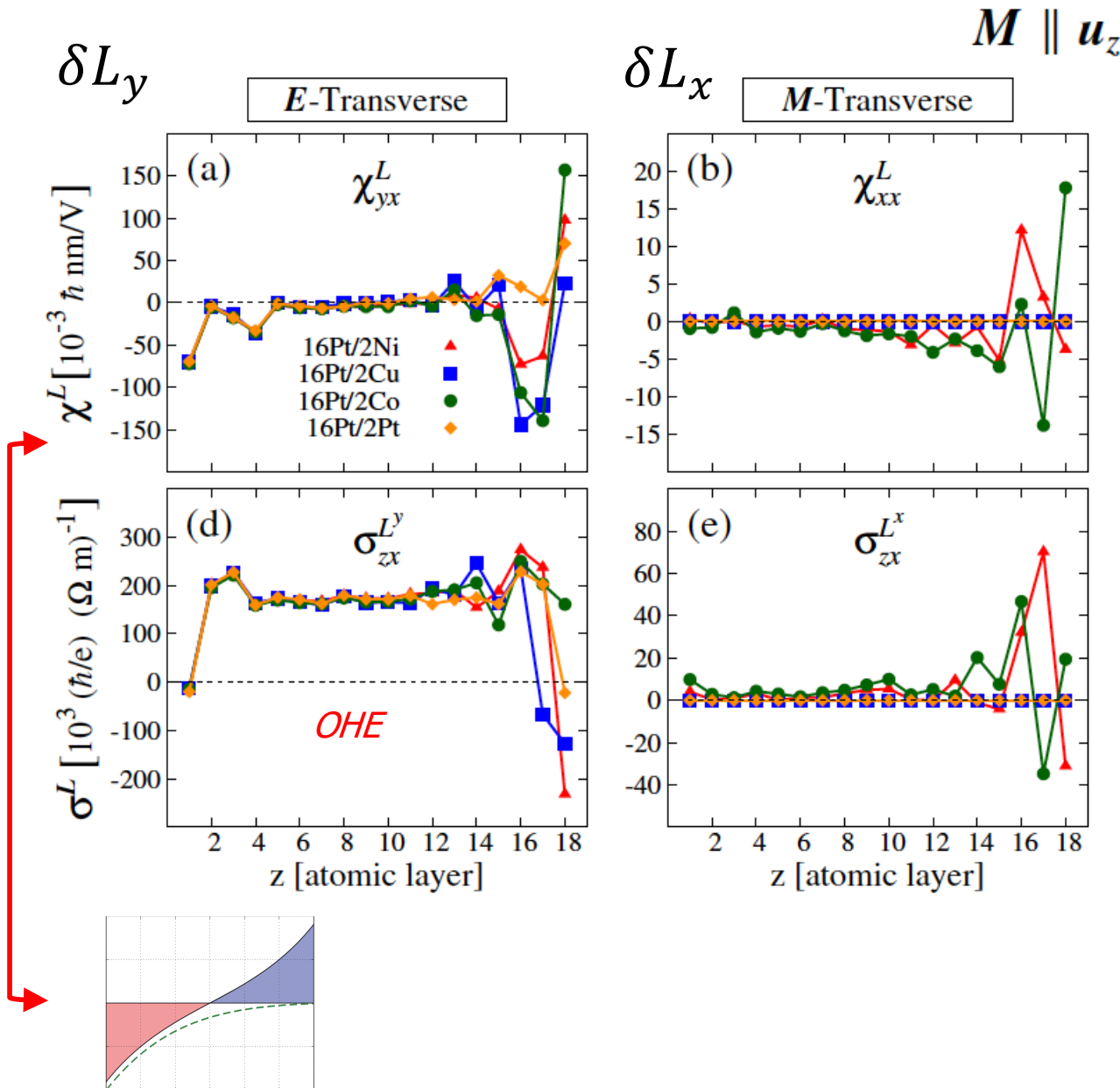
- ❖ Very local response at interface, along  $E_x$
- ❖ Only exists for magn. material (M, t-odd)
- ❖ Same size as transv.

“Magnetic SHE”

Kimata et al, Nature **565**, 627 (2019)



# Results Pt/3d-bilayers – *orbital* polarization & current



- ❖ Huge OHE
- ❖ Orb. accumulation profile quite different from spin
- ❖ Enlarged at the interface
- ❖ M, t-even effect

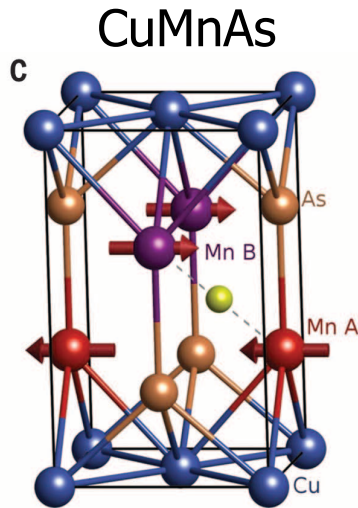
- ❖ Local response at interface, along  $E_x$
- ❖ Only exists for magn. material (M, t-odd)
- ❖ Smaller than transv.

“Magnetic OHE”

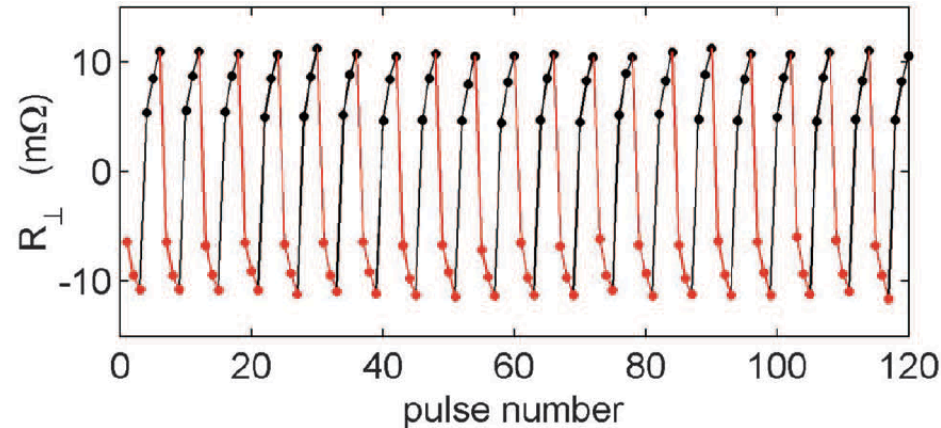
OHE and transv. orbital polarization not due to SOC



# Current-induced SOT switching in AFMs



Wadley et al, Science **351**, 587 (2016)

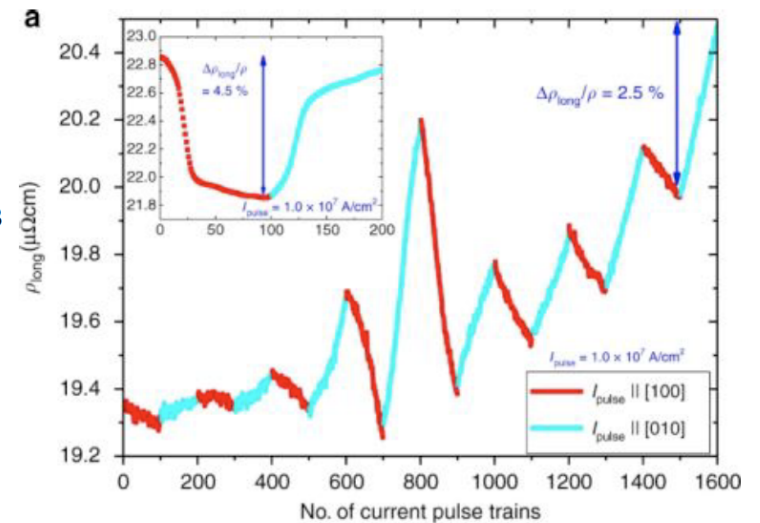
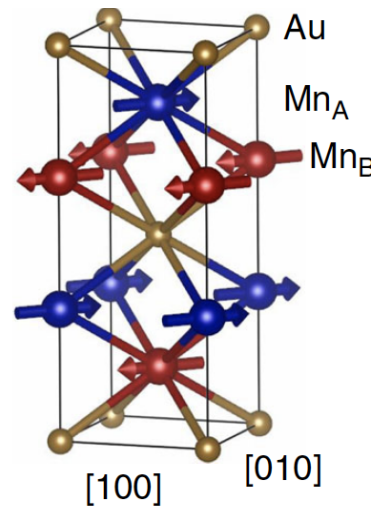


Proposed mechanism: staggered SO torque\*

$$dM_{A,B}/dt \sim M_{A,B} \times p_{A,B}$$

$$p_A = -p_B \quad (p^{ind} \perp j)$$

$$\text{due to } M_i^{ind} = \chi_{ij} \cdot E_j$$



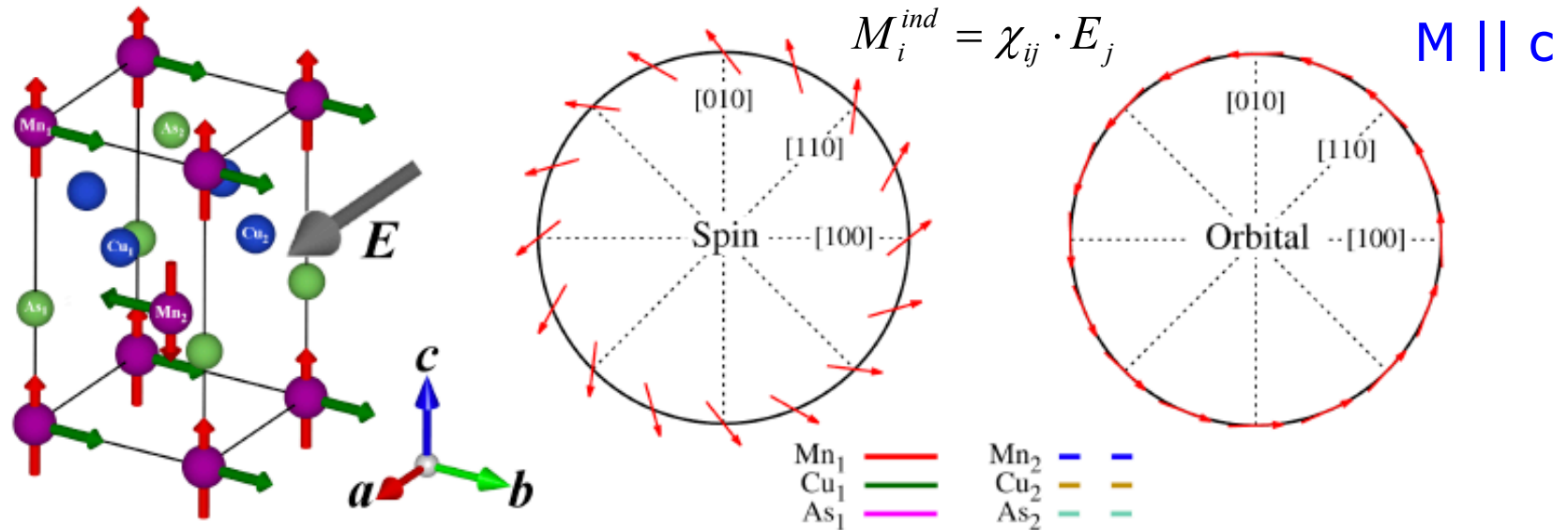
\*Zelezny et al, PRL **113**, 157201 (2014)  
Zelezny et al, PRB **95**, 014403 (2017)

Mn<sub>2</sub>Au

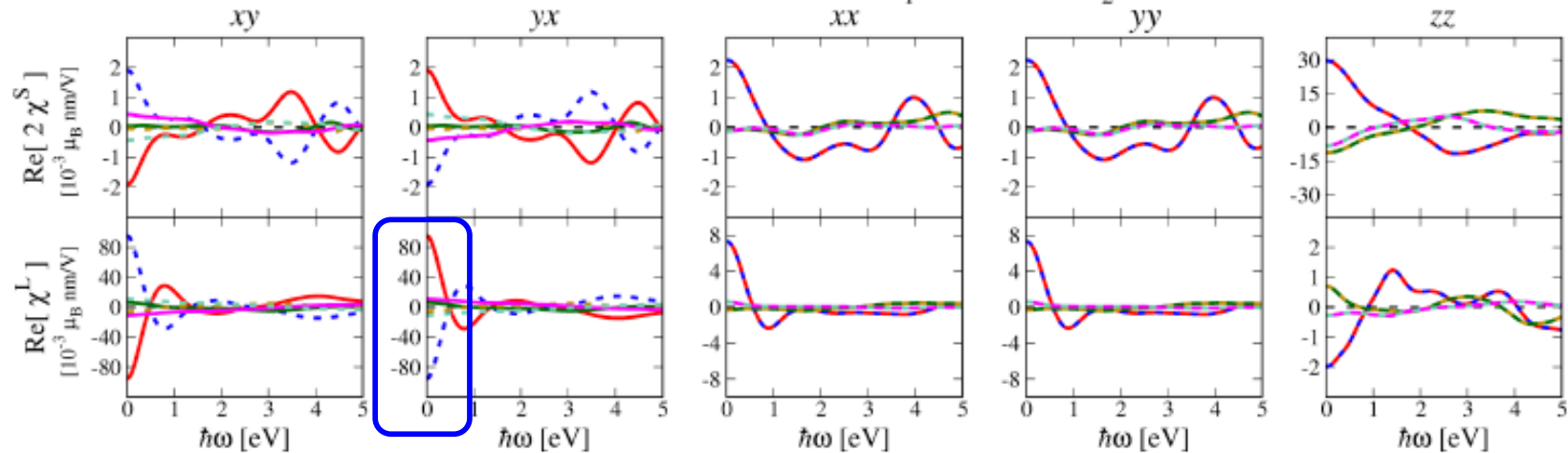
Bodnar et al, Nat. Commun. **9**, 348 (2018) 20



# Results for CuMnAs



SREE  
 $2\chi^S$   
 $\chi^L$   
OREE

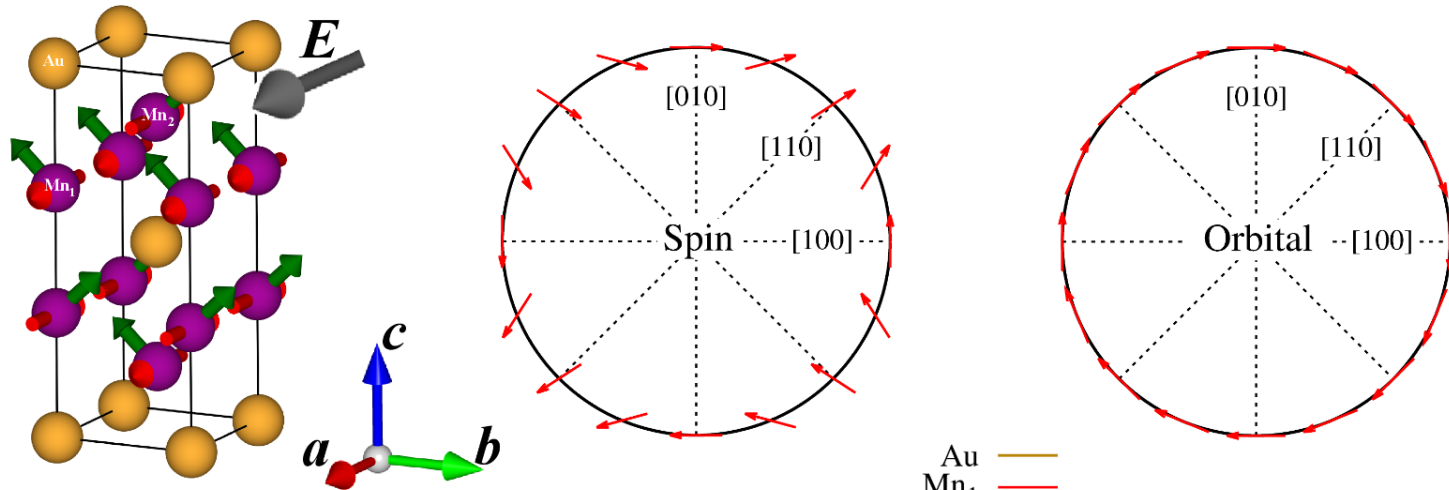


staggered

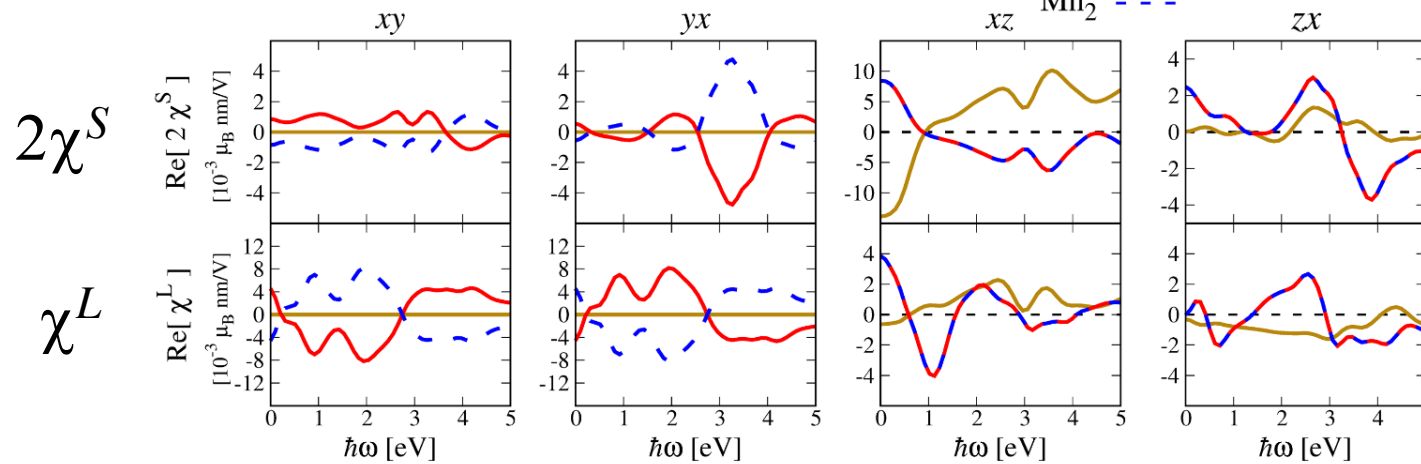
Non-staggered

- Dominant staggered **induced orb. polarization** (40 x larger) – not due to SOC
- Frequency dependent and *non-staggered*  $\chi$  elements also present

# Mn<sub>2</sub>Au – moments in basal plane










$M \parallel a$



- Dresselhaus-like spin response, Rashba-type orbital response
- Large *non-staggered*  $\chi$  elements present, give out-of-plane direction

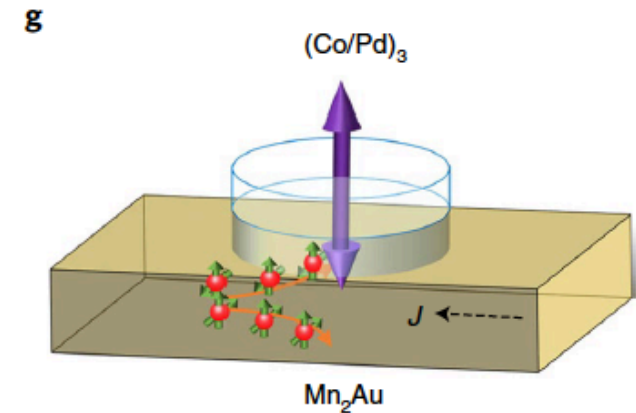
$$\mathcal{PT}\{L_{Mn_1}^{ind}\} = -L_{Mn_2}^{ind}$$

## Observation of the antiferromagnetic spin Hall effect

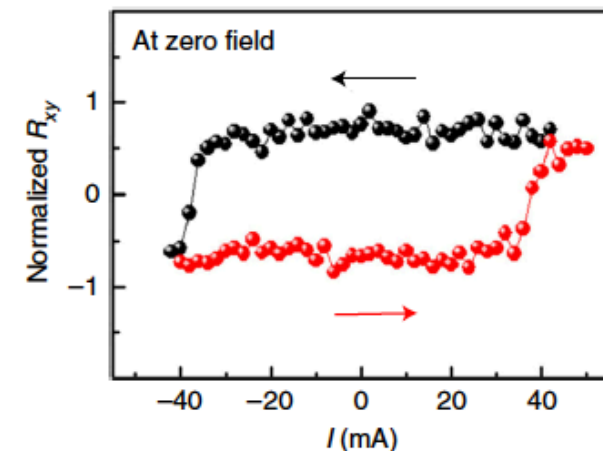
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- Non-staggered element  $zx$  that gives an induced spin polarization in  $z$ -direction for a charge current in  $x$ -direction
- Can be used for so-called field free switching



h Unusual spin polarization normal to layer; not normal SHE



Can switch the magnetization of Co/Pd layer

## Summarizing ...

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- ❖ LLG can be derived from and is consistent with Dirac-Kohn-Sham theory
- ❖ New relativistic SOT: optical SOT, field-derivative torque for time-dep. fields
- ❖ Intrinsic inertial torque can be due to relativistic effects
- ❖ Additional torques important and can extend LLG to shorter time scales
  
- ❖ Orbital Rashba-Edelstein effect huge (much larger than SREE) AFMs
- ❖ OREE staggered, not due to SOC, in symm.-broken AFMs
- ❖ SREE: large non-staggered elements
  
- ❖ Magn. SHE/OHE with induced spin/orbital pol. along E-field in Pt/3d layers
- ❖ Orbital accumulation *different* from spin accumulation
- ❖ Both  $\delta S_x$  and  $\delta S_y$  terms relevant for torques (dep. on magn. direction)