### Picosecond spin caloritronics

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#### Spintronics in 1985

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

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### 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.



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

### 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  $\Lambda$ 

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

• Interface thermal conductance G,  $\Delta T$ =temperature across an interface

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

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

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

### Outline

- 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

YIG GGG Cu Laser substrate or pulse Au  $C_{el}$ Laser Cu YIG  $C_m$ pulse electrons magnons  $G_{em}$  $g_{ep}$  $g_{mp}$ phonons Cu phonons  $C_{ph}$  $C_{ph}$  $G_{PP}$ 

## 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

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

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



Kimling et al., arXiv:1608.00702

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

$$j_{\rm S} = g_{\uparrow\downarrow} \frac{e^2}{h} S_{\rm S} (T_{\rm e} - T_{\rm m}) \quad S_{\rm S} = \left(\frac{\gamma h}{\pi M_{\rm s} V_{\rm a}}\right) \left(\frac{k_{\rm B}}{e}\right)$$

 $\alpha \equiv g_{\uparrow\downarrow} \frac{e^2}{h} S_{\rm S}$ 





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



### Long time decay of signal is consistent with signal proportional to interface $\Delta T$

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

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

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

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

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 Spin-dependent Seebeck effect



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



First approximation: treat as an interface spin source

$$G_S = -\left(\frac{\mu_{\rm B}}{eLT}\right)S_s J_{\rm Q}$$

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

- $S_{\rm S} = S_{\uparrow} S_{\downarrow}$ spin Seebeck coefficient
- spin current

$$\begin{array}{ll} \text{spin current} & j_{\uparrow} - j_{\downarrow} = \frac{2\sigma_{\uparrow}\sigma_{\downarrow}}{e(\sigma_{\uparrow} + \sigma_{\downarrow})} \left[ \frac{\partial\zeta_{\uparrow} - \zeta_{\downarrow}}{\partial z} - eS_{\mathrm{S}}\frac{\partial T}{\partial z} \right], \\ \text{spin chemical} & \frac{\partial(\zeta_{\uparrow} - \zeta_{\downarrow})}{\partial t} - D \left[ \frac{\partial^{2}(\zeta_{\uparrow} - \zeta_{\downarrow})}{\partial z^{2}} - eS_{\mathrm{S}}\frac{\partial^{2}T}{\partial z^{2}} \right] = -\frac{\zeta_{\uparrow} - \zeta_{\downarrow}}{\tau_{\mathrm{S}}} \end{array}$$

- 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  $S_{s}$

Kimling, et al. (submitted)

TS

### 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<sub>S</sub>
  - coefficient relating Kerr rotation and spin accumulation (10 nrad A<sup>-1</sup> m)

Choi et al. Nature Physics (2015); Kimling et al. (submitted)

### 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



Choi, et al. Nature Physics (2015), Kimling, et al. (submitted)

Two free parameters: spin relaxation time  $\tau_s$  and spin-dependent Seebeck coefficient  $S_s$ 

	Fitting parameter	[Co/Pt]	[Co/Ni]
$N_{ab} = 0$	$\tau_{s}$ (fs)	20	100
Nat. Phys (2015)	<i>S<sub>S</sub></i> (μV/K)	12	-24
	Fitting parameter	[Co/Pt]	[Co/Ni]
Pt/FM1/Cu	$\tau_{S}$ (fs)	22	108
Cu spin accumulation	<i>S<sub>S</sub></i> (μV/K)	22	-25
	Fitting parameter	[Co/Pt]	[Co/Ni]
Pt/FM1/Cu/FM2	$\tau_{S}$ (fs)	19	140
FM2 precession	<i>S<sub>S</sub></i> (μV/K)	19	-25

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

$$\begin{array}{ll} \text{spin current} & j_{\uparrow} - j_{\downarrow} = \frac{2\sigma_{\uparrow}\sigma_{\downarrow}}{(\sigma_{\uparrow} + \sigma_{\downarrow})} \left( -S_{\uparrow} \frac{\partial T_{\uparrow}}{\partial z} + S_{\downarrow} \frac{\partial T_{\downarrow}}{\partial z} \right) \\ \text{assume W-F law holds} & j_{\uparrow} - j_{\downarrow} = \frac{2(\Lambda_{\uparrow}S_{\downarrow}q_{\downarrow} - \Lambda_{\downarrow}S_{\uparrow}q_{\uparrow})}{(\Lambda_{\uparrow} + \Lambda_{\downarrow})L_{0}T_{\text{e}}} \end{array}$$

Rewrite in terms of asymmetry parameters  $\beta$  for material properties (Seebeck *S* and thermal conductivity  $\Lambda$ ) and instantaneous heat current *q*.

$$\beta_S = (S_{\uparrow} - S_{\downarrow})/(S_{\uparrow} + S_{\downarrow})$$
$$j_{\uparrow} - j_{\downarrow} = \frac{Sq_{\rm e}}{2L_0T_{\rm e}} \left[ (\beta_{\Lambda}\beta_S - 1)\beta_q + (\beta_{\Lambda} - \beta_S) \right]$$

Kimling *et al*. (submitted)

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

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





- 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 spindependent 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 spinvalve structure.
- S<sub>S</sub> 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).