

# Coherent sub-THz Spin Pumping from an Insulating Antiferromagnet

*Vaidya et al. Science* **368**, 160–165 (2020)

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*Department of Physics – UCF*  *Orlando*

de  Barco

Nanomagnetism and

LAB

Spintronics



*Work done in collaboration with:*

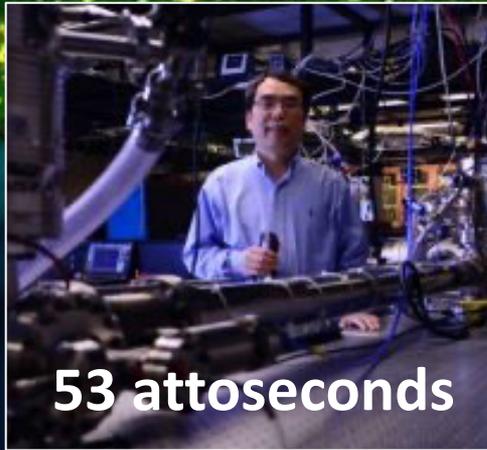
**David Lederman** (University of California – Santa Cruz)

**Ran Cheng** (University of California – Riverside)

**Arne Brataas** (Norwegian University of Science and Technology)

**Hans van Tol** (National High Magnetic Field Laboratory / NHMFL – Tallahassee/FL)

# College of Sciences



53 attoseconds



**UCF**

**ORLANDO**





Justin Zakhary



Ran Liu



Cameron Nickle



Francis Adoah



Yan Liu



Jaesuk Kwon



Priyanka Vaidya

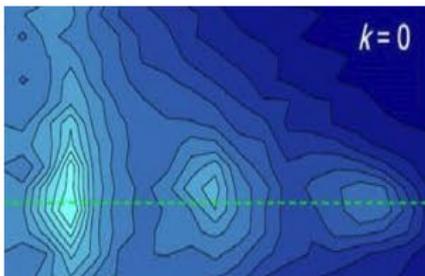


Gyan Khatri



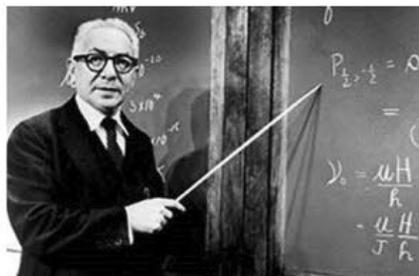
Greg Fritjofson

## MOLECULAR MAGNETISM – QUANTUM DYNAMICS OF SPIN



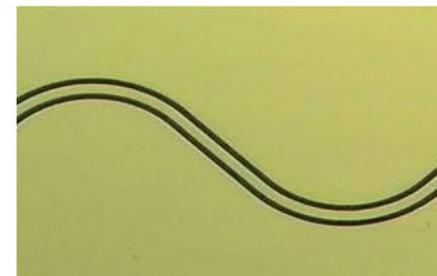
QUANTUM TUNNELING OF SPIN

Low-nuclearity molecules allow for a full numerical treatment of the multi-spin Hamiltonian, where degrees of freedom associated with the individual ions can be adequately tested.



RABI OSCILLATIONS IN ANISOTROPIC SPINS

The commutation between spin anisotropy operators leads to complex dynamics where the magnetic state of the molecule may develop excited precessional states.

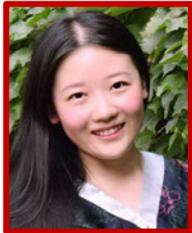


SPIN QUANTUM ELECTRODYNAMICS

High-Q superconducting resonators designed with nano-constrictions may allow the coupling a photon cavity mode with a single molecular spin.



Justin Zakhary



Ran Liu



Cameron Nickle



Francis Adoah



Yan Liu



Jaesuk Kwon



Priyanka Vaidya

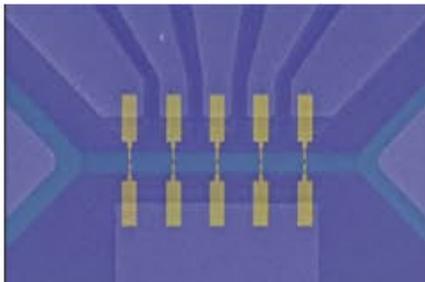


Gyan Khatri



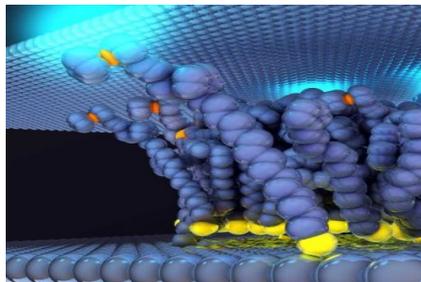
Greg Fritjofson

## MOLECULAR ELECTRONICS – SINGLE-MOLECULE TUNNEL JUNCTIONS



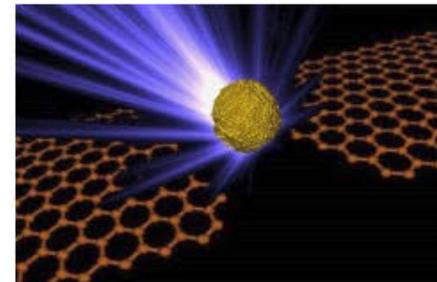
**SINGLE-ELECTRON TRANSISTORS**

Fabrication of electromigration-broken three-terminal single-electron transistors for transport spectroscopy at the individual molecule level.



**CHARGE RECTIFICATION IN MOLECULAR TRANSISTORS**

Study of charge rectification in tunnel junctions bridged by molecular chains including a ferrocene active conductive unit placed asymmetrically within the chain.



**OPTOELECTRONICS AT THE NANOSCALE**

Study of the transport properties of individual noble-metal nanoparticles in the presence of optical irradiation inducing plasmonic resonance.



Justin Zakhary



Ran Liu



Cameron Nickle



Francis Adoah



Yan Liu



Jaesuk Kwon



Priyanka Vaidya

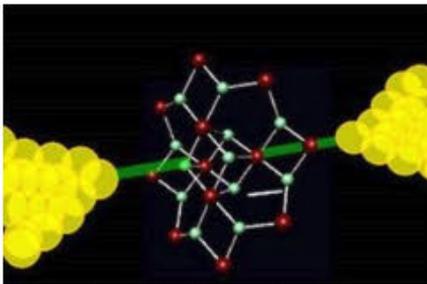


Gyan Khatri



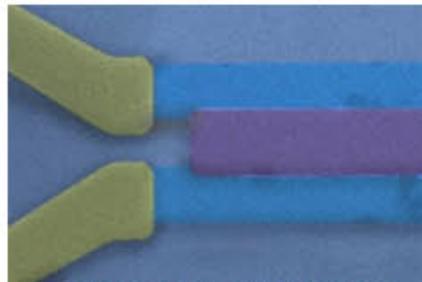
Greg Fritjofson

## NANOSCALE SPINTRONICS – 0D, 2D and 3D SPIN TRANSPORT



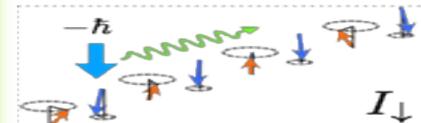
**SINGLE-ELECTRON TRANSPORT THROUGH MOLECULAR MAGNETS**

Investigation of the interplay between high-spin states of an individual SMM and conduction electrons in a three-terminal single-electron transistor.



**TWO-DIMENSIONAL SPINTRONICS**

An study aimed at understanding and controlling the injection of pure spin currents in graphene-based spintronics devices.



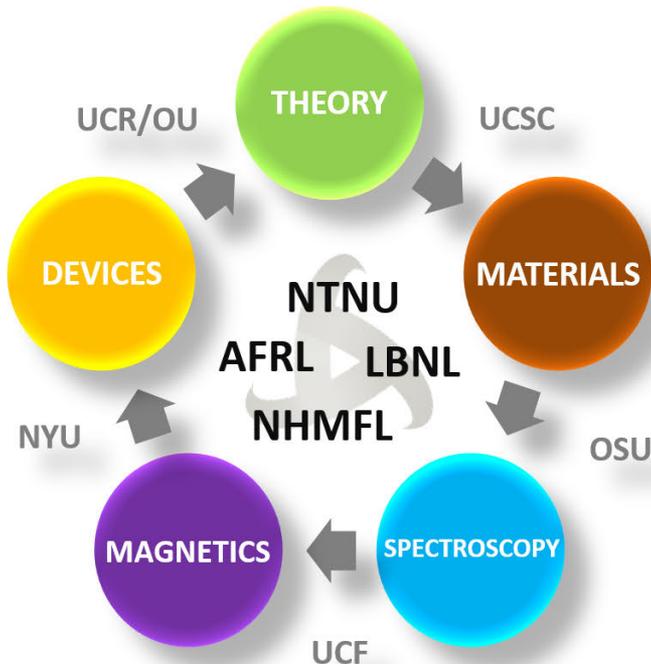
**ANTIFERROMAGNETIC SPINTRONICS**

Terahertz spintronics with antiferromagnetic insulators.

### Collaborative Effort – Spintronics with AFM insulators



# AFOSR MURI TEAM



#### Experiment:

- PI-Enrique del Barco (UCF)
- PI-Chris Hammel (OSU)
- PI-Andrew Kent (NYU)
- C-Hans van Tol (NHMFL)
- C-Simranjeet Singh (CMU)
- C-Robert Peale (UCF)
- C-Michael Chini (UCF)

#### Materials:

- PI-David Lederman (UCSC)
- PI-Fengyuan Yang (OSU)
- C-Michael Page (AFRL)
- C-Madhab Neupane (UCF)
- C-Hendrik Ohldag (LBNL)

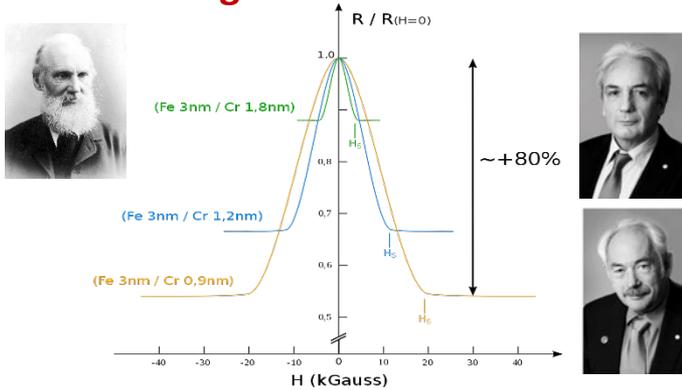
#### Theory:

- PI-Ran Cheng (UCR)
- PI-Andrei Slavin (OU)
- C-Arne Brataas (NTNU–Norway)

### What AFM materials have to offer in spintronics?

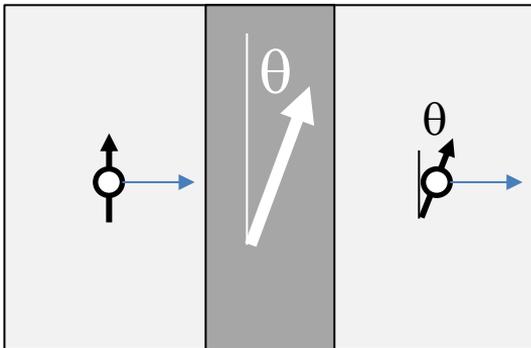
### Ferromagnetic spintronics / STT-based MRAM

#### Magnetoresistance



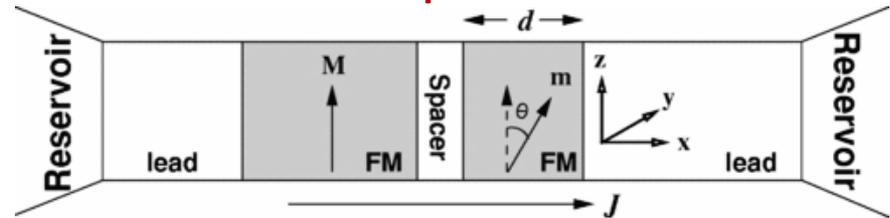
W. Thompson-5% (1856) 2007 Noble Prize: Fert-Grünberg-80% (1988)

#### Spin Transfer Torque

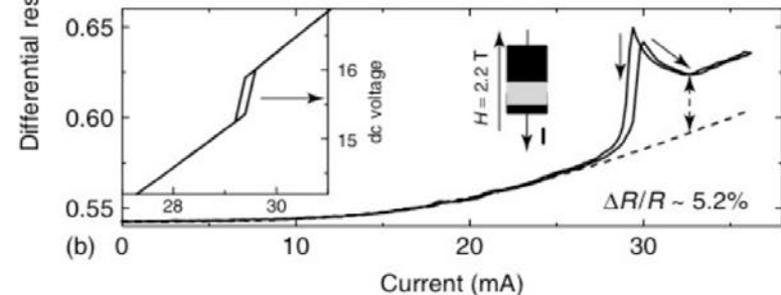
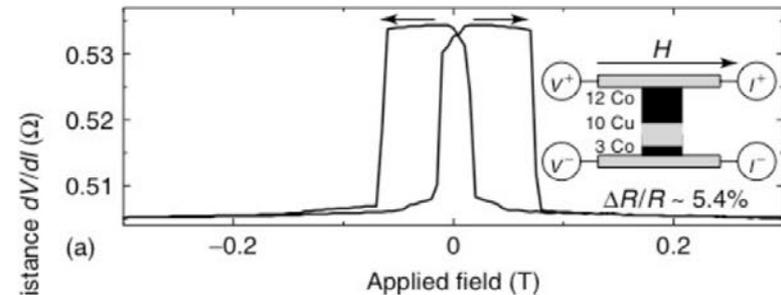


Slonczewski (1989), (1996) and Berger (1996)

#### STT spin valve



Jiang Xiao, A. Zangwill, and M. D. Stiles. PRB (2005)

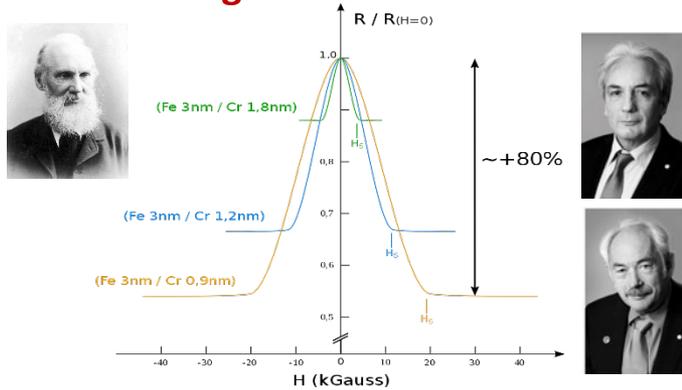


Ozylmaz, Kent, Monsma, Sun, Rooks, and Koch. PRL (2003)

### What AFM materials have to offer in spintronics?

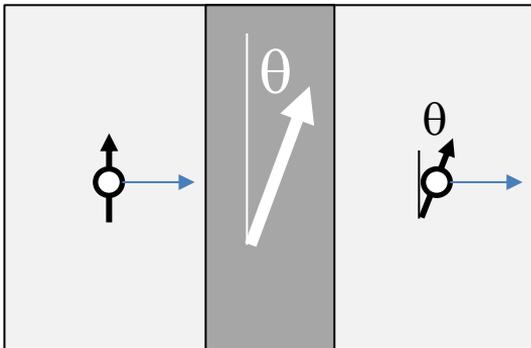
### Ferromagnetic spintronics / STT-based GHz oscillator

#### Magnetoresistance

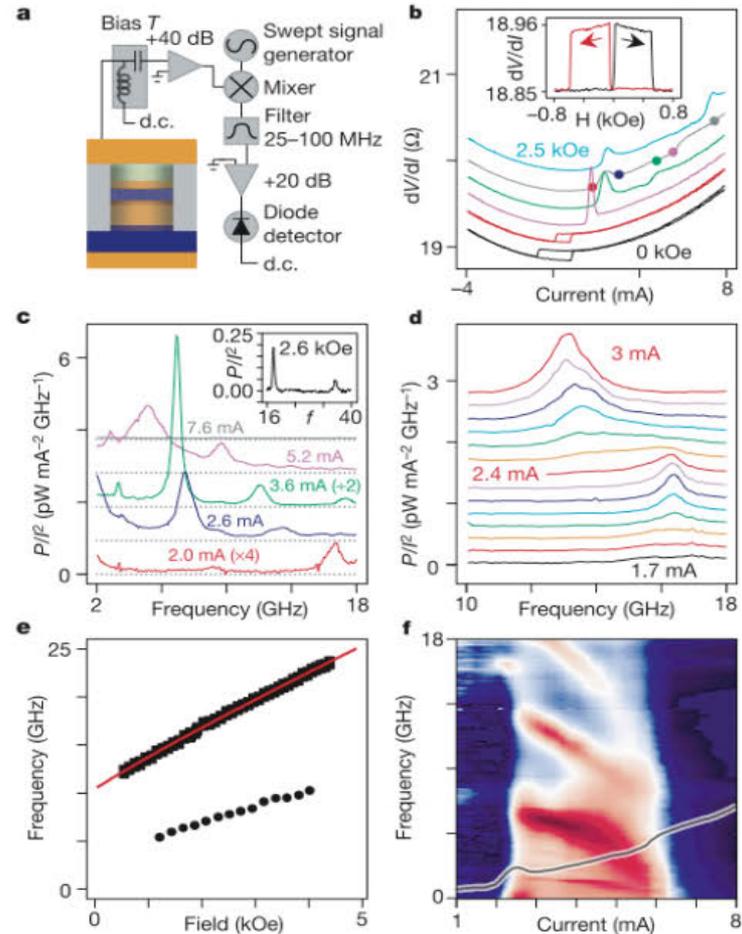


W. Thomson-5% (1856) 2007 Noble Prize: Fert-Grünberg-80% (1988)

#### Spin Transfer Torque



Slonczewski (1989), (1996) and Berger (1996)

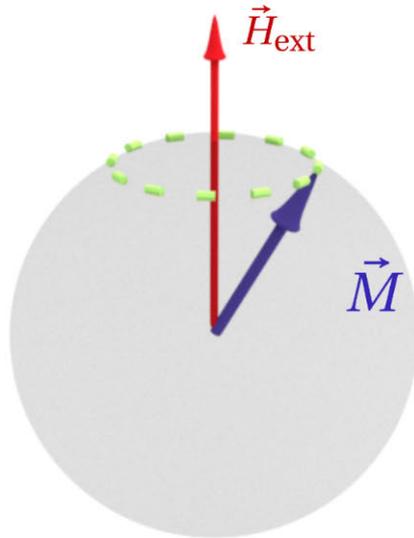


Kiselev et al. Nature (2003)

### What AFM materials have to offer in spintronics?

- ✓  $f = 0.3\text{-}3\text{ THz}$   
Operation in the THz gap
- ✓ No stray fields / No cross-talk
- ✓ nm-scale magnon waves  
Miniaturization of magnonic circuits
- ✓ Group velocity of magnons  $\propto \sqrt{f}$   
Faster data transmission rates
- ✓ High frequency / Short Times  
Fast operations (picoseconds)

What AFM materials have to offer in spintronics?



$$H_{\text{ext}} \approx 1\text{T}$$

$$\omega_{\text{FMR}} = \gamma \sqrt{H_{\text{ext}} H_{\text{dip}}} \approx 1 - 20\text{GHz}$$

Ferromagnet

### What AFM materials have to offer in spintronics?

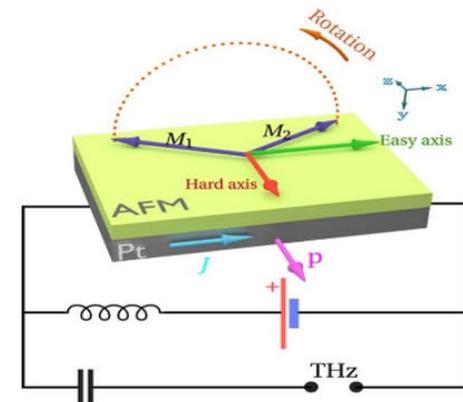
### TUNABLE AFM OSCILLATOR

Cheng et al. PRL 2016 / Khymyn et al. Sci. Rep. (2017)

$$\begin{aligned}
 d\mathbf{M}_1/dt &= \underbrace{\gamma[\mathbf{H}_1 \times \mathbf{M}_1]}_{\text{damping}} + \frac{\alpha}{M_s} [\mathbf{M}_1 \times d\mathbf{M}_1/dt] + \underbrace{\frac{\tau}{M_s} [\mathbf{M}_1 \times [\mathbf{M}_1 \times \mathbf{p}]]}_{\text{Spin torque}}, \\
 d\mathbf{M}_2/dt &= \gamma[\mathbf{H}_2 \times \mathbf{M}_2] + \frac{\alpha}{M_s} [\mathbf{M}_2 \times d\mathbf{M}_2/dt] + \frac{\tau}{M_s} [\mathbf{M}_2 \times [\mathbf{M}_2 \times \mathbf{p}]]
 \end{aligned}$$

$$\begin{aligned}
 \mathbf{H}_1 &= \frac{1}{M_s} \left[ -\frac{1}{2} H_{\text{ex}} \mathbf{M}_2 + H_h \mathbf{n}_h (\mathbf{n}_h \cdot \mathbf{M}_1) - H_e \mathbf{n}_e (\mathbf{n}_e \cdot \mathbf{M}_1) \right] \\
 \mathbf{H}_2 &= \frac{1}{M_s} \left[ -\frac{1}{2} H_{\text{ex}} \mathbf{M}_1 + H_h \mathbf{n}_h (\mathbf{n}_h \cdot \mathbf{M}_2) - H_e \mathbf{n}_e (\mathbf{n}_e \cdot \mathbf{M}_2) \right]
 \end{aligned}$$

exchange
biaxial anisotropy



### What AFM materials have to offer in spintronics?

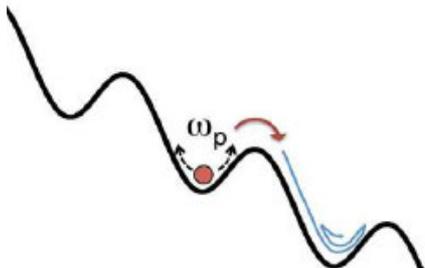
### TUNABLE AFM OSCILLATOR

Cheng et al. PRL 2016 / Khymyn et al. Sci. Rep. (2017)

$$\vec{l} = \frac{M_1 - M_2}{2M_s} = \vec{x} \cos \phi(t) + \vec{y} \sin \phi(t)$$

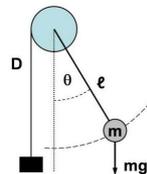
$$\underbrace{\frac{1}{\omega_{ex}} \ddot{\phi}}_{\substack{\text{exchange} \\ (\text{inertia}=\ll\text{mass}\gg)}} + \underbrace{\alpha \dot{\phi}}_{\text{damping}} + \underbrace{\frac{\omega_e}{2} \sin 2\phi}_{\text{anisotropy}} = \underbrace{\sigma j}_{\text{Spin torque}}$$

#### Tilted washboard potential



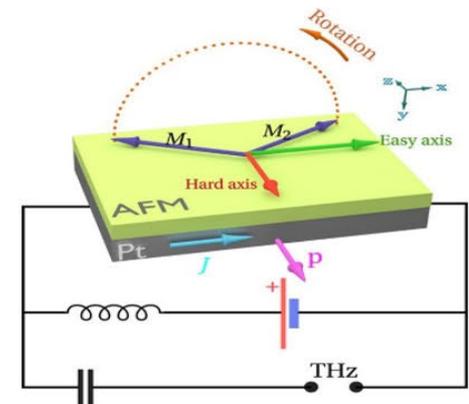
#### RC Josephson Junction

$$\ddot{\phi} + \frac{1}{RC} \dot{\phi} + \omega_J^2 \sin \phi = \omega_J^2 \gamma$$



#### Torqued pendulum

$$\Theta \ddot{\theta} + \Gamma \dot{\theta} + mg\ell \sin \theta = D$$



### What AFM materials have to offer in spintronics?

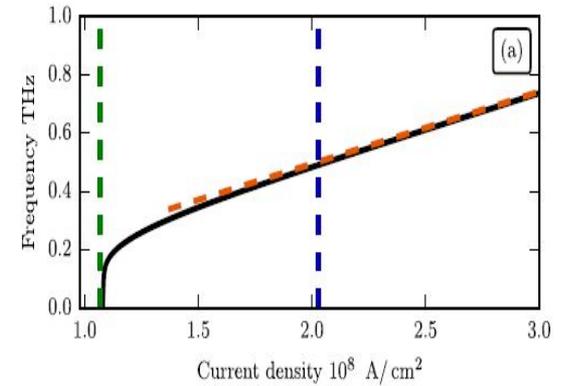
### TUNABLE AFM OSCILLATOR

Cheng et al. PRL 2016 / Khymyn et al. Sci. Rep. (2017)

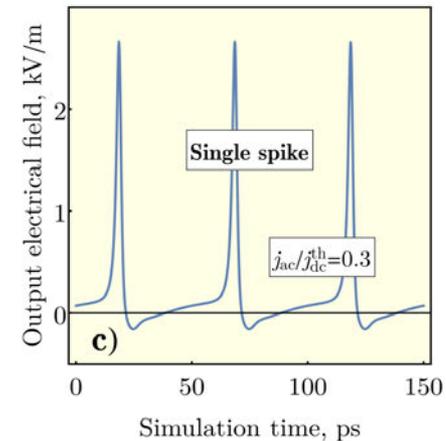
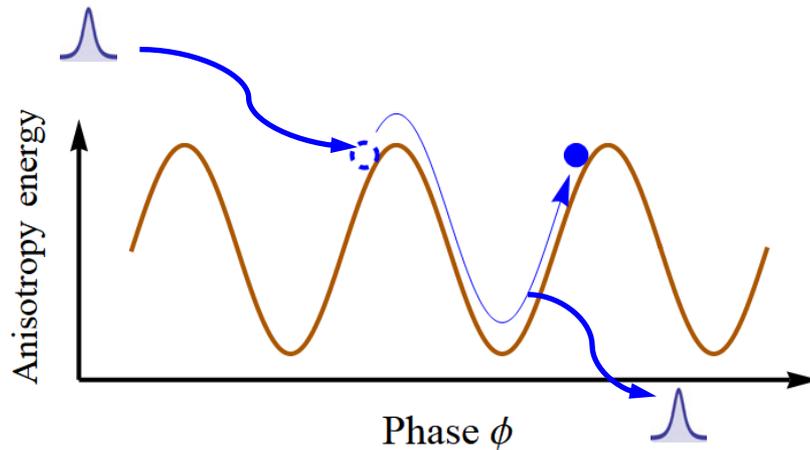
**Super-critical regime: Driven dynamics**  
(above a threshold current)

$$j_1^{\text{th}} = \frac{\omega_e}{2\sigma}$$

$$\dot{\phi}(t) \approx \frac{\omega_{\text{gen}}}{2} + \frac{\omega_e \omega_{\text{ex}}}{4\sqrt{\alpha^2 \omega_{\text{ex}}^2 + \omega_{\text{gen}}^2}} \cos \omega_{\text{gen}} t$$



**Sub-critical regime: Inertial dynamics**  
(below a threshold current)



### What AFM materials have to offer in spintronics?

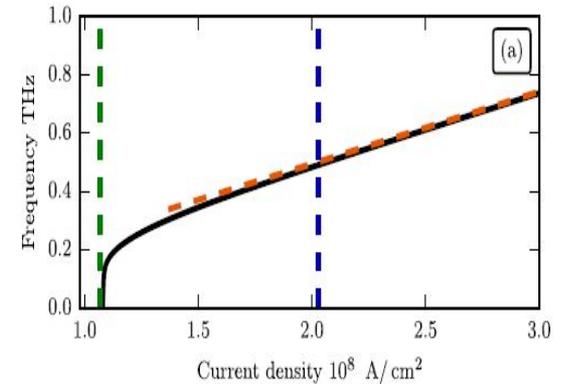
## TUNABLE AFM OSCILLATOR

Cheng et al. PRL 2016 / Khymyn et al. Sci. Rep. (2017)

**Super-critical regime: Driven dynamics**  
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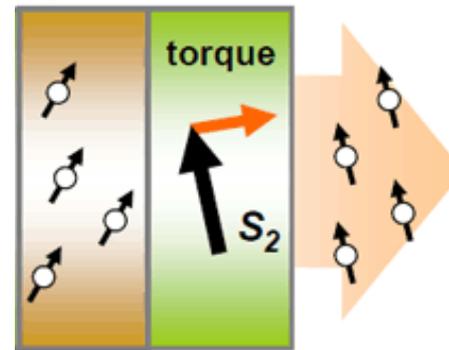
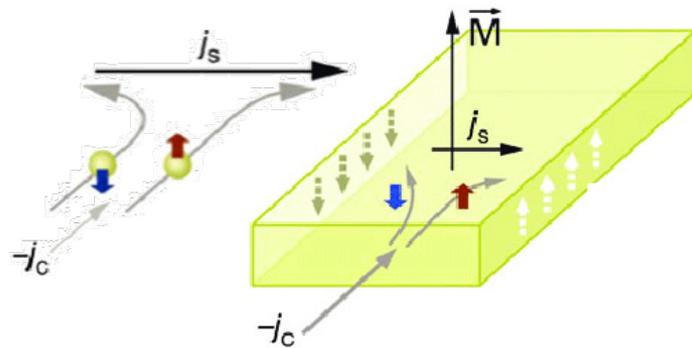


## CONDITIONS

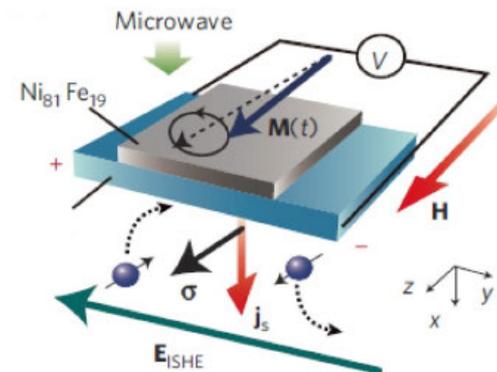
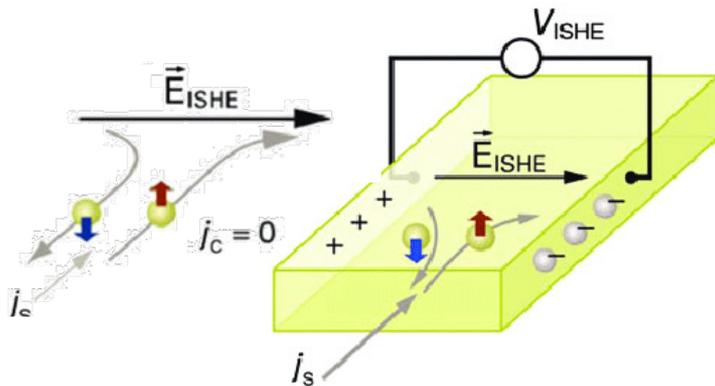
- Single domain AFM
- Very specific magnetic anisotropy and AFM/NM device configuration
- Low anisotropy
- **Efficient spin injection – Spin transfer torque**  
(Threshold charge current in Pt/NiO > 10<sup>8</sup> A/cm<sup>2</sup>)

### Spin-Charge interconversion with AFMs

Spin Hall Effect (SHE) + Spin Transfer Torque (STT)



Inverse Spin Hall Effect (ISHE) + Dynamical Spin Pumping (DSP)



### Magnetization dynamics in a uniaxial antiferromagnet (e.g., MnF<sub>2</sub>)

Free energy (macrospin)

$$F = \underbrace{\hbar\omega_E \mathbf{m}_1 \cdot \mathbf{m}_2}_{\text{exchange}} - \underbrace{\hbar\omega_H \mathbf{e}_z \cdot (\mathbf{m}_1 + \mathbf{m}_2)}_{\text{magnetic field}} - \underbrace{\frac{1}{2} \hbar\omega_A \left[ (\mathbf{m}_1 \cdot \mathbf{e}_z)^2 + (\mathbf{m}_2 \cdot \mathbf{e}_z)^2 \right]}_{\text{anisotropy}}$$

Equations of motion

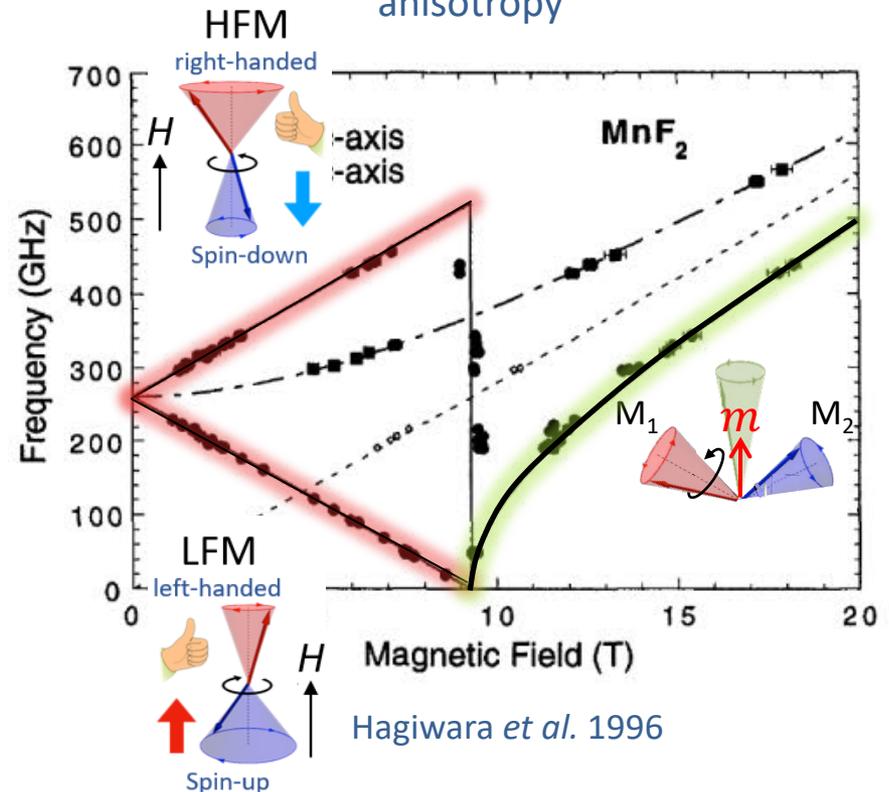
$$\dot{\mathbf{m}}_1 = \mathbf{m}_1 \times \left[ -\omega_E \mathbf{m}_2 + \omega_H \mathbf{e}_z - \omega_A \mathbf{e}_z (\mathbf{m}_1 \cdot \mathbf{e}_z) \right]$$

$$\dot{\mathbf{m}}_2 = \mathbf{m}_2 \times \left[ -\omega_E \mathbf{m}_1 + \omega_H \mathbf{e}_z - \omega_A \mathbf{e}_z (\mathbf{m}_2 \cdot \mathbf{e}_z) \right]$$

Resonance frequency

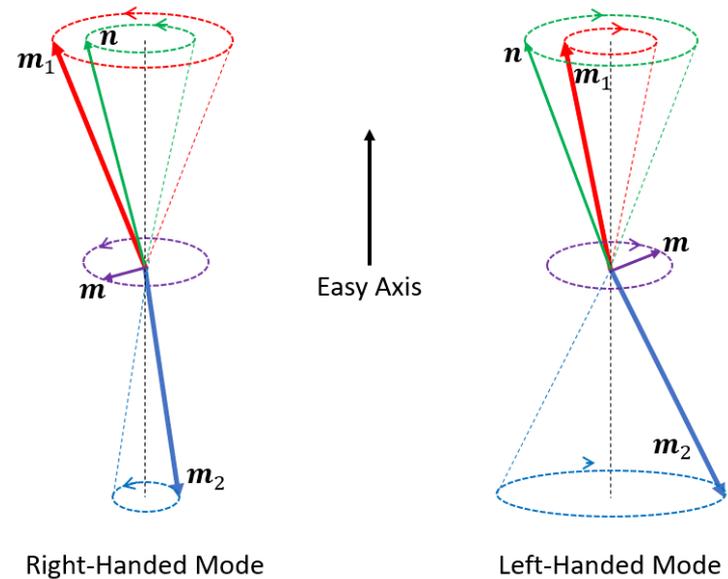
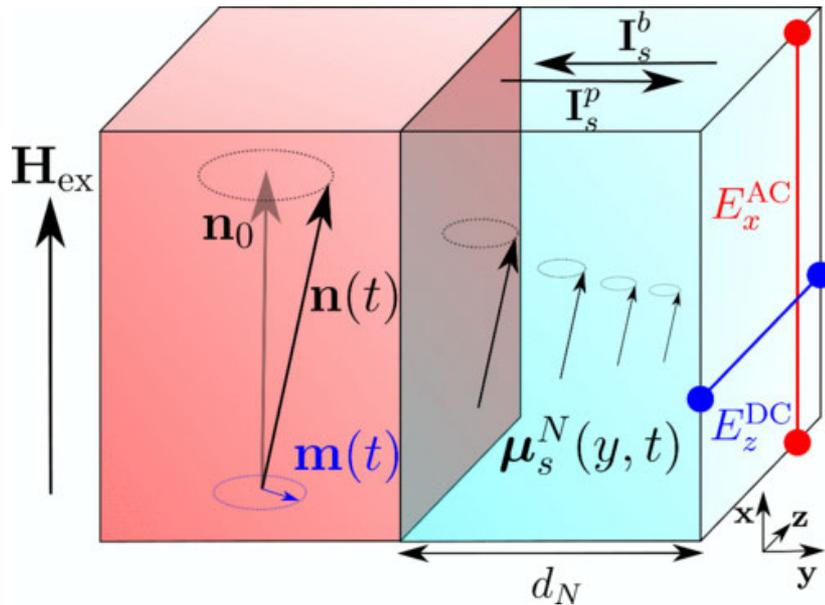
$$\omega_{res} = \gamma\mu_0 \sqrt{H_A(2H_E + H_A)} \pm \gamma\mu_0 H$$

$$\omega_{res} = \gamma\mu_0 \sqrt{H^2 - 2H_E H_A}$$



### Spin pumping at AFM/NM interfaces

Cheng/Braatas PRL 113, 057601 (2014)



$$I_s \propto \mathbf{m}_1 \times \dot{\mathbf{m}}_1 + \mathbf{m}_2 \times \dot{\mathbf{m}}_2$$

$$\mathbf{m}_1 = -\mathbf{m}_2$$

$$\dot{\mathbf{m}}_1 \sim -\dot{\mathbf{m}}_2$$

$$\mathbf{n} = \frac{(\mathbf{m}_1 - \mathbf{m}_2)}{2}$$

Staggered Magnetization

$$I_s \propto \mathbf{n} \times \dot{\mathbf{n}}$$

$$\mathbf{m} = \frac{(\mathbf{m}_1 + \mathbf{m}_2)}{2}$$

Net Magnetization

$$I_s \propto \mathbf{m} \times \dot{\mathbf{m}}$$

### Spin pumping at AFM/NM interfaces

Cheng/Braatas PRL 113, 057601 (2014)

$$F = \hbar\omega_E \mathbf{m}_1 \cdot \mathbf{m}_2 - \hbar\omega_H \mathbf{e}_z \cdot (\mathbf{m}_1 + \mathbf{m}_2) - \frac{1}{2} \hbar\omega_A \left[ (\mathbf{m}_1 \cdot \mathbf{e}_z)^2 + (\mathbf{m}_2 \cdot \mathbf{e}_z)^2 \right] + \underbrace{\delta F}_{\text{a.c. field}}$$

$$\delta F = (\mathbf{m}_1 + \mathbf{m}_2) \cdot \left[ \omega_{Hx} \mathbf{e}_x \cos(\omega t + \theta_x) + \omega_{Hy} \mathbf{e}_y \cos(\omega t + \theta_y) \right]$$

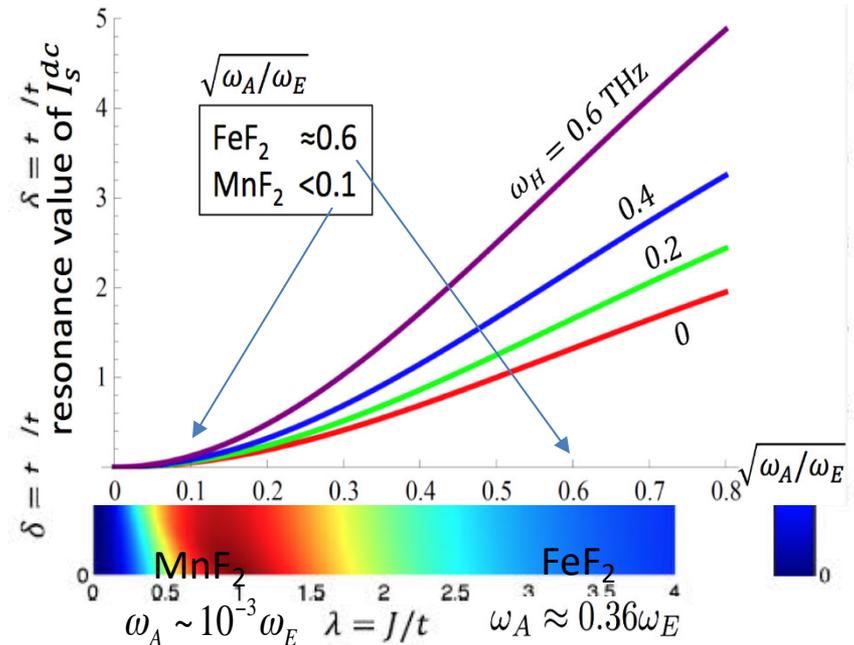
$$\frac{e}{\hbar} \mathbf{I}_s = G_r (\mathbf{n} \times \dot{\mathbf{n}} + \mathbf{m} \times \dot{\mathbf{m}}) - G_i \dot{\mathbf{m}}$$

Spin pumping (at resonance  $\omega = \omega_R$ ,  $\omega_H = 0$ )

$$\mathbf{m}_1 \times \dot{\mathbf{m}}_1 + \mathbf{m}_2 \times \dot{\mathbf{m}}_2 = \sqrt{\frac{\omega_A}{\omega_E}} \frac{1}{\alpha^2} \frac{\omega_{Hx} \omega_{Hy}}{\omega_E} \sin(\theta_x - \theta_y)$$

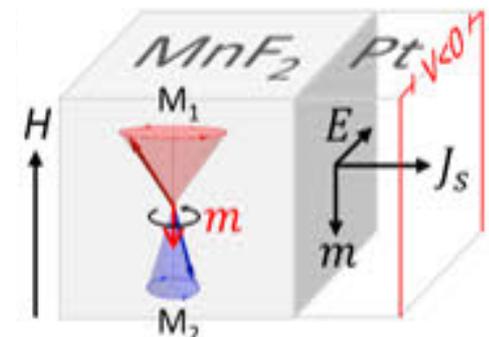
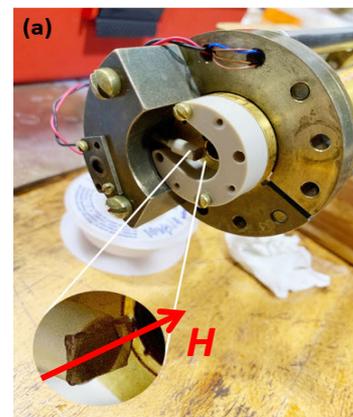
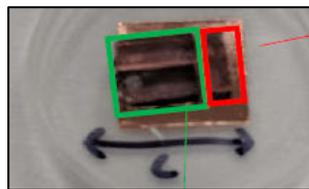
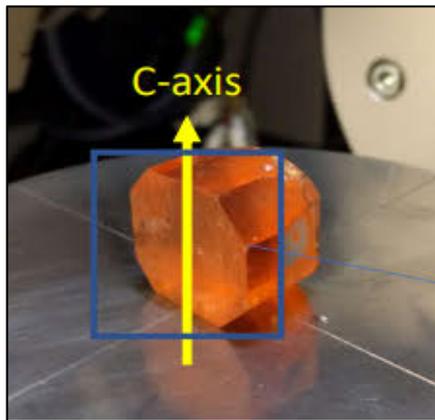
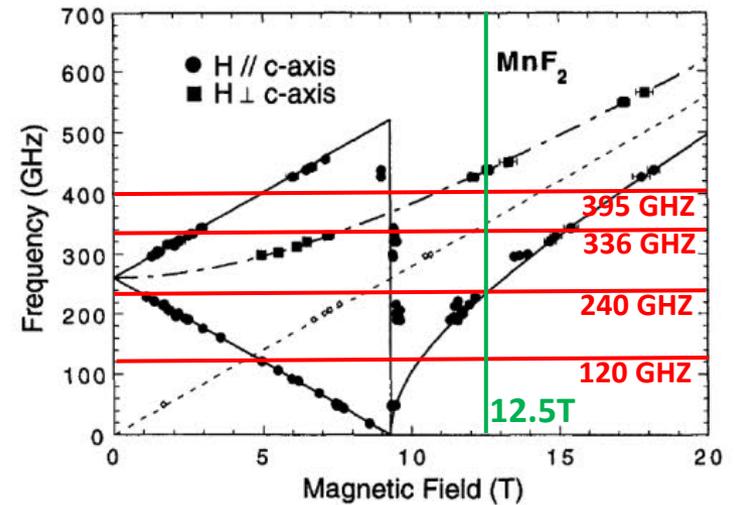
**anisotropy  
enhancement**

**polarization  
dependence  
(no SP with linear)**



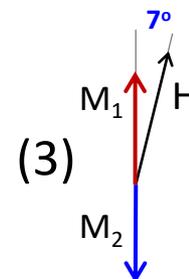
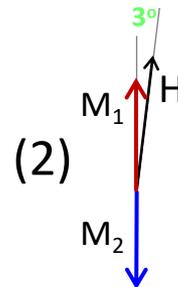
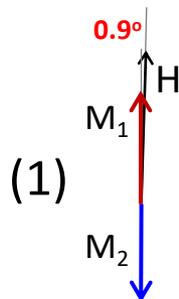
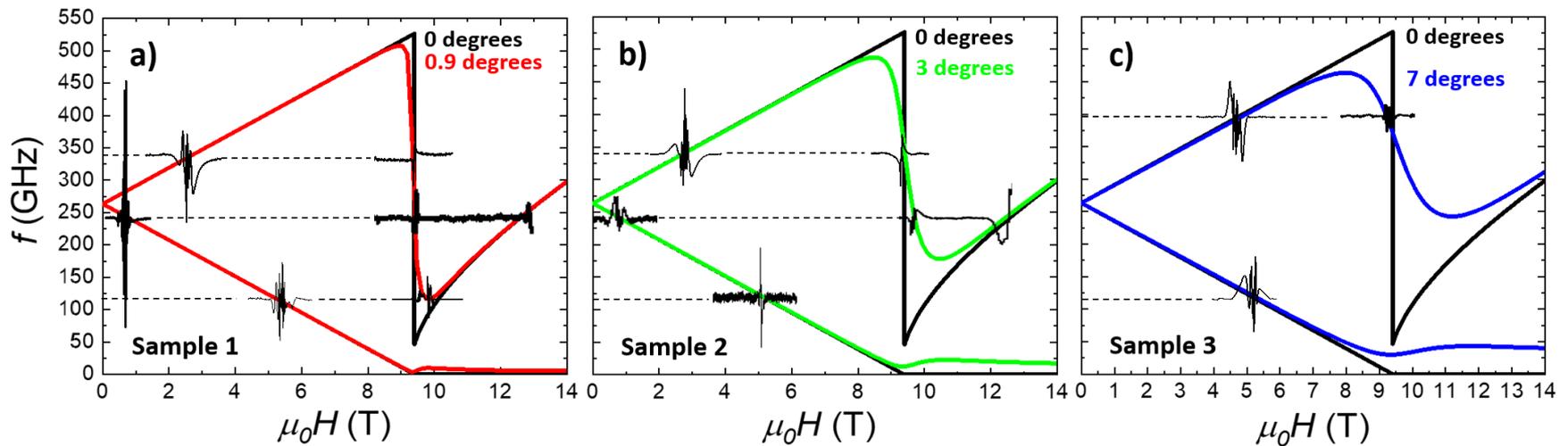
### Spin pumping at $MnF_2/Pt$ interfaces

UCF/UCSC/UCR/NTNU/NHMFL results -  $MnF_2(300\mu m+10nm)/Pt(4nm)$

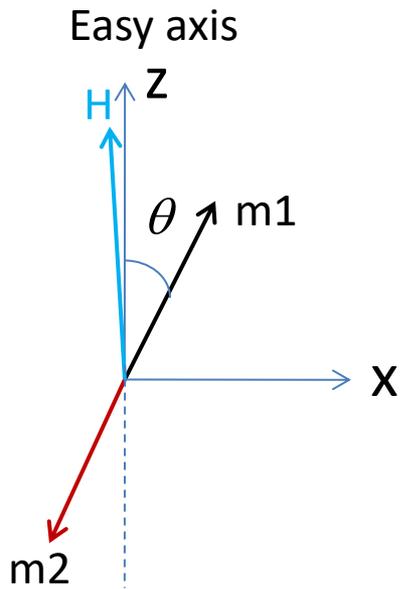


### Spin pumping at $\text{MnF}_2/\text{Pt}$ interfaces

### EPR SPECTROSCOPY



### Spin pumping at MnF<sub>2</sub>/Pt interfaces



$$\frac{d\mathbf{M}_i}{dt} = -\gamma \left[ \mathbf{M}_i \times \mu_0 \mathbf{H}_{\text{eff},i} \right] - \alpha \frac{\gamma}{M} \left[ \mathbf{M}_i \times \left[ \mathbf{M}_i \times \mu_0 \mathbf{H}_{\text{eff},i} \right] \right]$$

$$H_{\text{eff}} = H_{\text{ex}} + H_k + H + H_p$$

### Spin pumping at MnF<sub>2</sub>/Pt interfaces

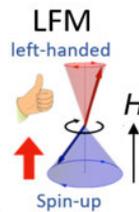
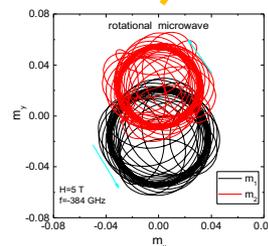
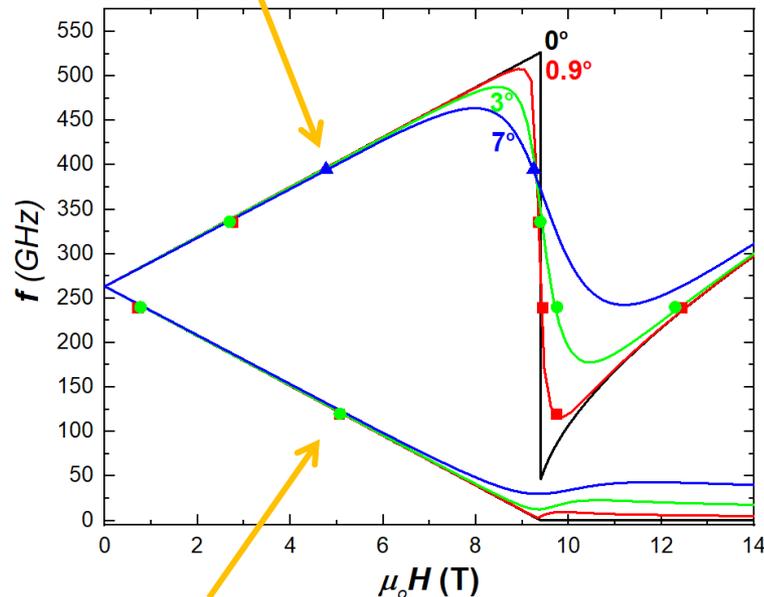
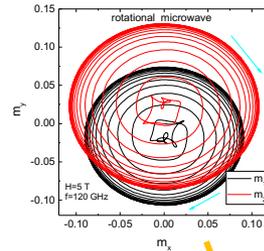
$$M_S = 0.06T$$

$$H_E = 47.05T$$

$$H_A = 0.8T$$

$$\gamma = \gamma_e$$

$$\alpha = 0.001$$



# Spin Pumping from AFM Insulator MnF<sub>2</sub>

## Terahertz Spintronics with Antiferromagnetic Insulators

### Spin pumping at MnF<sub>2</sub>/Pt interface

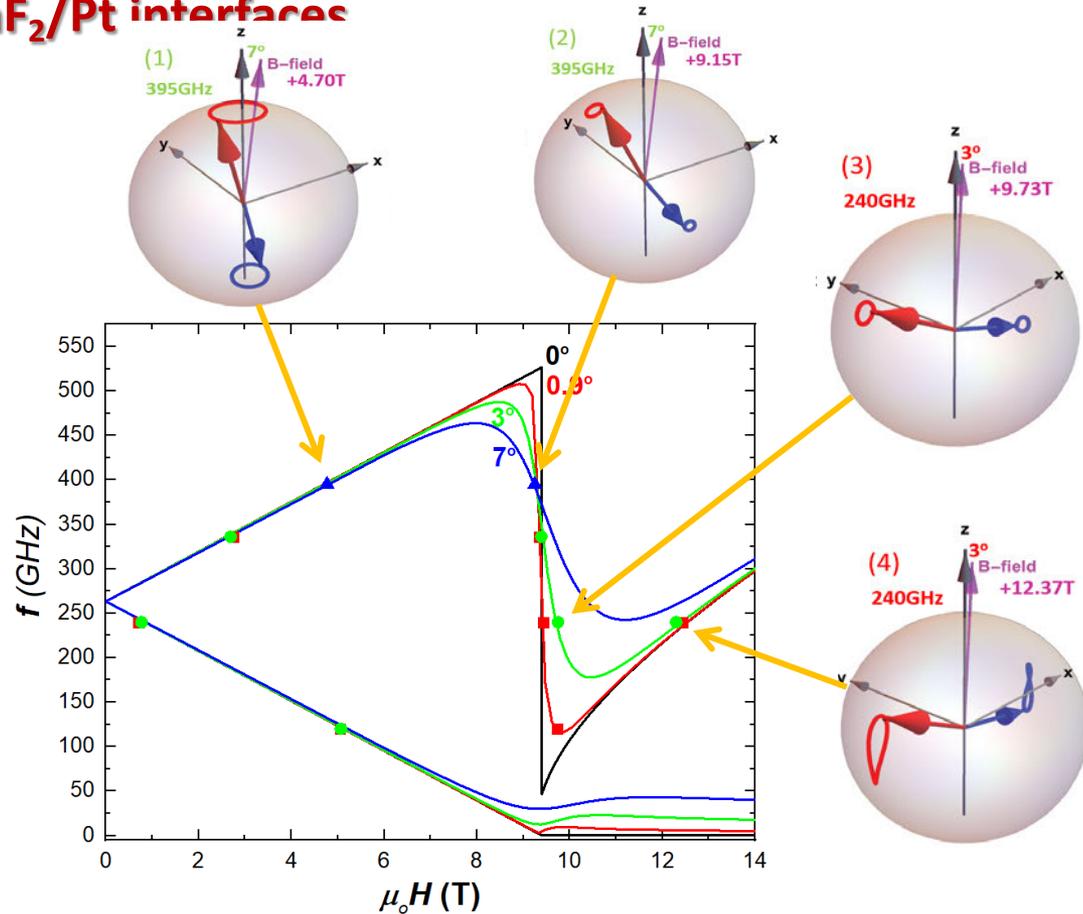
$$M_S = 0.06\text{T}$$

$$H_E = 47.05\text{T}$$

$$H_A = 0.8\text{T}$$

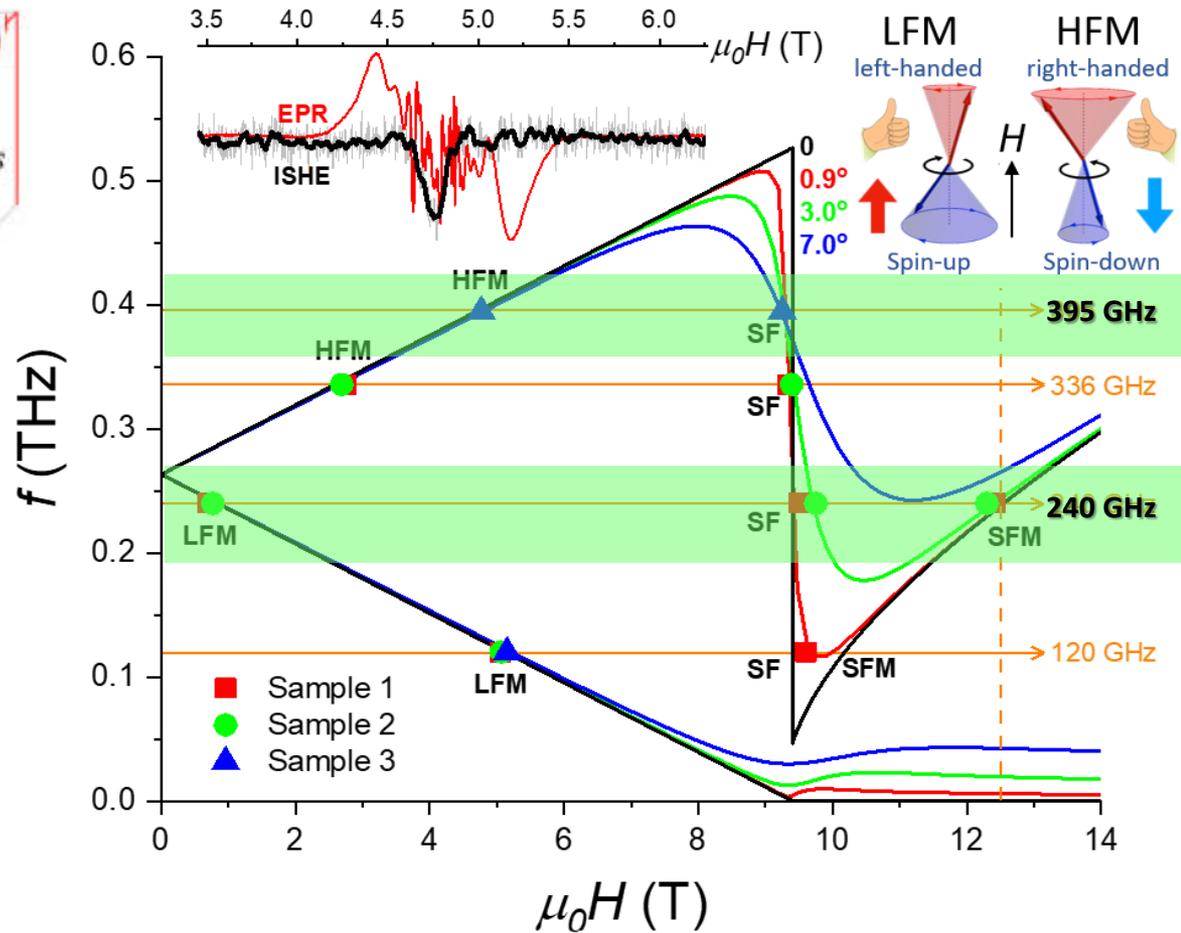
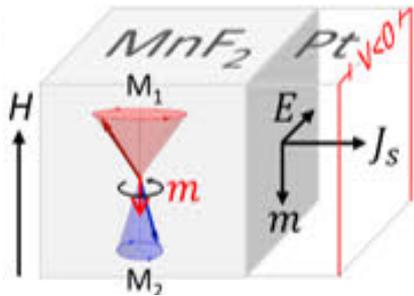
$$\gamma = \gamma_e$$

$$\alpha = 0.001$$



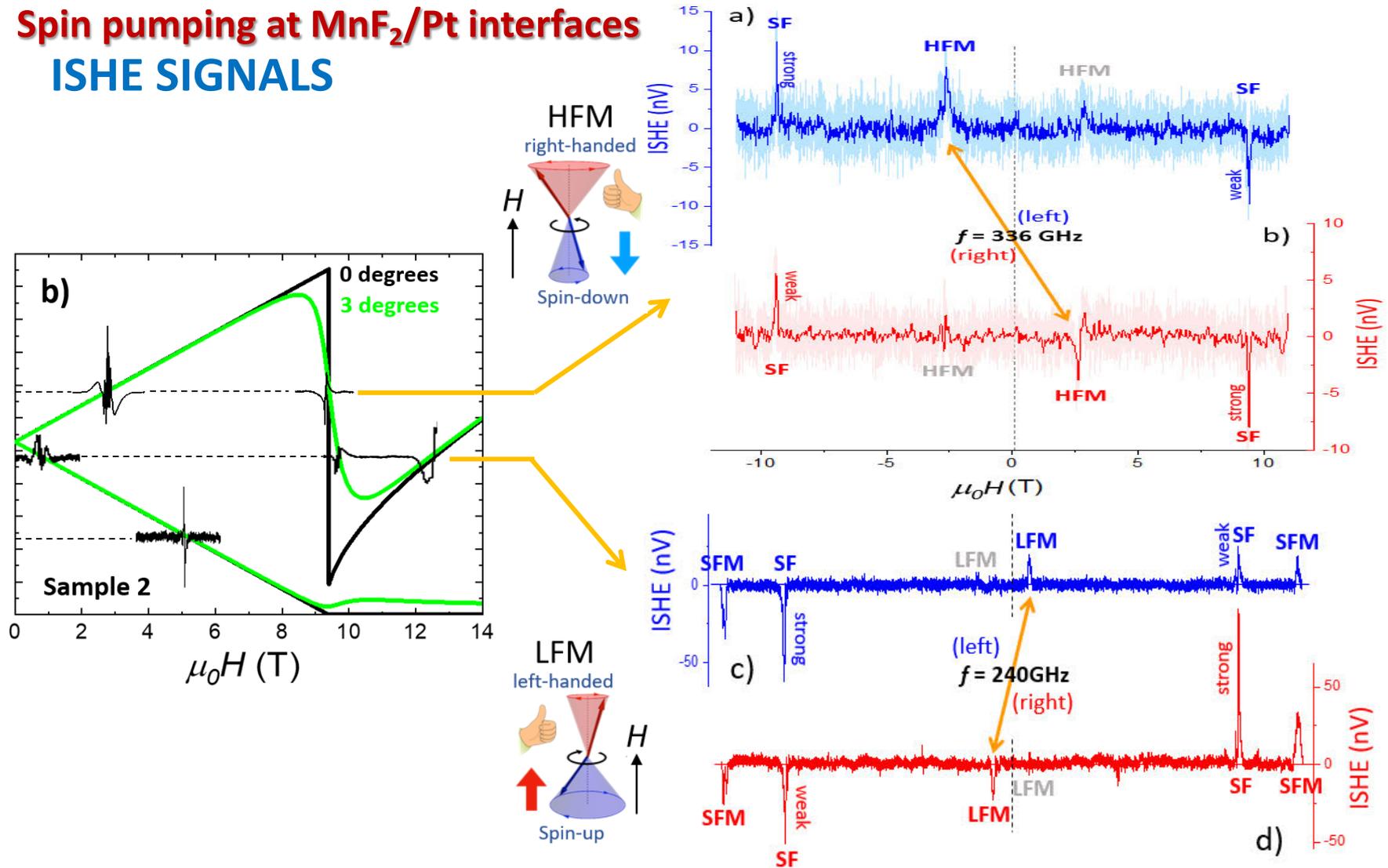
### Spin pumping at $MnF_2/Pt$ interfaces

### ISHE SIGNALS



### Spin pumping at $MnF_2/Pt$ interfaces

### ISHE SIGNALS



# Spin Pumping from AFM Insulator $\text{MnF}_2$

## Terahertz Spintronics with Antiferromagnetic Insulators

**Spin pumping at  $\text{MnF}_2/\text{Pt}$  interfaces**  
**ISHE SIGNALS**

# Spin Pumping from AFM Insulator $\text{MnF}_2$

## Terahertz Spintronics with Antiferromagnetic Insulators

**Spin pumping at  $\text{MnF}_2/\text{Pt}$  interfaces**  
**ISHE SIGNALS**

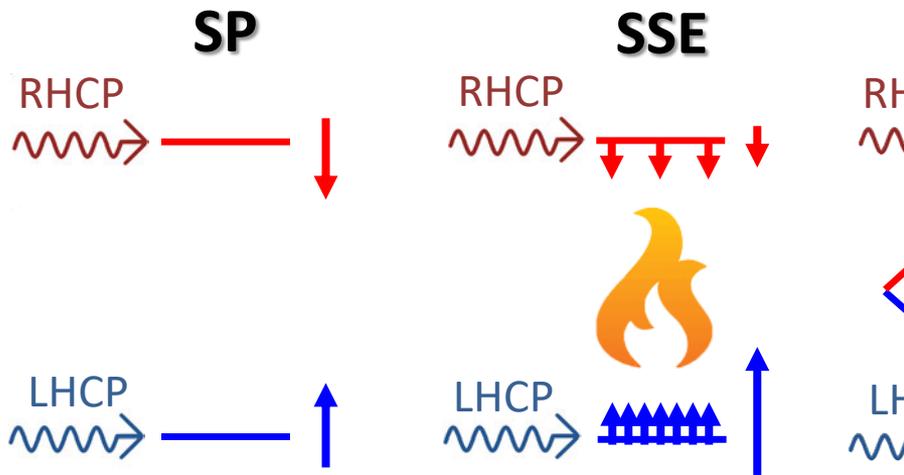
# Spin Pumping from AFM Insulator $\text{MnF}_2$

## Terahertz Spintronics with Antiferromagnetic Insulators

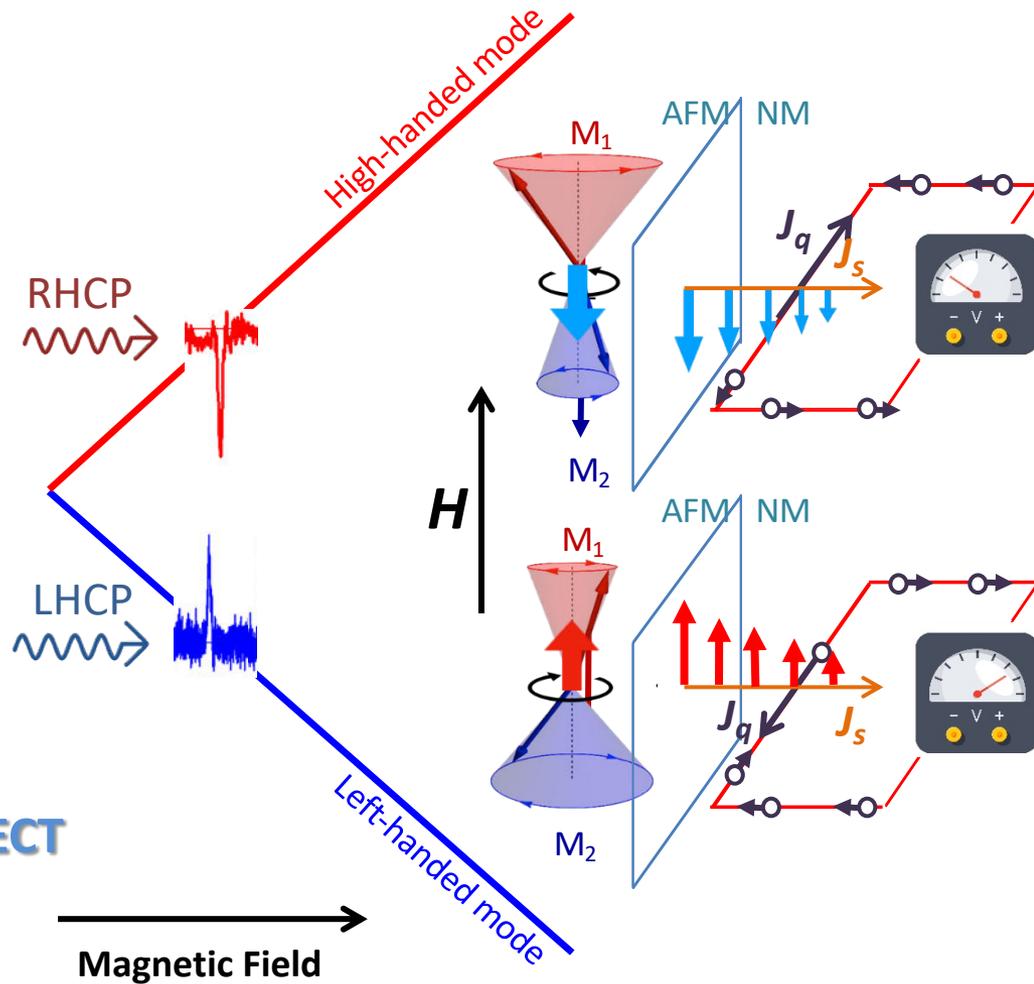
**Spin pumping at  $\text{MnF}_2/\text{Pt}$  interfaces**  
**ISHE SIGNALS**

### Spin pumping at $\text{MnF}_2/\text{Pt}$ interfaces

### ISHE SIGNALS



**PROOF of COHERENT SPIN PUMPING**  
**NOT INCOHERENT SPIN SEEBECK EFFECT**



### Spin pumping at MnF<sub>2</sub>/Pt interfaces

### ISHE SIGNALS

## SPIN-MIXING CONDUCTANCE

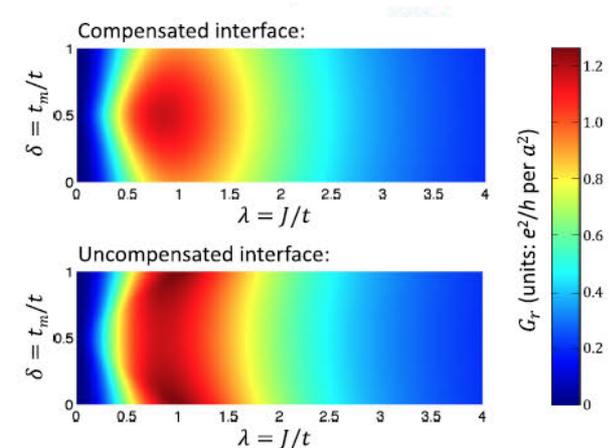
$$V_{ISHE} = L\theta_S \left(\frac{H_A}{H_E}\right) \left(\frac{\lambda}{d_N}\right) \frac{\hbar e(\gamma B_{\perp})^2}{\alpha^2 \omega_R} \frac{g_r \tanh \frac{d_N}{2\lambda}}{h\sigma + 2\lambda e^2 g_r \coth \frac{d_N}{2\lambda}}$$

$$\begin{aligned} \frac{H_A}{H_E} &\approx 1.8\% \\ \alpha &\approx 0.5 \times 10^{-3} \\ d_N &\approx 4 \text{ nm} \\ \theta_S &\approx 0.04 \\ \lambda &\approx 4 \text{ nm} \\ \sigma &\approx 2 \times 10^6 \text{ S/m} \\ B_{\perp} &\approx 200 \text{ mG} \\ V_{ISHE} &\approx 25 \text{ nV} \end{aligned}$$

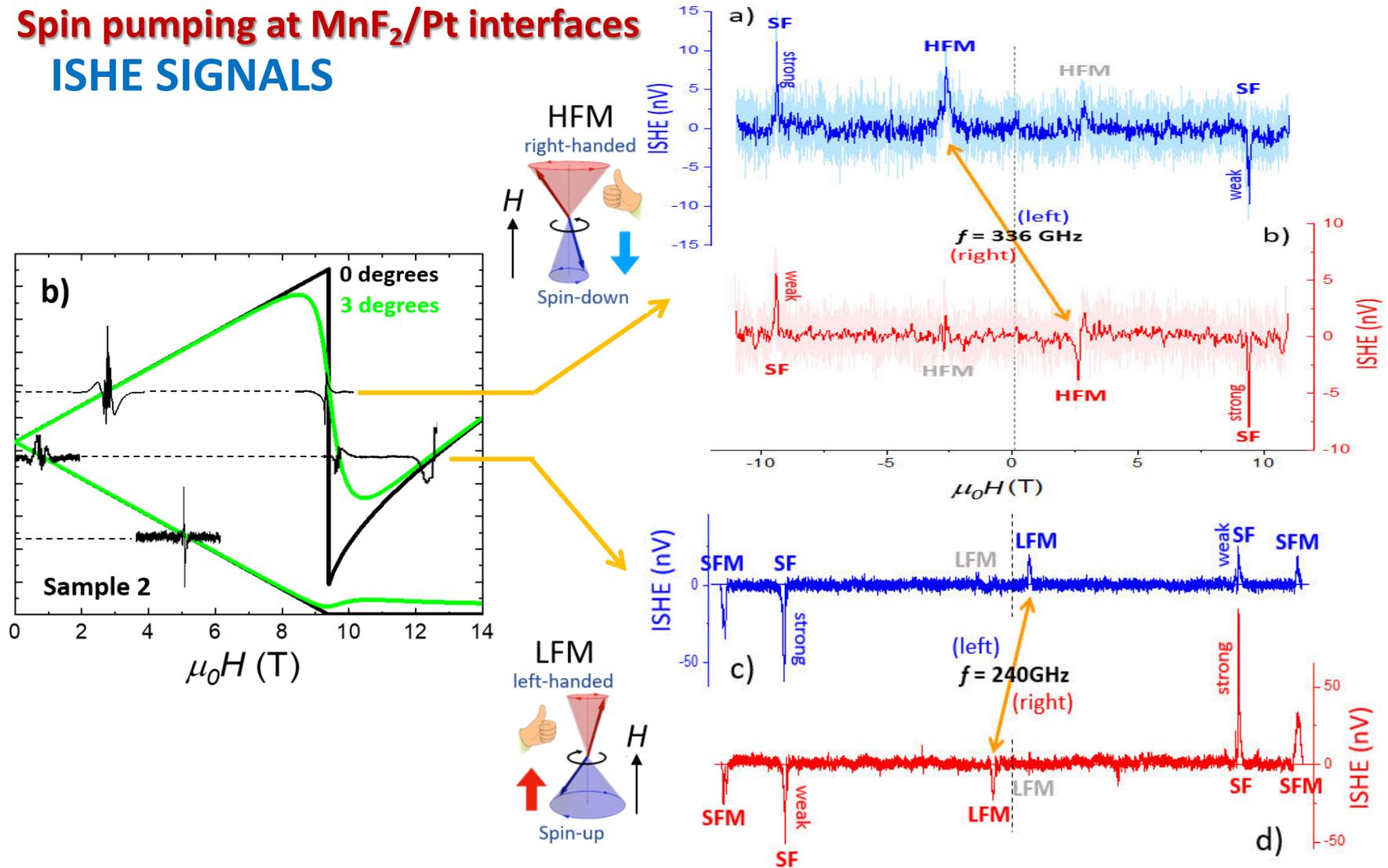
$$g_r \approx 4.3 \times 10^{18} \text{ m}^{-2}$$

$$\approx 1 e^2/h \text{ per unit cell area}$$

Similar than in FM/Pt interfaces!

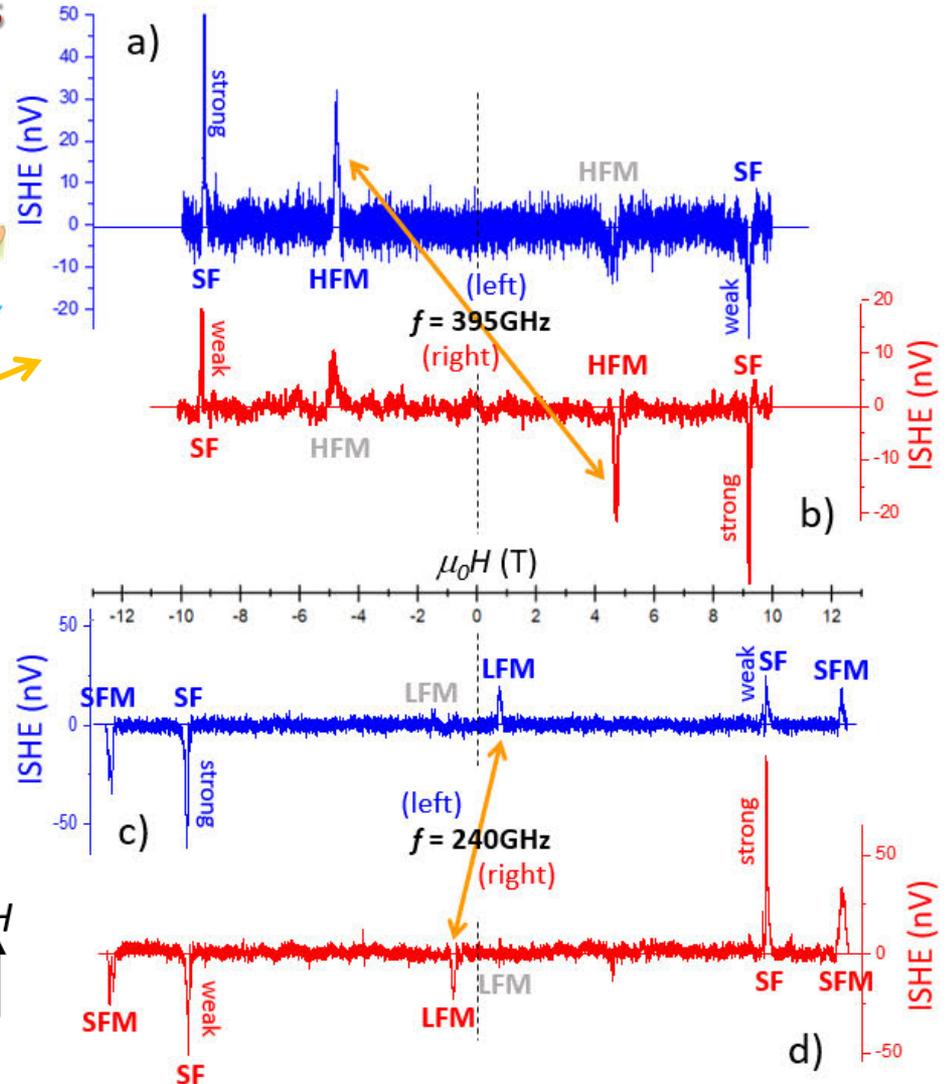
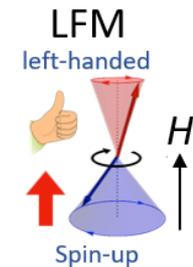
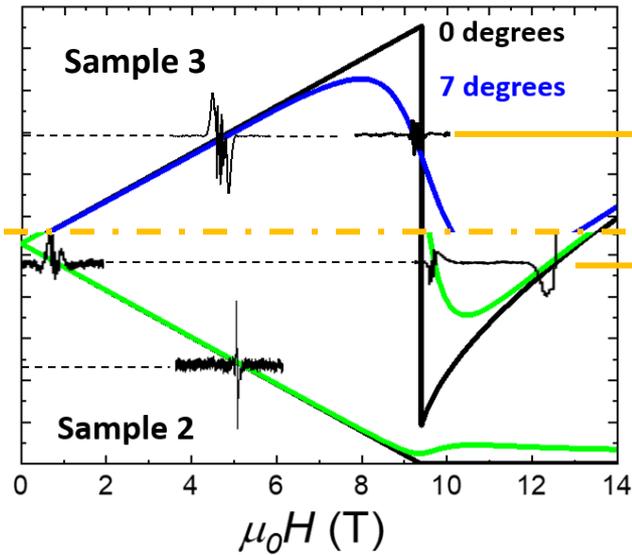


### Spin pumping at $MnF_2/Pt$ interfaces ISHE SIGNALS

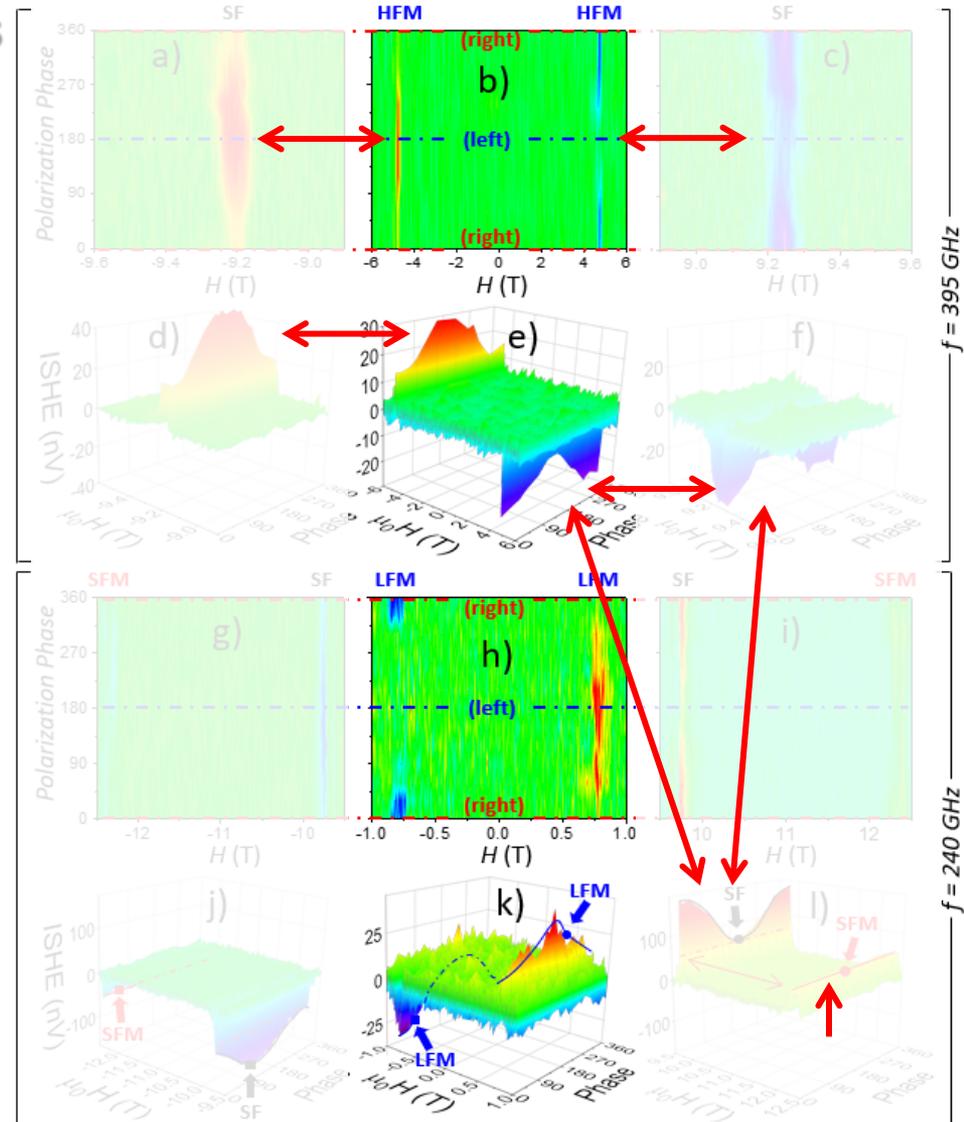
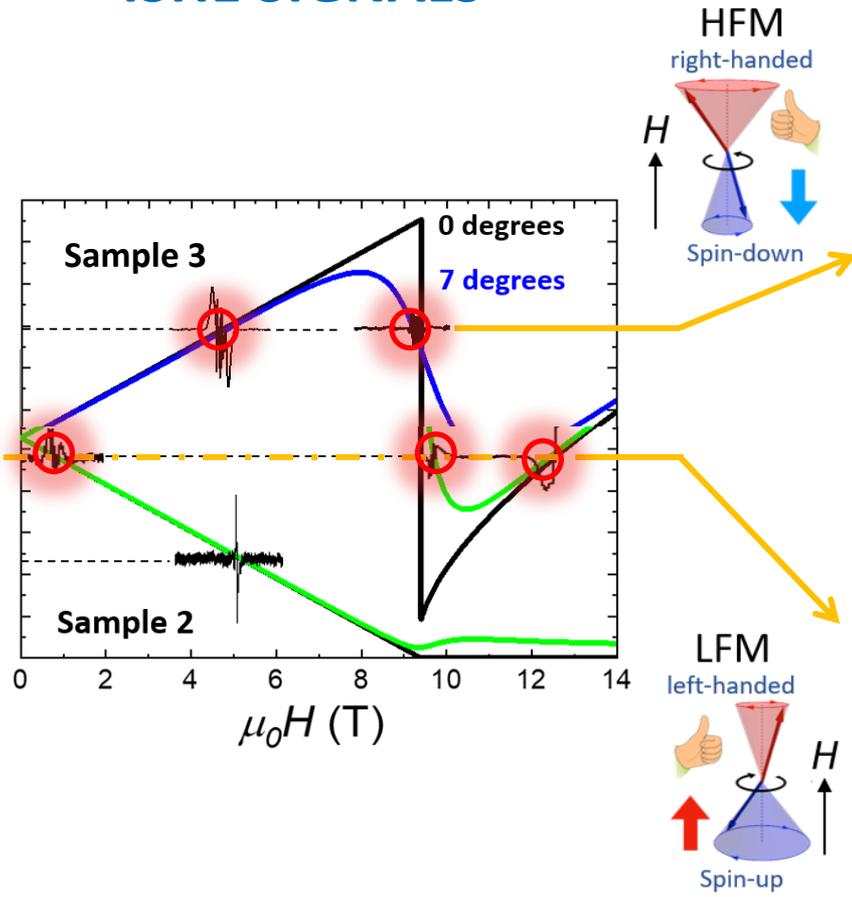


### Spin pumping at $MnF_2/Pt$ interfaces

### ISHE SIGNALS

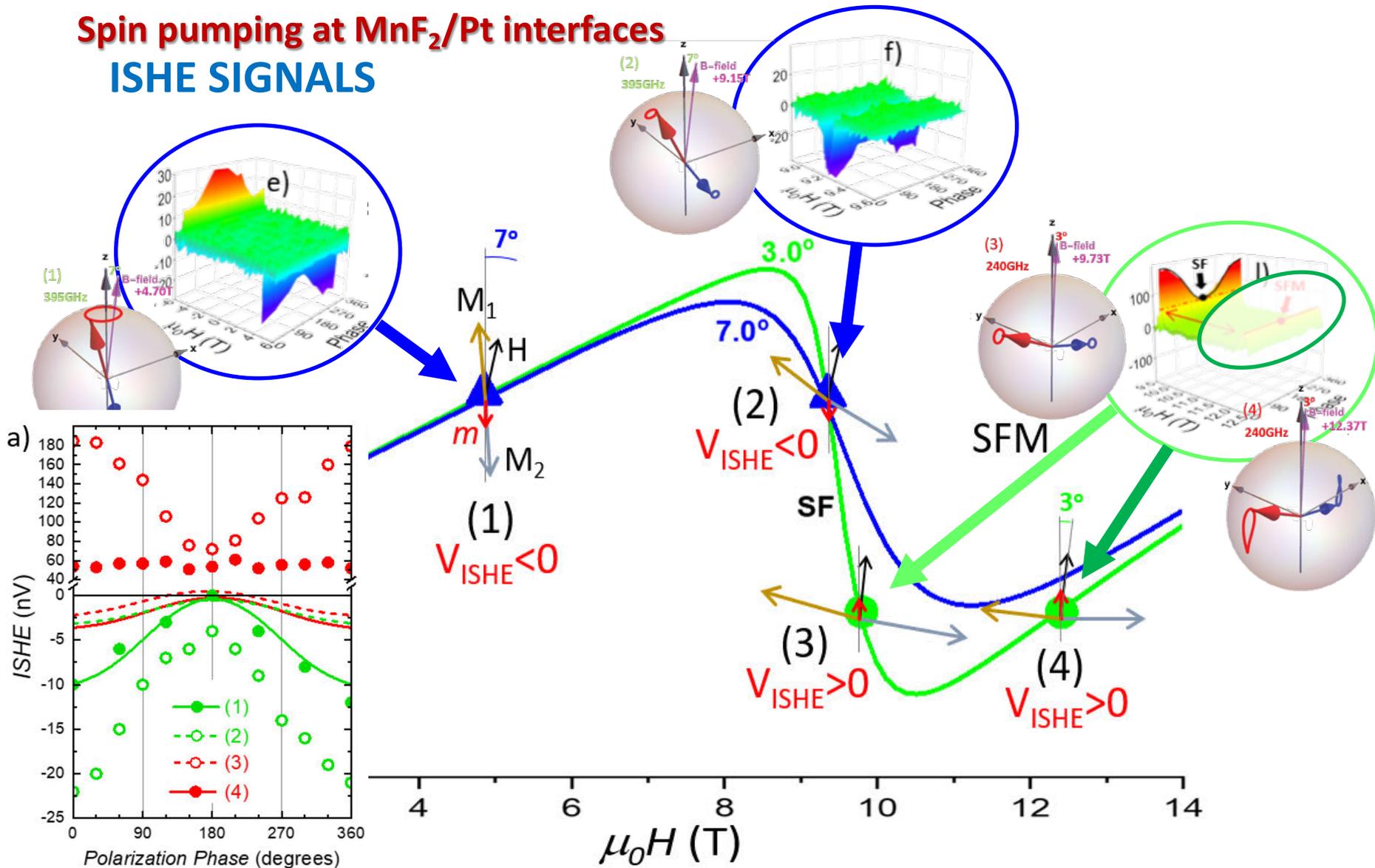


### Spin pumping at $MnF_2/Pt$ interfaces ISHE SIGNALS



### Spin pumping at $\text{MnF}_2/\text{Pt}$ interfaces

### ISHE SIGNALS



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Priyanka Vaidya



Gyan Khatri



Greg Fritjofson

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