

Picosecond spin caloritronics

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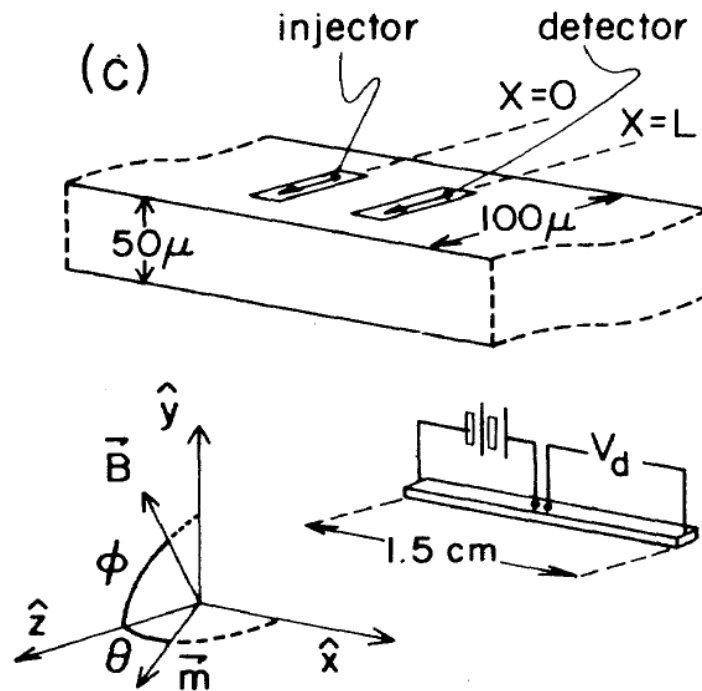


Interfacial Charge-Spin Coupling: Injection and Detection of Spin Magnetization in Metals

Mark Johnson and R. H. Silsbee

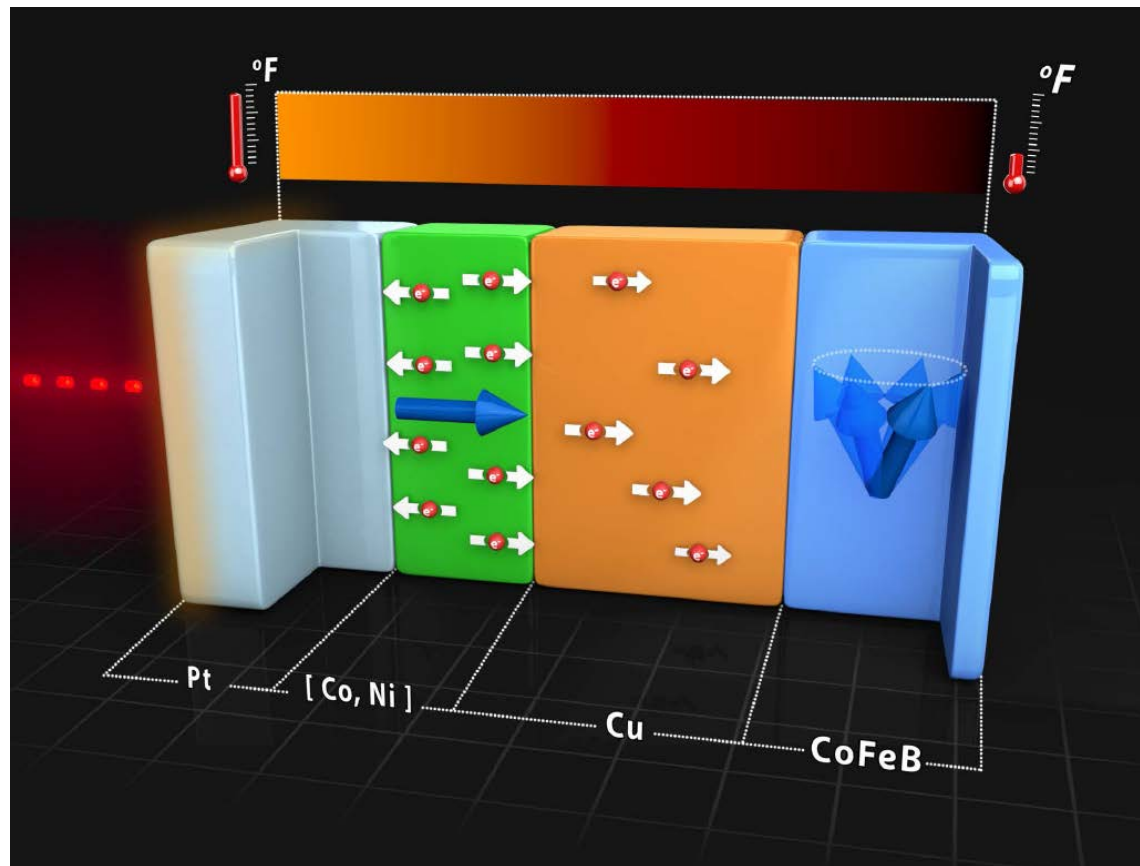
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(Received 1 July 1985)



Drive spin currents with heat instead of charge

- In “picosecond spin caloritronics”, we inject spin current using heat transport and detect spin optically, all on picosecond time scales.



Picosecond time-scales enable enormous heat currents (unit of time in the denominator)

- Conventional heat currents, e.g., heat diffusion equation, Fourier's law in steady-state governed by thermal conductivity Λ

$$J_Q = -\Lambda \nabla T \quad \Lambda \propto \text{W m}^{-1} \text{K}^{-1}$$

- Interface thermal conductance G , ΔT =temperature across an interface

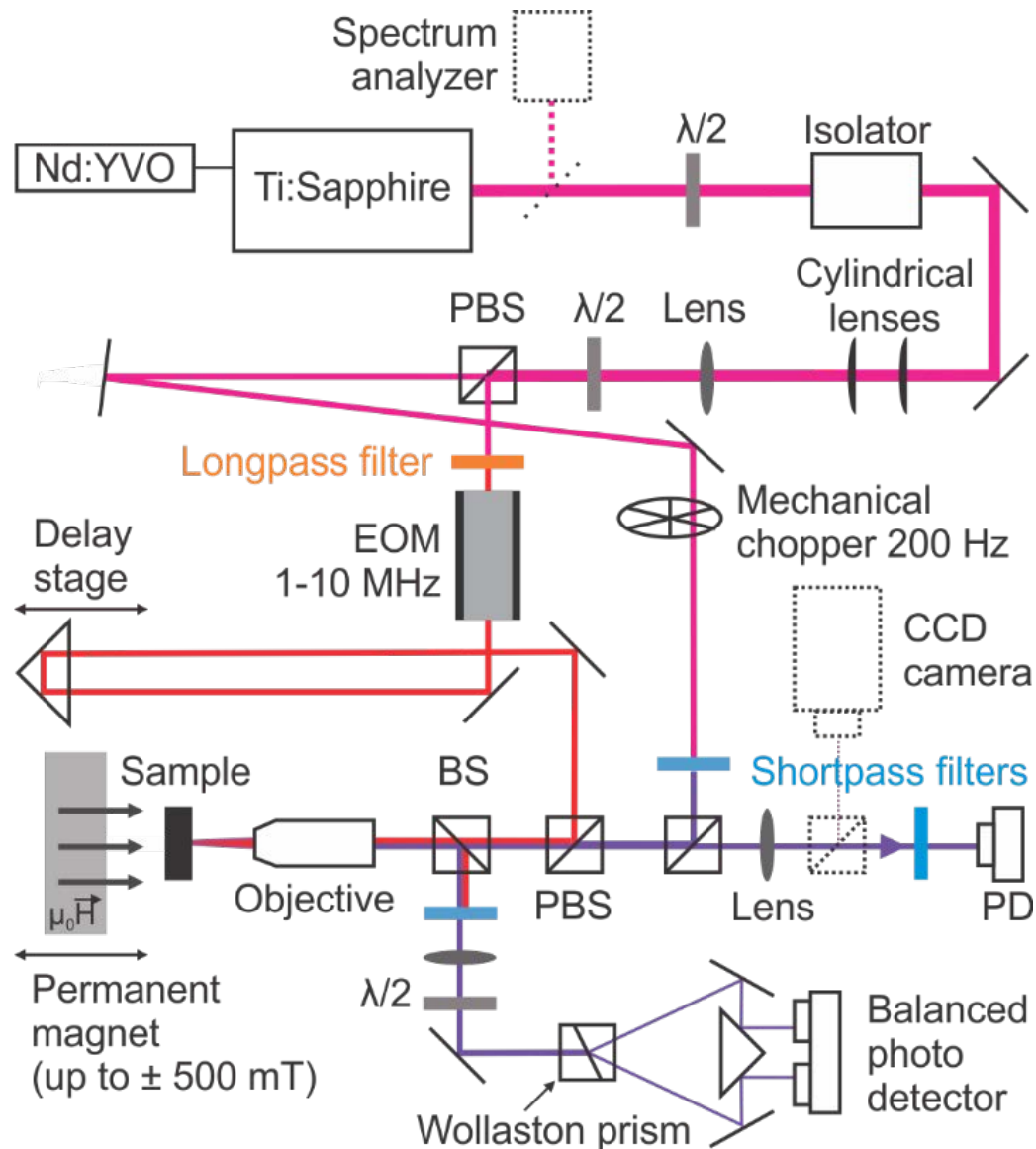
$$J_Q = G \Delta T \quad G \propto \text{W m}^{-2} \text{K}^{-1}$$

- Volumetric heat currents exchanged between excitation, e.g., two-temperature model of electrons and magnons

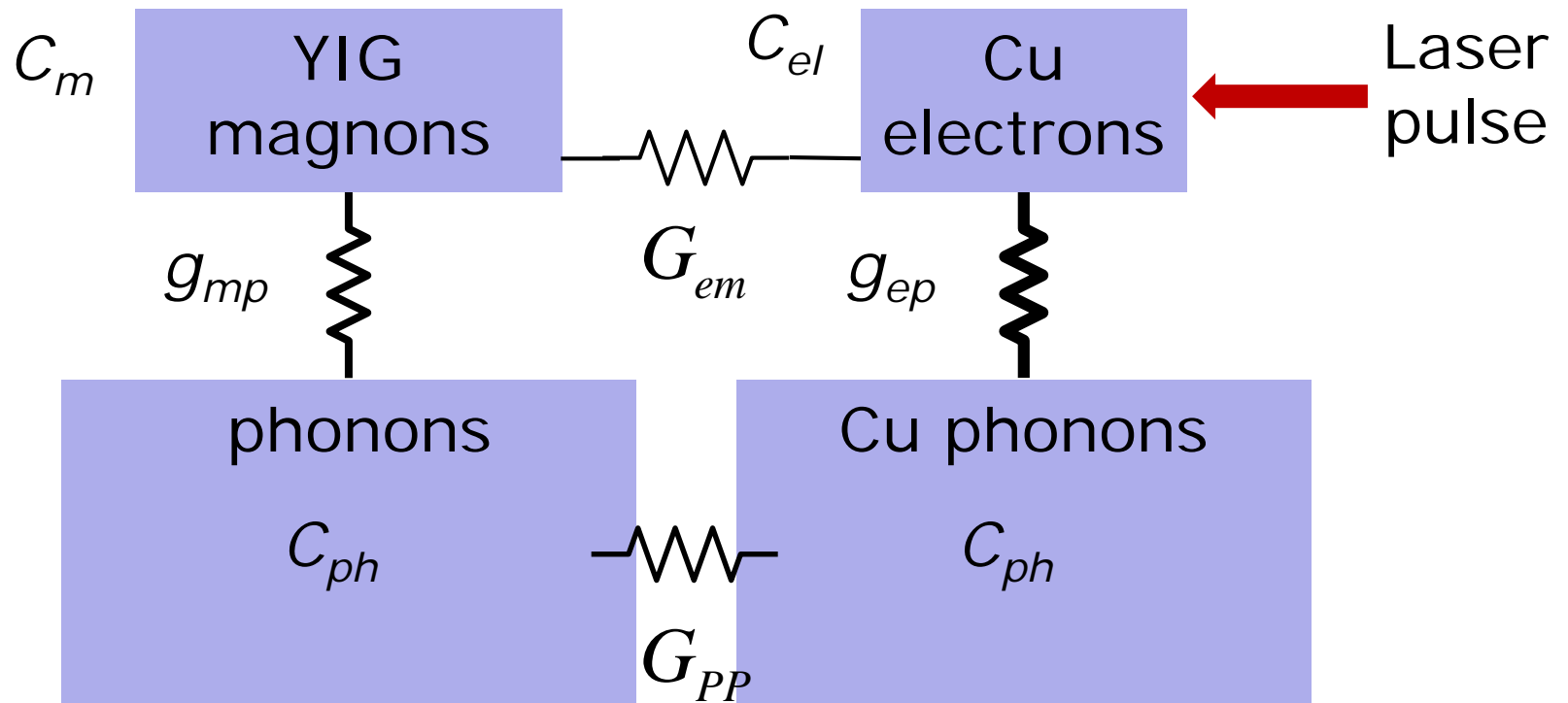
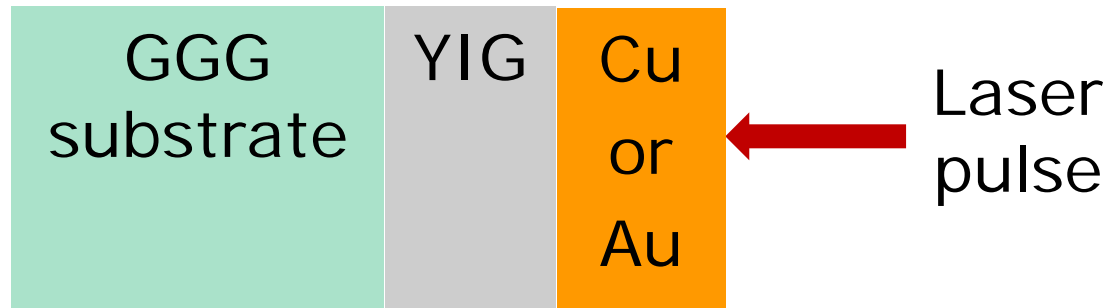
$$\dot{j}_Q = g_{em} (T_e - T_m) \quad g_{em} \propto \text{W m}^{-3} \text{K}^{-1}$$

- **2 Layers** Au/YIG
 - Measure time-resolved spin accumulation in a rapidly heated normal metal by magneto-optic Kerr effect (MOKE)
 - Source of spin is the **interfacial spin-Seebeck effect**
- **3 Layers** Pt/[Co,Pt]/Cu
 - Measure time-resolved spin accumulation in a normal metal by MOKE
 - Dominant source of spin accumulation is **thermally-driven demagnetization**
- **4 Layers** Pt/[Co,Pt]/Cu/CoFeB
 - Measure magnetization dynamics by MOKE
 - Additional spin transfer torque coming from the **spin-dependent Seebeck effect**

Detect spin accumulation and magnetization dynamics by time-resolved magneto-optic Kerr effect (TR-MOKE)



Thermal circuit for picosecond spin Seebeck effect



Difficult to directly compare the volume and interface thermal conductances (different units)

- At room temperature

- electron-phonon coupling in Cu

$$g_{ep} \approx 8 \times 10^{16} \text{ W m}^{-3} \text{ K}^{-1}$$

- magnon-phonon coupling in a cuprate spin ladder

$$g_{em} \approx 5 \times 10^{15} \text{ W m}^{-3} \text{ K}^{-1}$$

- phonon-phonon interface conductance

$$G_{pp} \approx 200 \text{ MW m}^{-2} \text{ K}^{-1}$$

- tentative estimate from our data

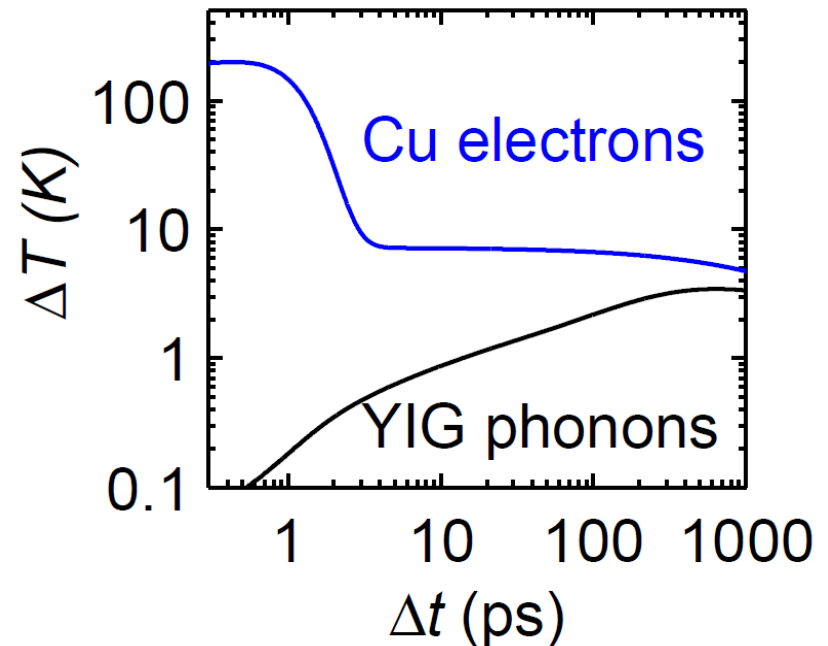
$$G_{em} \approx \left(10^8 \text{ A m}^{-2} \text{ K}^{-1} \right) \left(\frac{k_B T}{e} \right) \sim 2 \text{ MW m}^{-2} \text{ K}^{-1}$$

First solve the heat problem

Solve two-temperature model in Cu and couple to phonons in YIG through an interface thermal conductance.

For Cu:

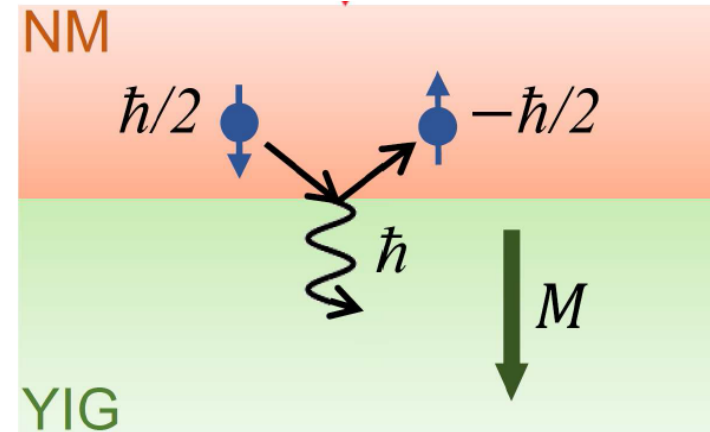
$$C_e \frac{\partial T_e}{\partial t} - \Lambda_e \frac{\partial^2 T_e}{\partial z^2} = g_{ep}(T_p - T_e)$$
$$C_p \frac{\partial T_p}{\partial t} - \Lambda_p \frac{\partial^2 T_p}{\partial z^2} = g_{ep}(T_e - T_p)$$



Then solve the spin diffusion problem using the spin-Seebeck effect as a boundary condition at the Cu/YIG interface

$$j_S = g_{\uparrow\downarrow} \frac{e^2}{h} S_S (T_e - T_m) \quad S_S = \left(\frac{\gamma \hbar}{\pi M_s V_a} \right) \left(\frac{k_B}{e} \right)$$

$$\alpha \equiv g_{\uparrow\downarrow} \frac{e^2}{h} S_S$$



$$j_{\uparrow} - j_{\downarrow} = \frac{\sigma}{2e} \frac{\partial(\xi_{\uparrow} - \xi_{\downarrow})}{\partial x}$$

$$\frac{\partial(\xi_{\uparrow} - \xi_{\downarrow})}{\partial t} = \frac{2}{eN} \frac{\partial(j_{\uparrow} - j_{\downarrow})}{\partial x} = - \frac{\xi_{\uparrow} - \xi_{\downarrow}}{\tau}$$

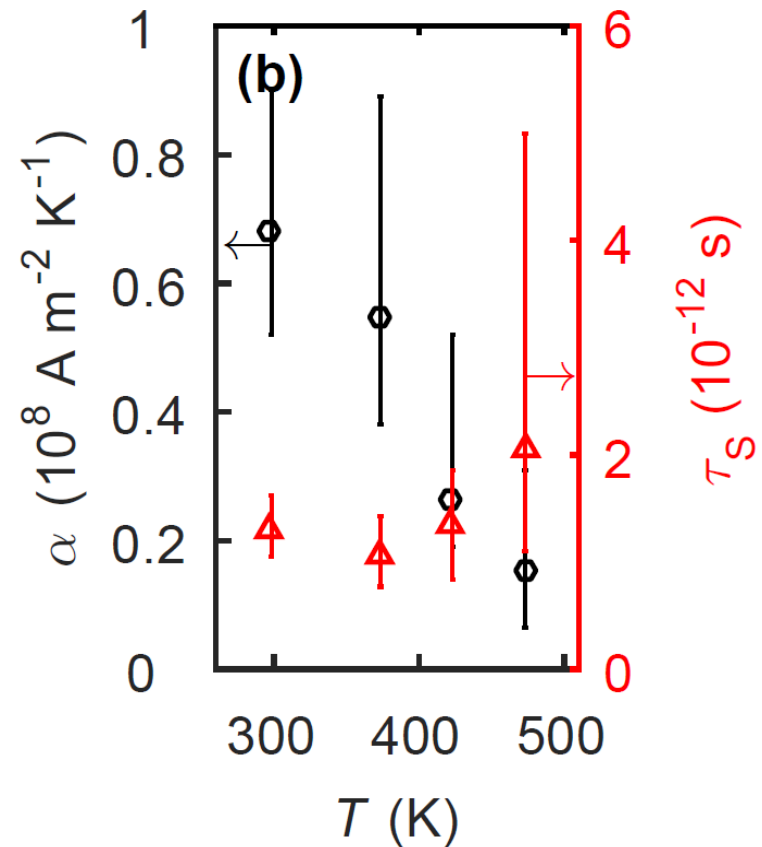
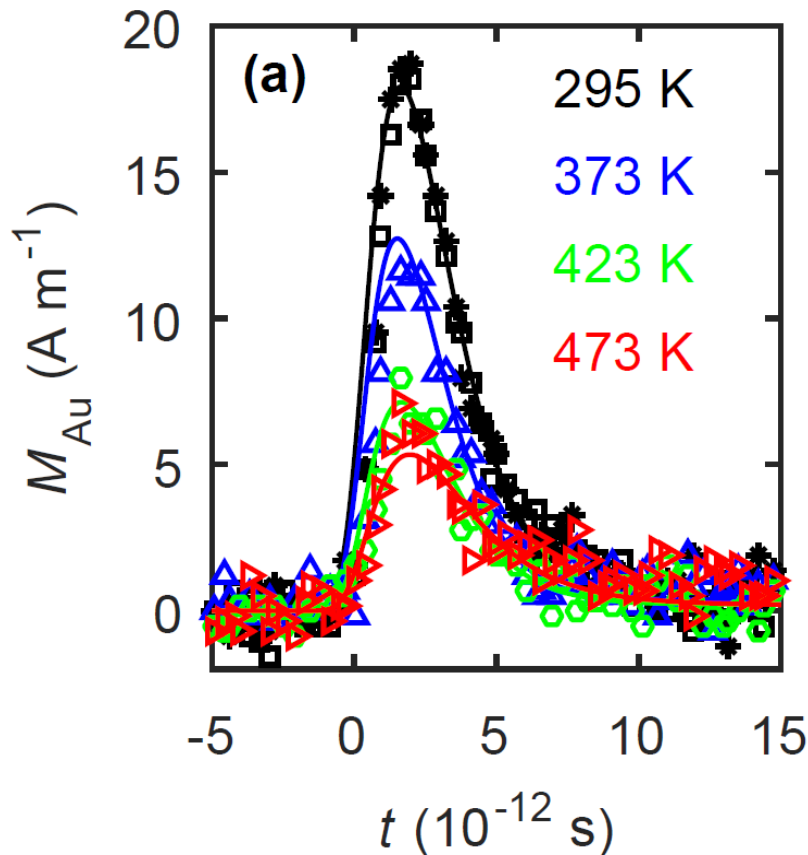
Spin relaxation time
Fit parameter

Measure spin accumulation in Cu or Au by the polar Kerr effect and convert to magnetization using a previously determined calibration

$$\text{Cu: } \theta_K \approx (10 \text{ nrad m A}^{-1})M$$

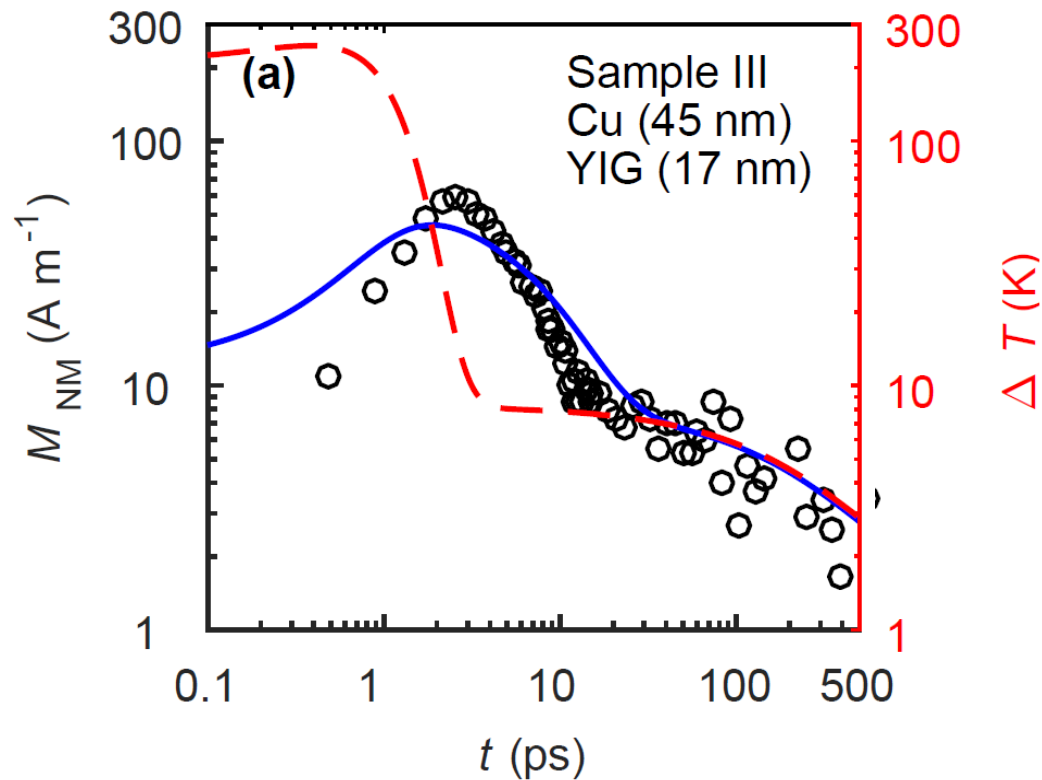
$$\text{Au: } \theta_K \approx (50 \text{ nrad m A}^{-1})M$$

Au(60 nm)/YIG(20 nm)



Long time decay of signal is consistent with signal proportional to interface ΔT

At $t > 3$ ps, $(T_e - T_m) \approx (T_{Cu} - T_{YIG})$



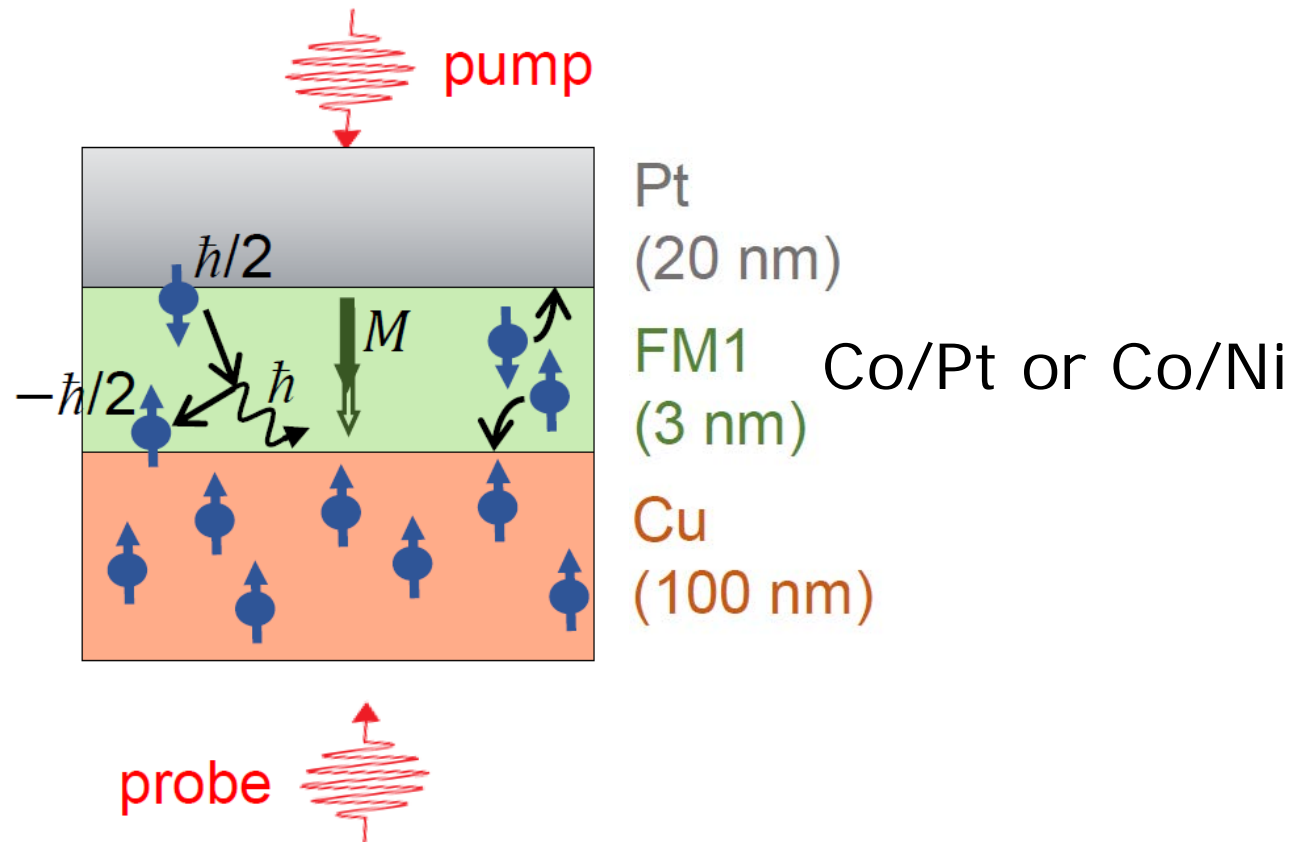
Need better systematic control of the metal/YIG interface. Some tentative conclusions from 6 samples

$$\alpha \sim 10^8 \text{ A m}^{-2} \text{ K}^{-1}$$

- α independent of YIG thickness
- α independent of metal thickness
- α larger for Cu/YIG than for Au/YIG
- in-situ deposited Au has higher α than ex-situ

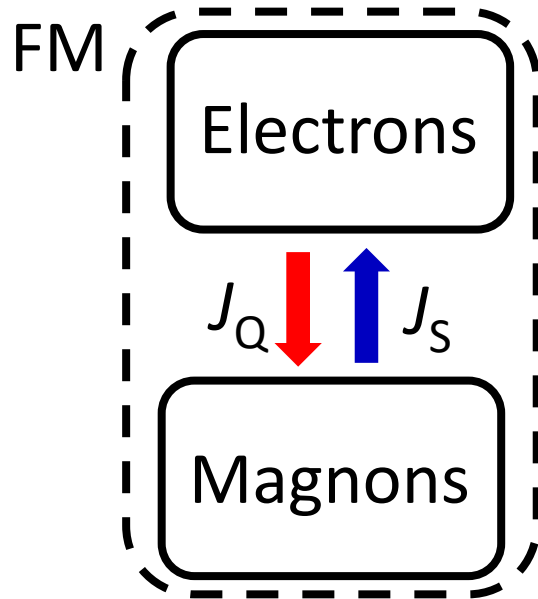
	Sample I	Sample II	Sample III	Sample IV	Sample V	Sample VI
NM	Au	Au	Cu	Au	Au	Cu
h_{NM} (nm)	60	60	45	103	29	35
h_{YIG} (nm)	20	100	17	50	51	17
α ($10^8 \text{ A m}^{-2} \text{ K}^{-1}$)	0.84 ± 0.12	0.66 ± 0.29	3.02 ± 1.05	0.29 ± 0.11	0.30 ± 0.05	2.32 ± 0.24
τ_{S} (ps)	1.14 ± 0.13	0.99 ± 0.26	3.79 ± 0.85	2.67 ± 0.91	1.74 ± 0.29	2.52 ± 0.27

3 layer metal structure: Place a perpendicular metallic ferromagnet layer between a Pt heater and a Cu heat sink



Two mechanisms for thermally-driven spin generation

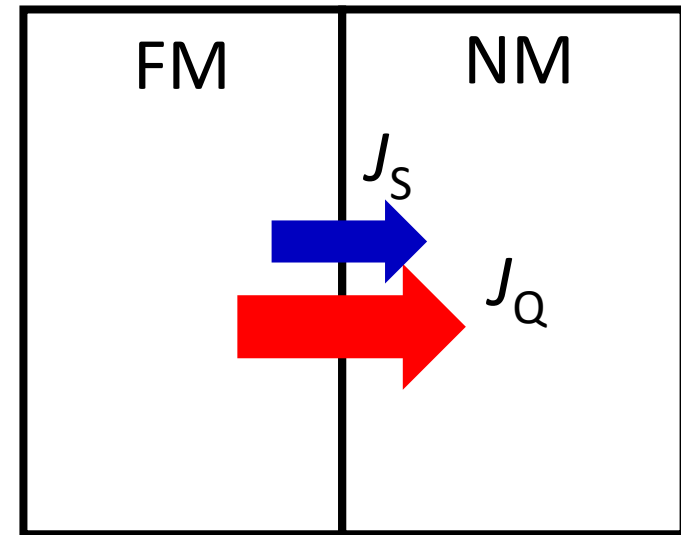
Ultrafast demagnetization



$$g_S = -\frac{dM}{dt}$$

Choi, *et al.* Nature Commun. **5**, 4334 (2014)

Spin-dependent Seebeck effect



First approximation: treat as an interface spin source

$$G_S = -\left(\frac{\mu_B}{eLT}\right) S_S J_Q$$

Slachter, *et al.* Nature Phys. **6**, 879 (2010)

Choi, *et al.* Nature Phys. **11**, 576 (2015)

FM layer thickness is actually comparable to the spin diffusion length (particularly for Co/Ni) drop the boundary condition approximation

spin Seebeck coefficient

$$S_S = S_{\uparrow} - S_{\downarrow}$$

spin current

$$j_{\uparrow} - j_{\downarrow} = \frac{2\sigma_{\uparrow}\sigma_{\downarrow}}{e(\sigma_{\uparrow} + \sigma_{\downarrow})} \left[\frac{\partial\zeta_{\uparrow} - \zeta_{\downarrow}}{\partial z} - eS_S \frac{\partial T}{\partial z} \right],$$

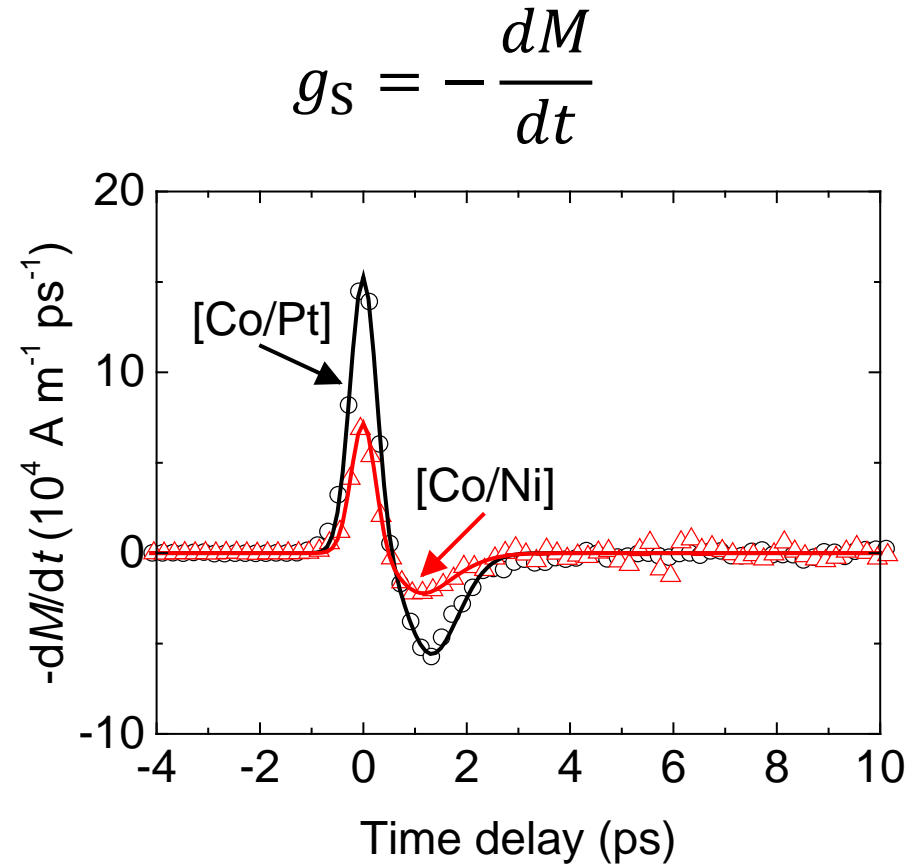
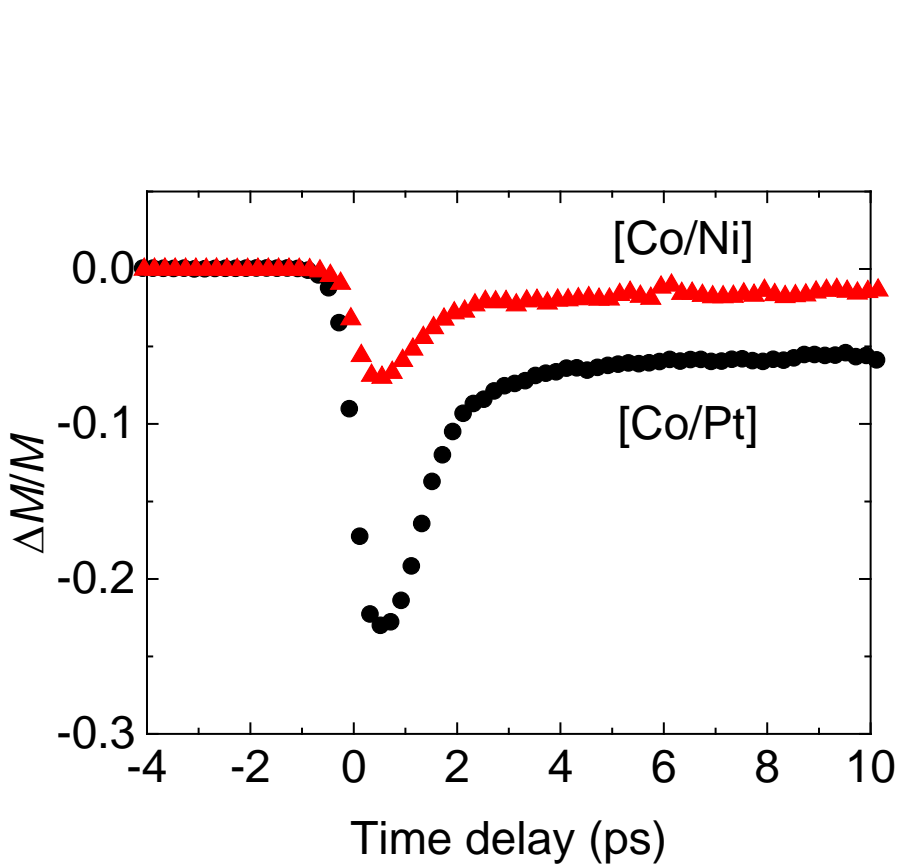
spin chemical potential

$$\frac{\partial(\zeta_{\uparrow} - \zeta_{\downarrow})}{\partial t} - D \left[\frac{\partial^2(\zeta_{\uparrow} - \zeta_{\downarrow})}{\partial z^2} - eS_S \frac{\partial^2 T}{\partial z^2} \right] = -\frac{\zeta_{\uparrow} - \zeta_{\downarrow}}{\tau_S}$$

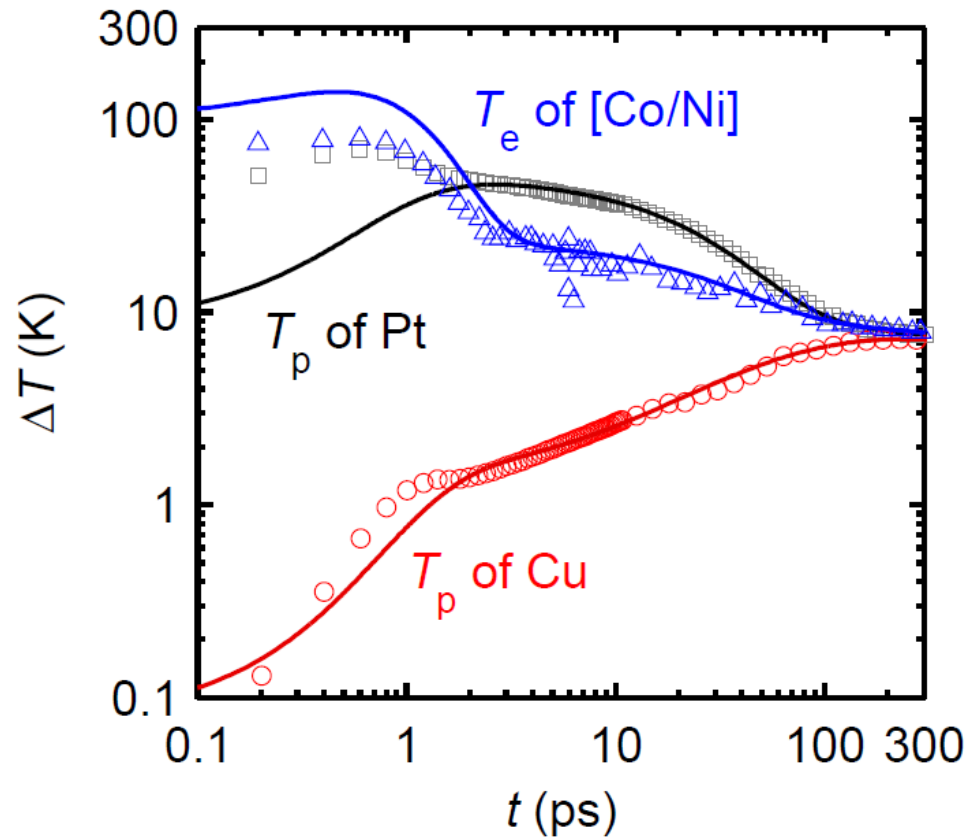
• Notes $j = j_{\uparrow} + j_{\downarrow} = 0$

- Zero charge current is a good approximation
- These equations assume the same T for both spin populations. Will return to this point later...
- Choi et al. (2015) used a different definition of S_S

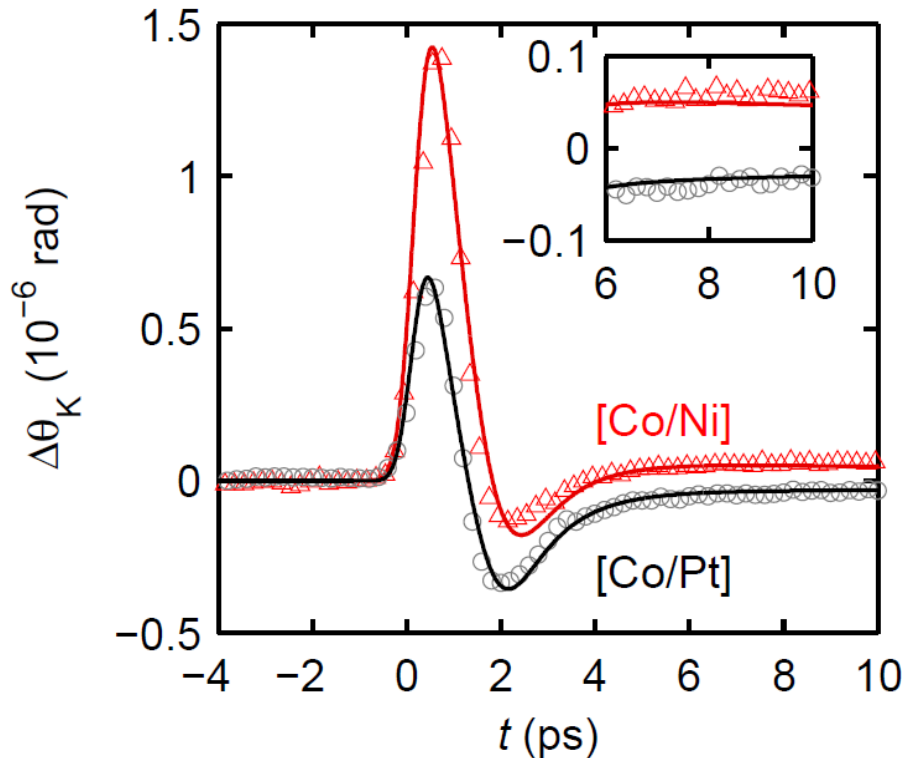
Measure $M(t)$ of FM1 by TR-MOKE



Measure temperatures of Pt and Cu by time domain thermoreflectance (TDTR) and use data to refine thermal model for electron and phonon temperatures



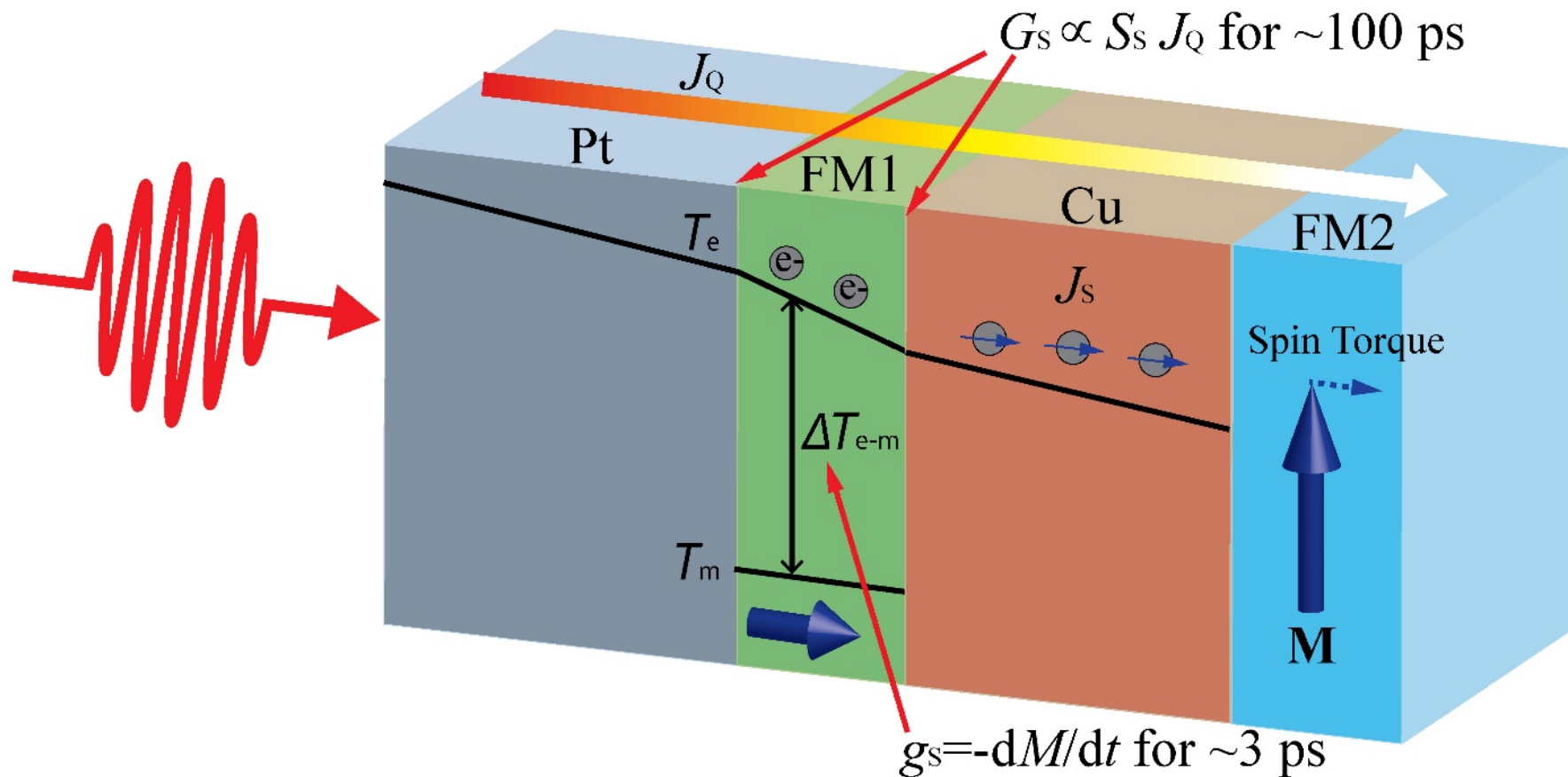
Measure spin accumulation in Cu by TR-MOKE and compare to spin diffusion model



- Three fitting parameters for each sample.
 1. spin relaxation time in FM1
 2. S_S
 3. coefficient relating Kerr rotation and spin accumulation ($10 \text{ nrad A}^{-1} \text{ m}$)

4-layer structure: Thermal spin-transfer torque

Pt (20)/ [Co/Pt] or [Co/Ni] (3)/ Cu (100)/ CoFeB (2) (nm)

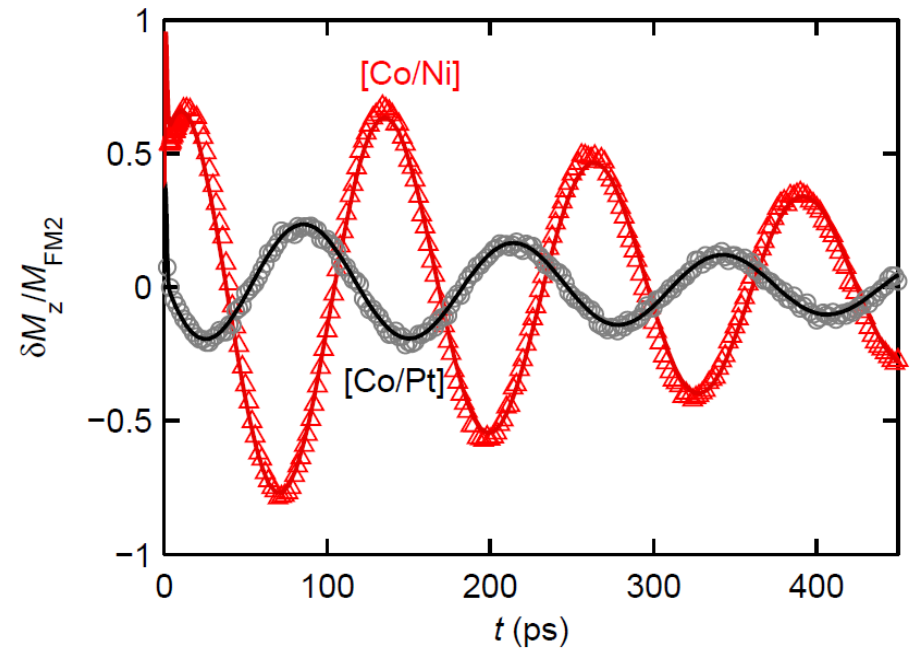
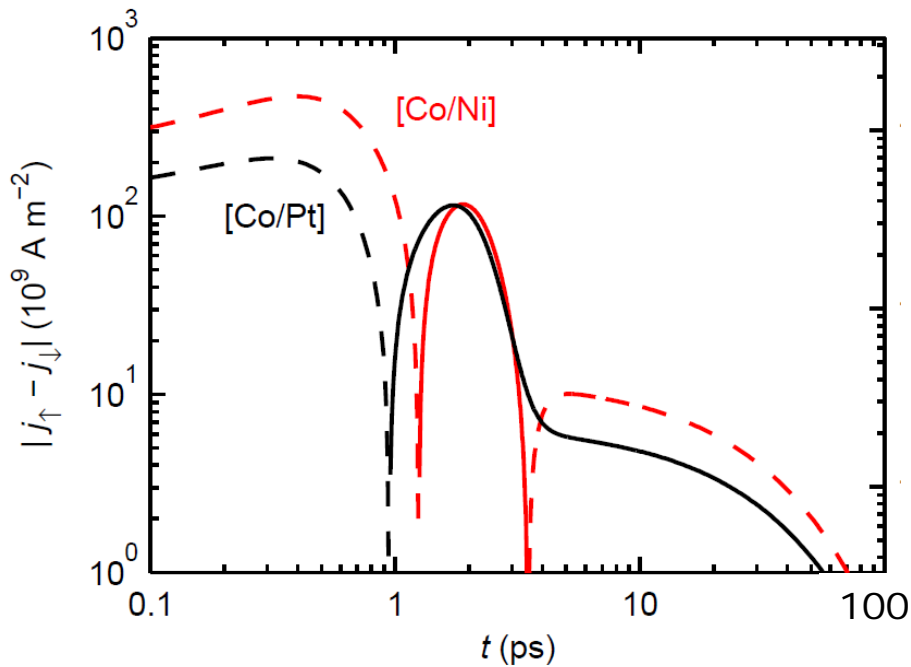


Choi, *et al.* Nature Physics (2015)

Kimling, *et al.* (submitted)

Model spin current (FM2 is a perfect sink of spin) and magnetization dynamics created by the spin transfer torque. Compare to experiment

$$\frac{\partial \mathbf{m}}{\partial t} = -\gamma_e \mu_0 \mathbf{m} \times \mathbf{H}_{\text{eff}} + \alpha_G \mathbf{m} \times \frac{\partial \mathbf{m}}{\partial t} + \frac{\mu_B (j_{\uparrow} - j_{\downarrow})}{e M_{\text{Sh}}} \mathbf{m} \times (\mathbf{m} \times \mathbf{e}_z)$$



Two free parameters: spin relaxation time τ_s and spin-dependent Seebeck coefficient S_s

	Fitting parameter	[Co/Pt]	[Co/Ni]
Nat. Phys (2015)	τ_s (fs)	20	100
	S_s ($\mu\text{V}/\text{K}$)	12	-24
	Fitting parameter	[Co/Pt]	[Co/Ni]
Pt/FM1/Cu Cu spin accumulation	τ_s (fs)	22	108
	S_s ($\mu\text{V}/\text{K}$)	22	-25
	Fitting parameter	[Co/Pt]	[Co/Ni]
Pt/FM1/Cu/FM2 FM2 precession	τ_s (fs)	19	140
	S_s ($\mu\text{V}/\text{K}$)	19	-25

Assumption of equal temperatures for up and down spin channels is probably not a valid approximation

spin current

$$j_{\uparrow} - j_{\downarrow} = \frac{2\sigma_{\uparrow}\sigma_{\downarrow}}{(\sigma_{\uparrow} + \sigma_{\downarrow})} \left(-S_{\uparrow} \frac{\partial T_{\uparrow}}{\partial z} + S_{\downarrow} \frac{\partial T_{\downarrow}}{\partial z} \right)$$

assume W-F law holds for both channel

$$j_{\uparrow} - j_{\downarrow} = \frac{2(\Lambda_{\uparrow}S_{\downarrow}q_{\downarrow} - \Lambda_{\downarrow}S_{\uparrow}q_{\uparrow})}{(\Lambda_{\uparrow} + \Lambda_{\downarrow})L_0T_e}$$

Rewrite in terms of asymmetry parameters β for material properties (Seebeck S and thermal conductivity Λ) and instantaneous heat current q .

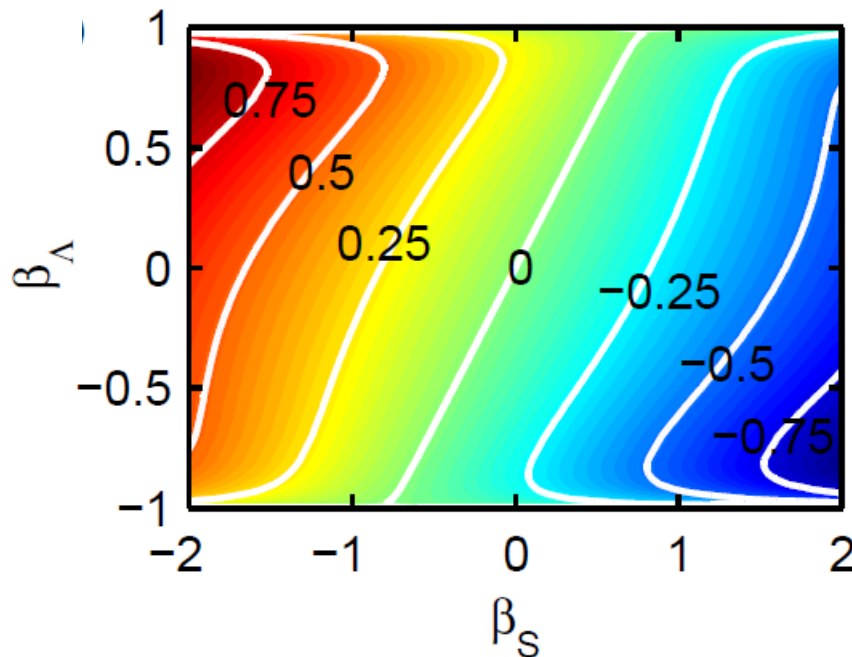
$$\beta_S = (S_{\uparrow} - S_{\downarrow}) / (S_{\uparrow} + S_{\downarrow})$$

$$j_{\uparrow} - j_{\downarrow} = \frac{Sq_e}{2L_0T_e} [(\beta_{\Lambda}\beta_S - 1)\beta_q + (\beta_{\Lambda} - \beta_S)]$$

Complicated phase space of material parameters.
Take home message is that spin heat accumulation
can help generate larger spin currents

Precession amplitude increases with
the time integral of spin current

$$I = \int (j_{\uparrow} - j_{\downarrow}) dt$$



Contours of constant I
in units (A s m^{-2})

Summary

- Picosecond second time-scale isolates the interface contribution to spin Seebeck effect at normal-metal/ferromagnetic-insulator interfaces.
 - Order of magnitude of the coefficient is consistent with theory and prior measurements of spin mixing conductance.
- In metallic structures, ultrafast demagnetization and spin-dependent Seebeck S_S are of similar absolute magnitude.
 - For [Co,Ni] the two mechanisms reinforce each other and produce a 1% tilting of the magnetization in spin-valve structure.
- S_S coefficient can be measured accurately.
 - Difficult, however, to relate to microscope parameters that describe the Seebeck coefficients and thermal conductivities of the spin-up and spin-down channels (if the two channels are indeed not at the same temperature).