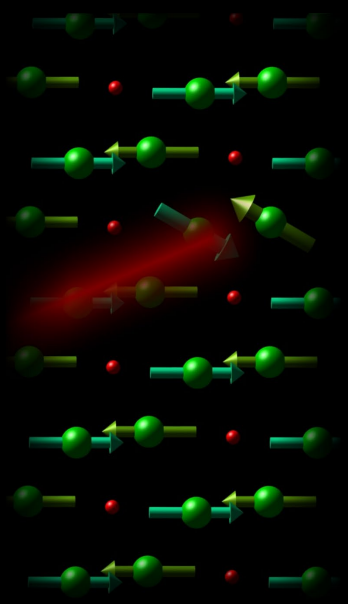

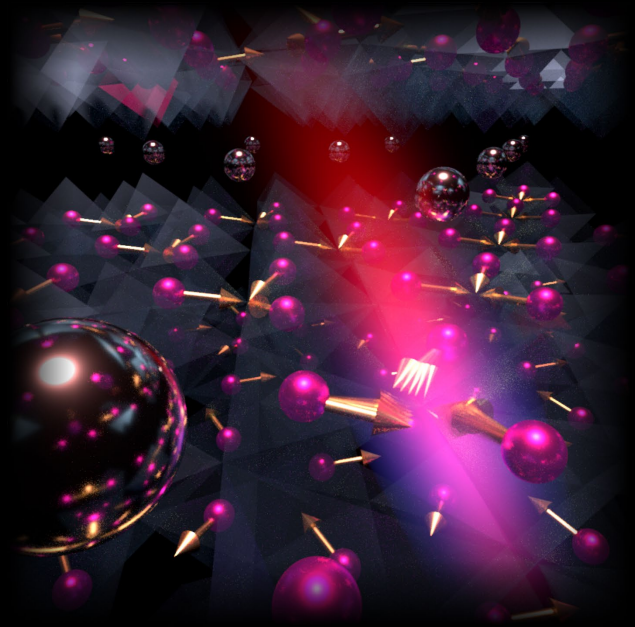

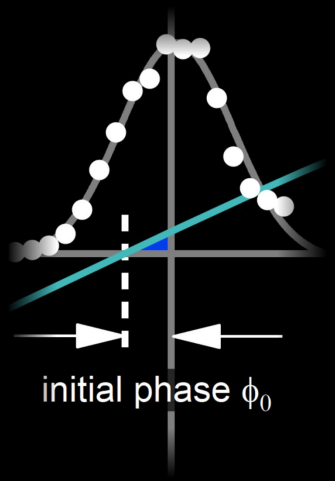

Ultrafast optical excitation and probing of coherent antiferromagnetic spin dynamics

CHRISTIAN TZSCHASCHEL, HARVARD UNIVERSITY

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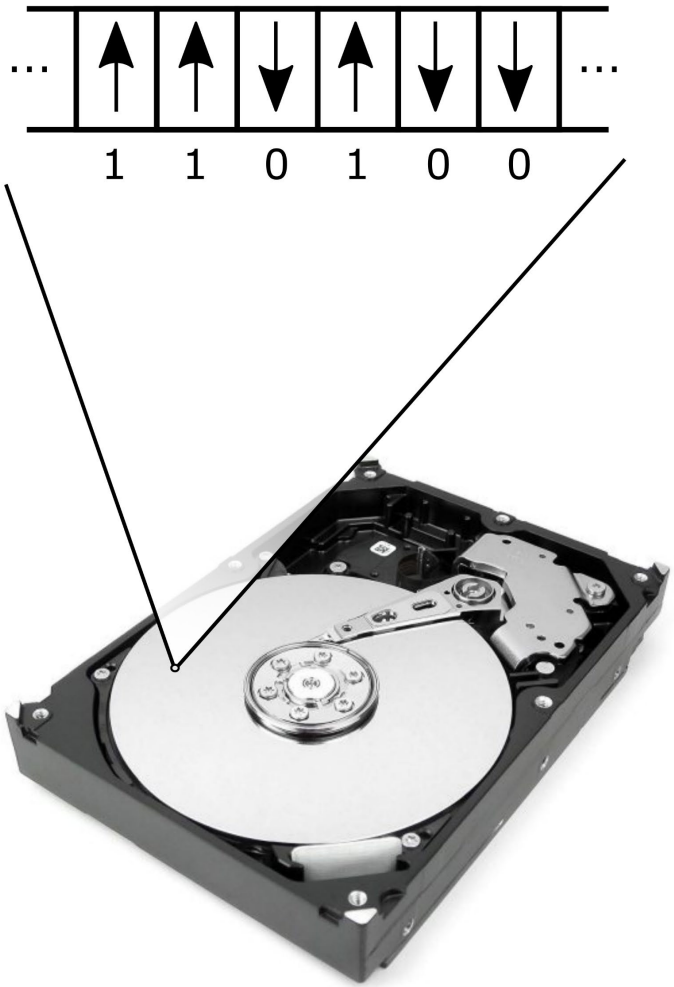
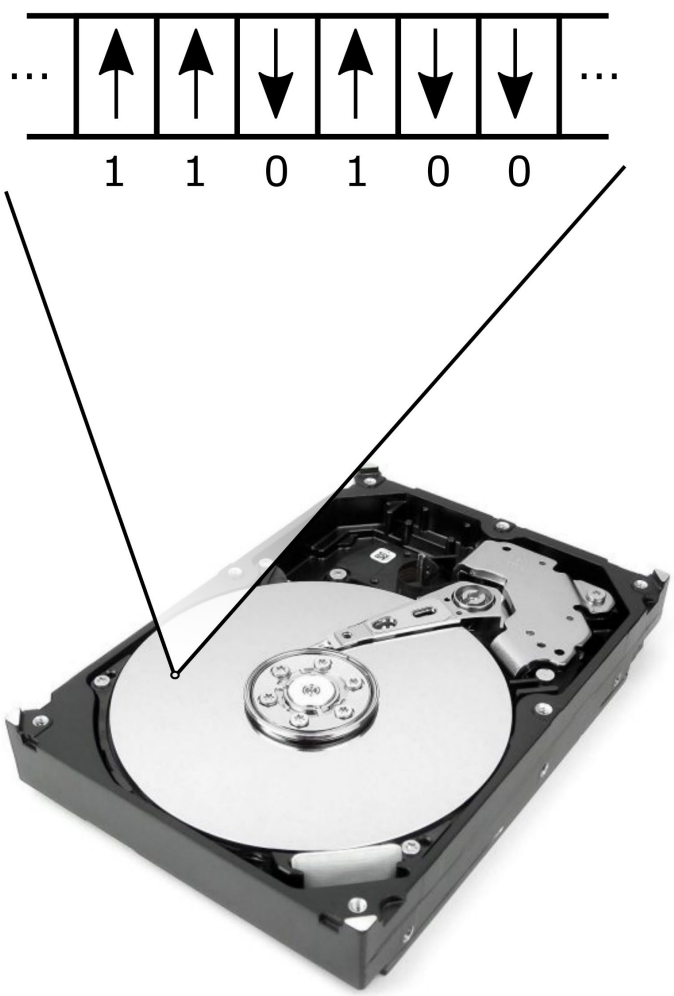
HARVARD UNIVERSITY
Office of the Vice Provost for Research
Climate Change Solutions Fund

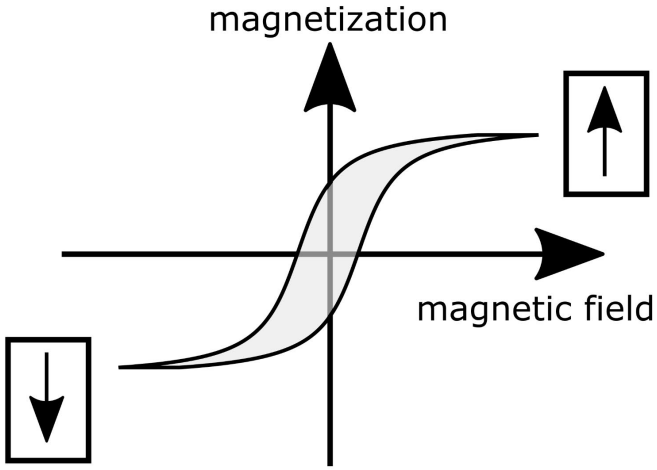
Information Technology

Information Technology



Information Technology





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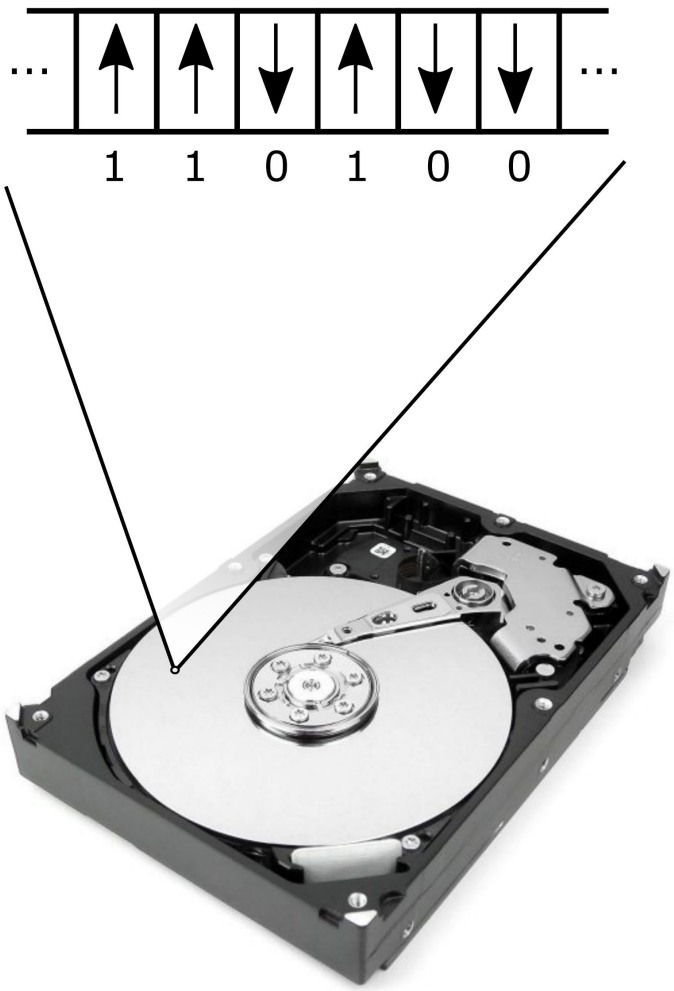
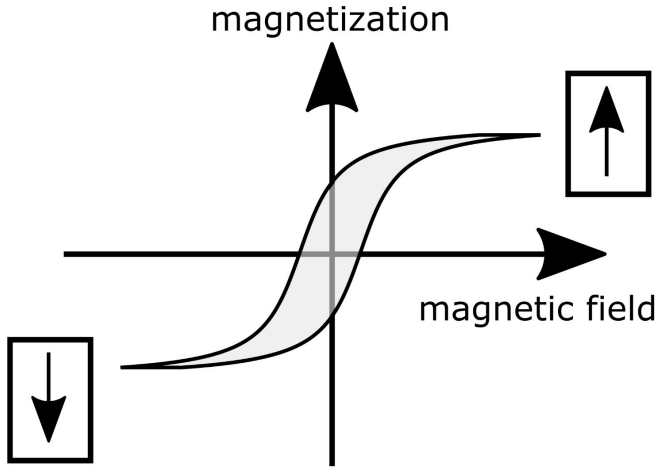
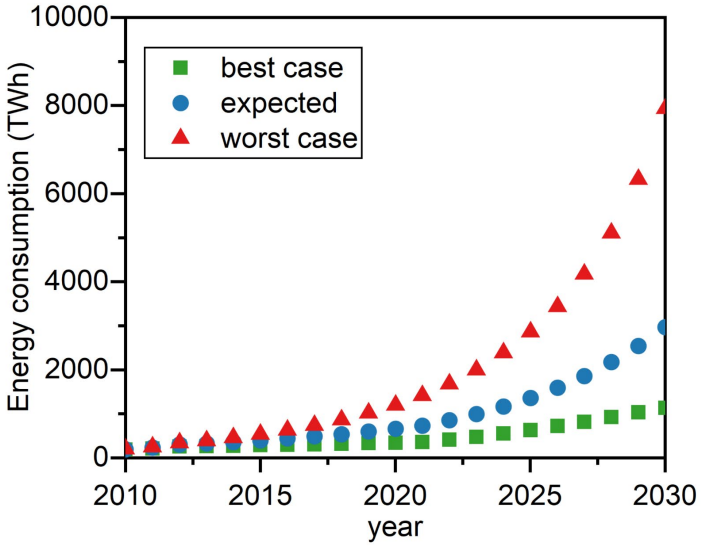
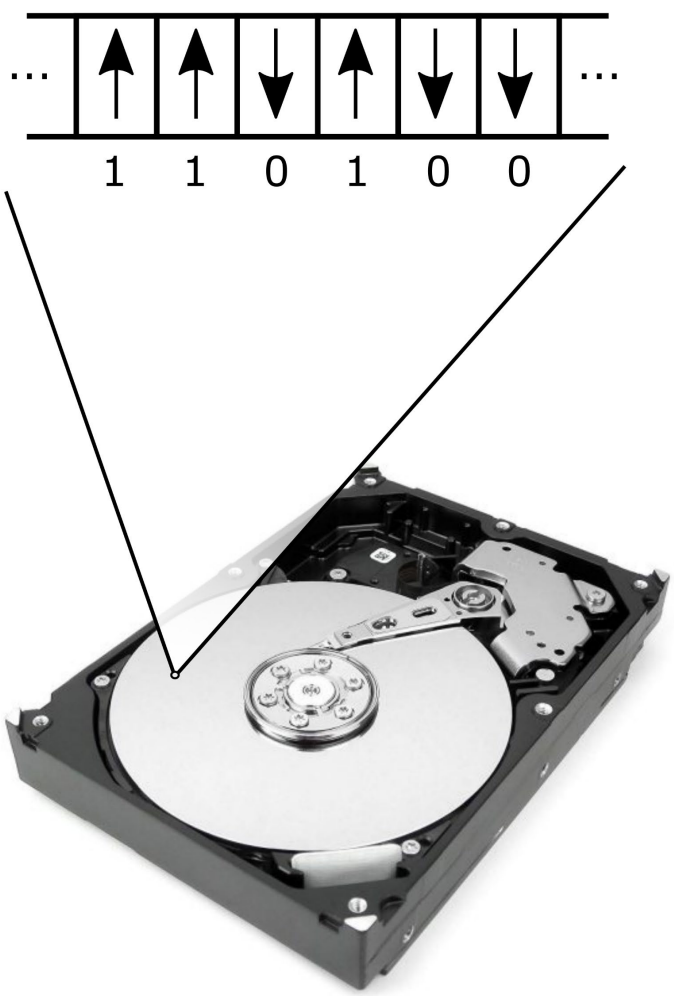


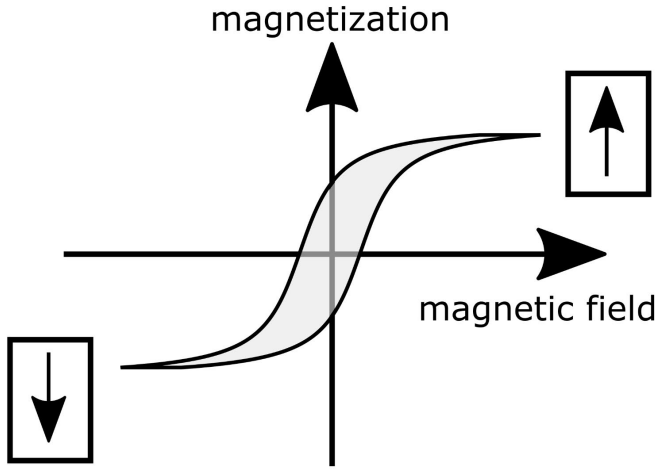


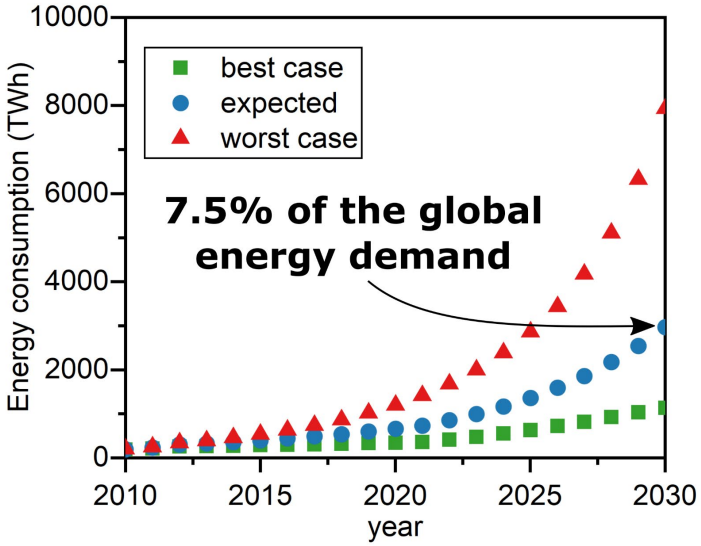


Andrae et al., Challenges **6,** 117 (2015)

Information Technology



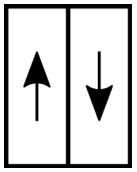




Andrae et al., Challenges **6,** 117 (2015)

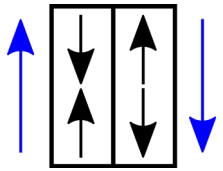
Antiferromagnets

Ferromagnetic materials



Magnetization **M**

Antiferromagnetic materials



OP **L** = $\mathbf{M}_1 - \mathbf{M}_2$

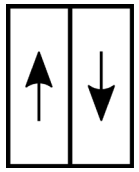
"Antiferromagnets are extremely interesting from the theoretical viewpoint, but do not seem to have any application" – Louis Néel, 1970

- Robust
- Abundant
- Versatile
- Fast

Antiferromagnets envisaged for novel logic devices

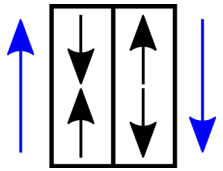
Coherent dynamics

Ferromagnetic materials



Magnetization **M**

Antiferromagnetic materials



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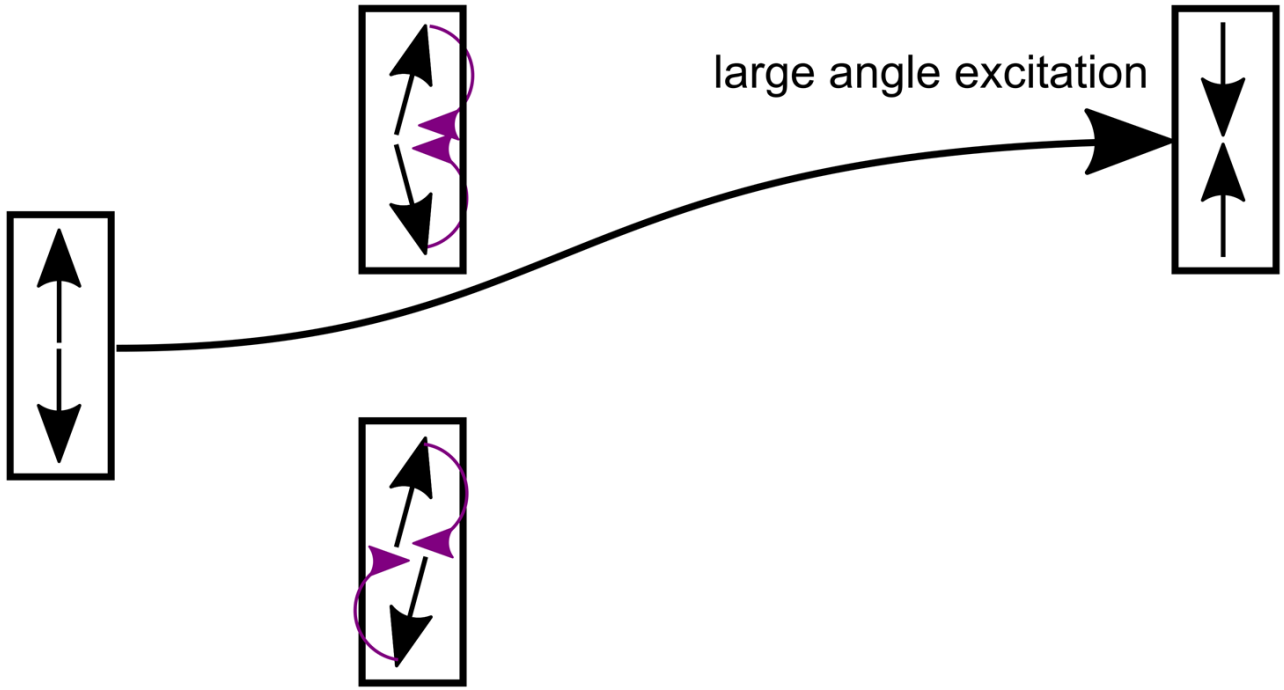
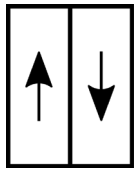


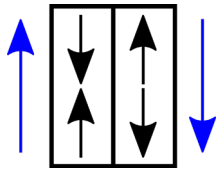
Coherent dynamics

Ferromagnetic materials



Magnetization **M**

Antiferromagnetic materials



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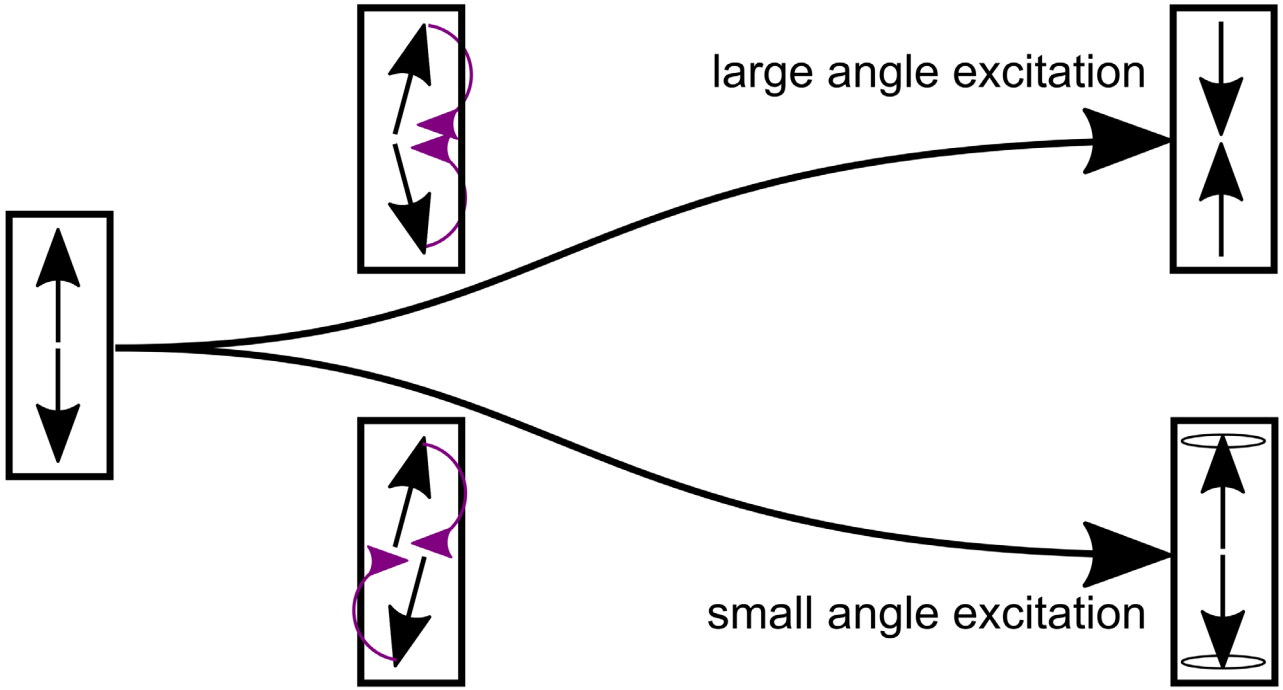
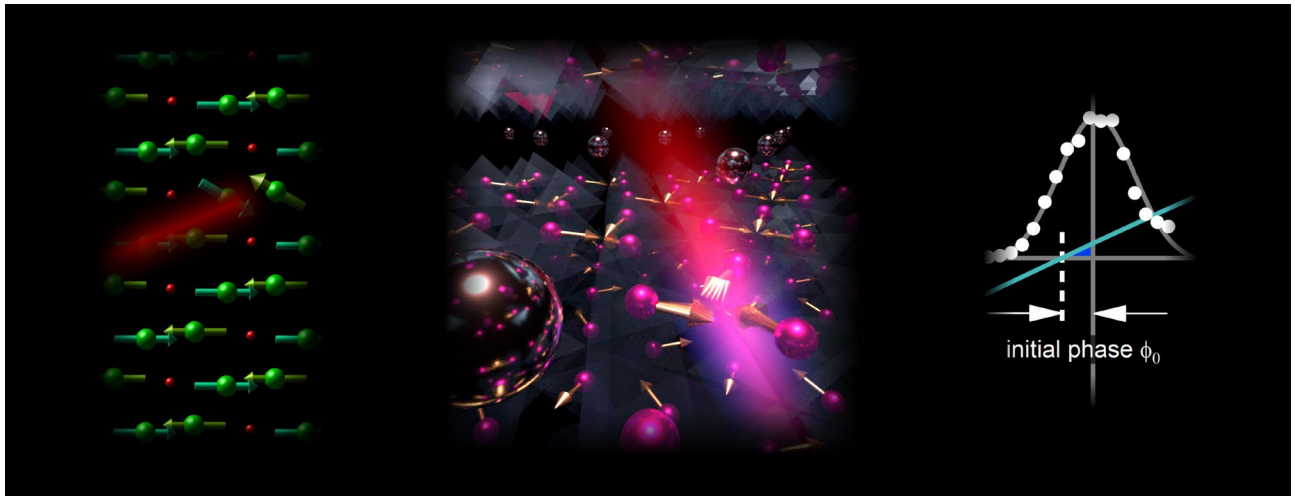


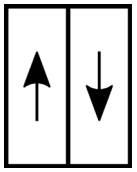
Three questions

- How can we excite antiferromagnets?
- How can we probe an AFM order parameter?
- What's next?



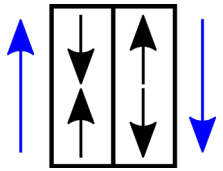
Coherent dynamics

Ferromagnetic materials

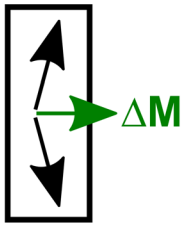


Magnetization **M**

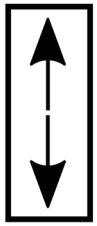
Antiferromagnetic materials



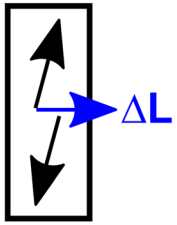
OP **L** = $\mathbf{M}_1 - \mathbf{M}_2$



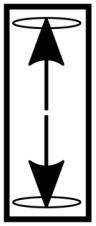
equilibrium



excitation

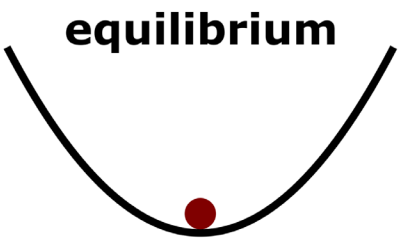


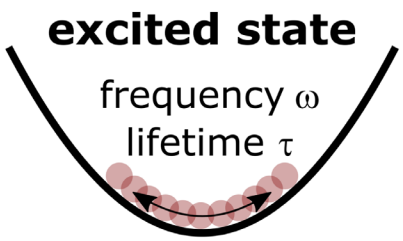
excited state



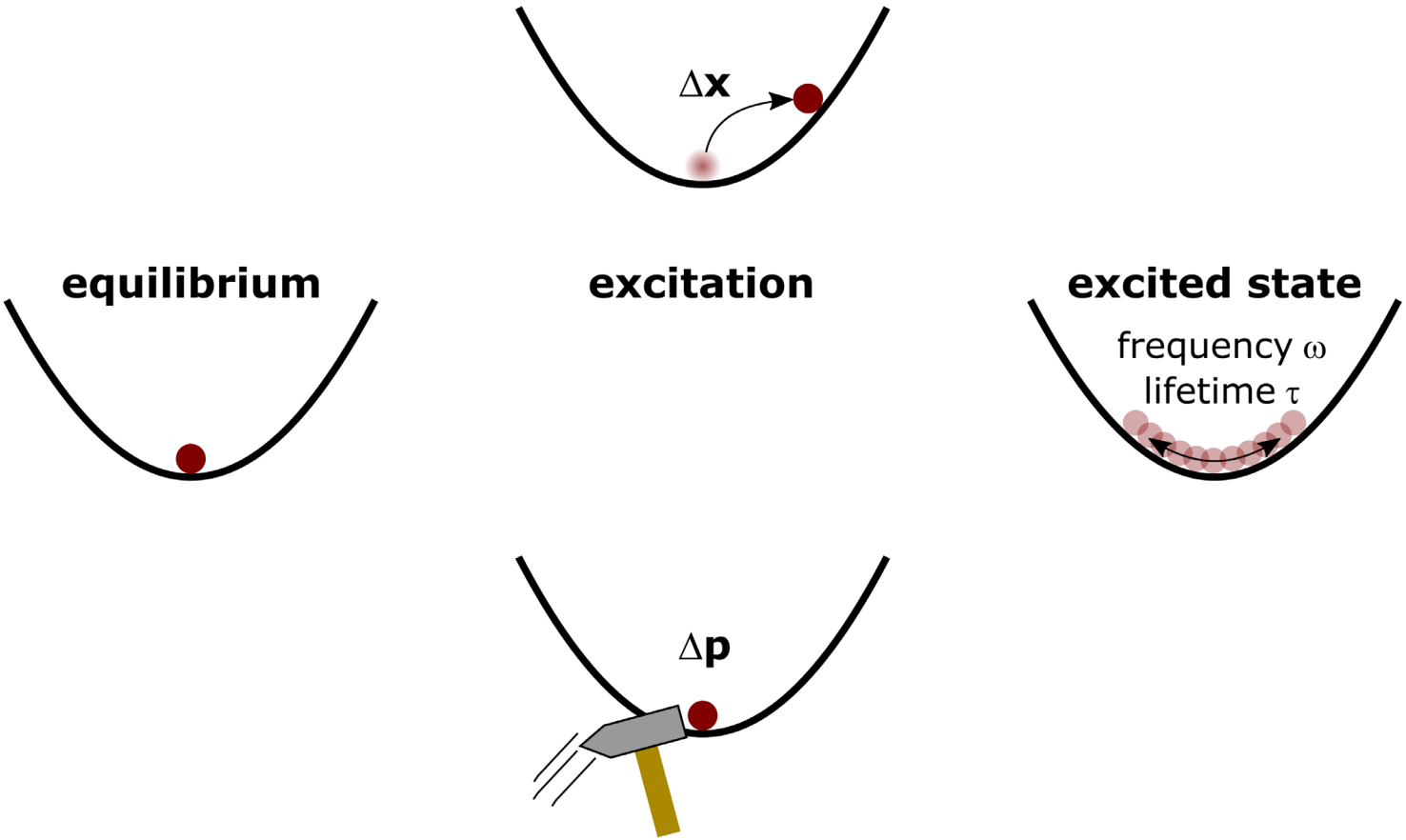
frequency ω
lifetime τ

Coherent dynamics

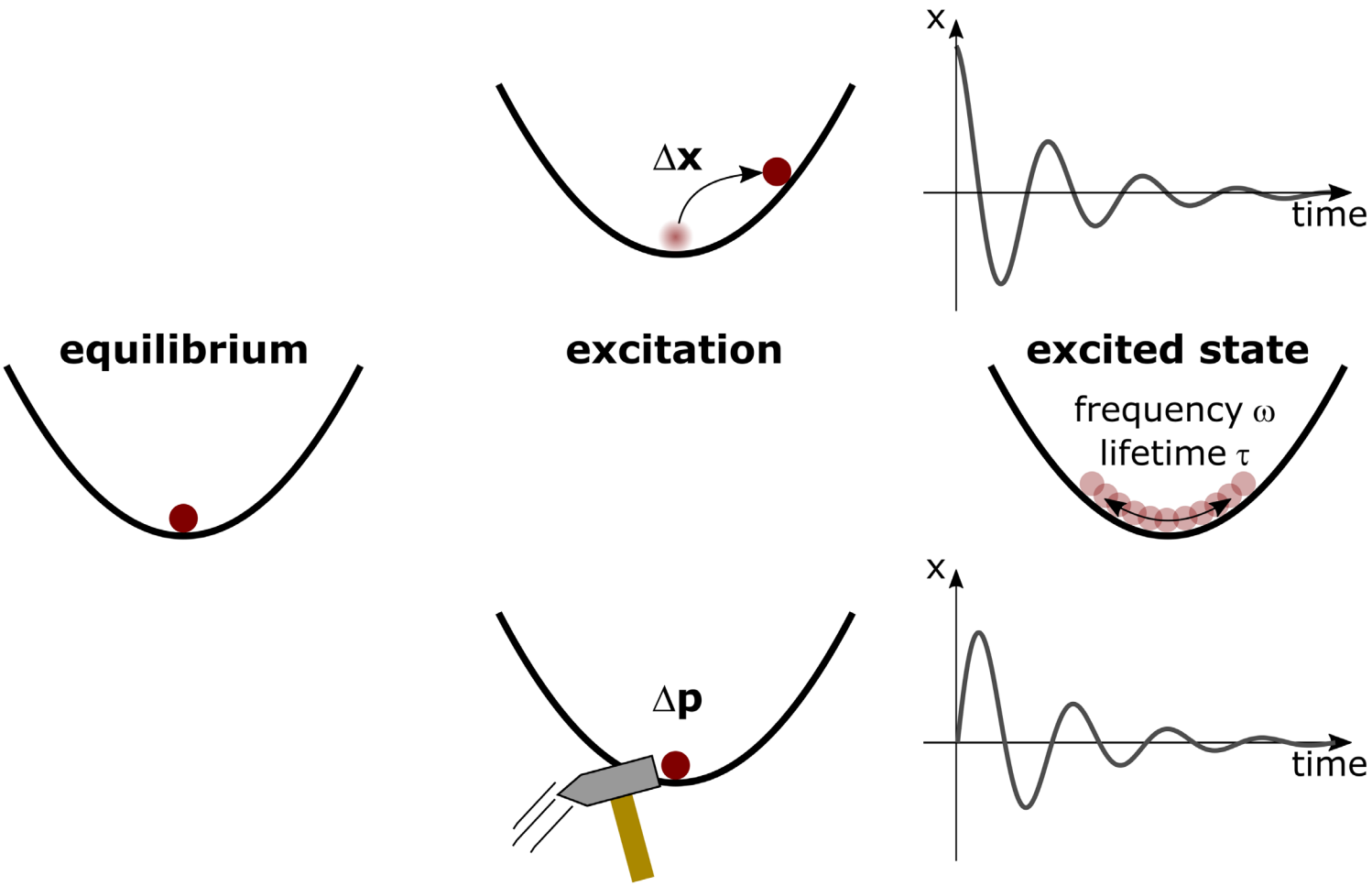




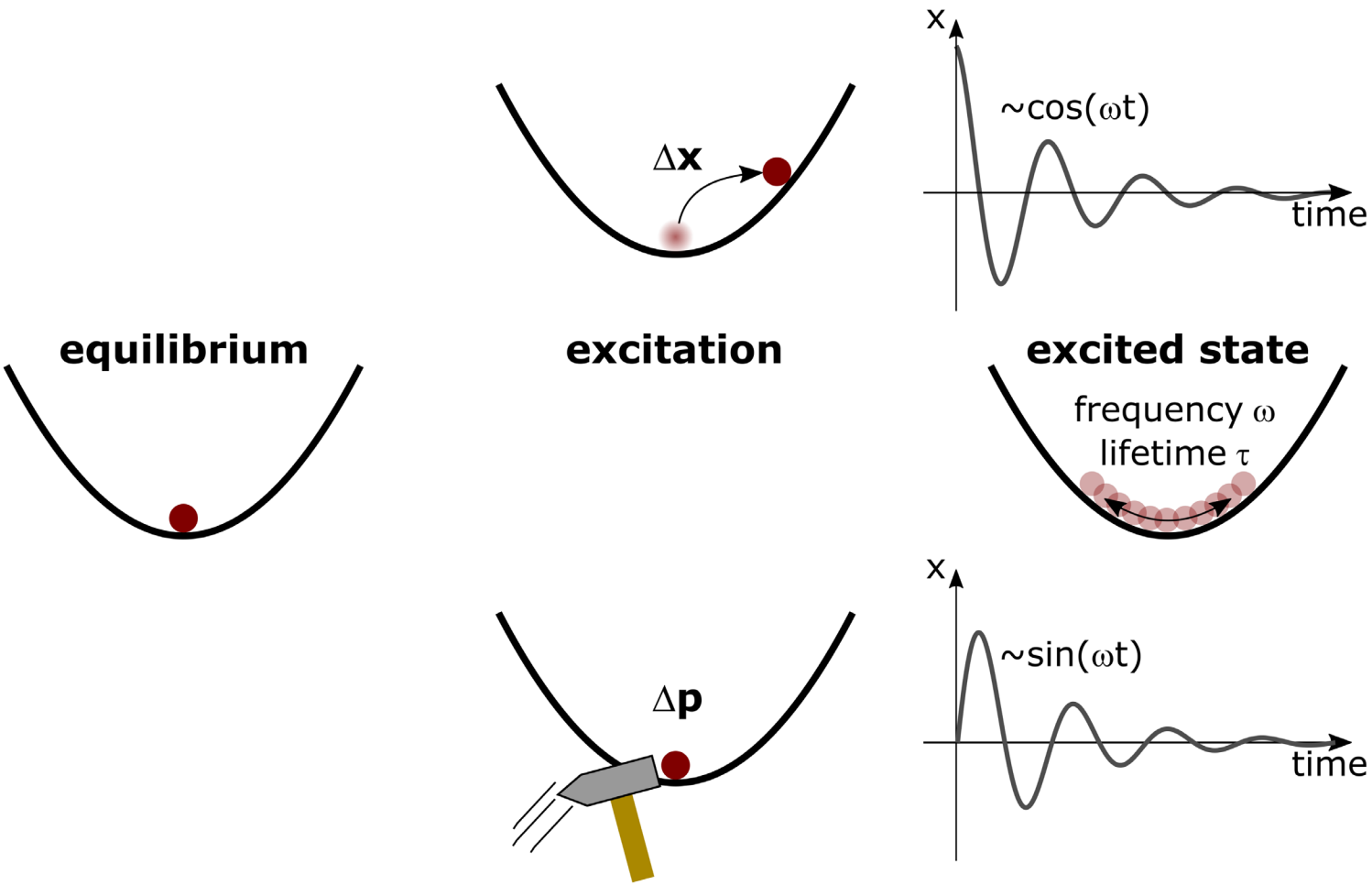
Coherent dynamics



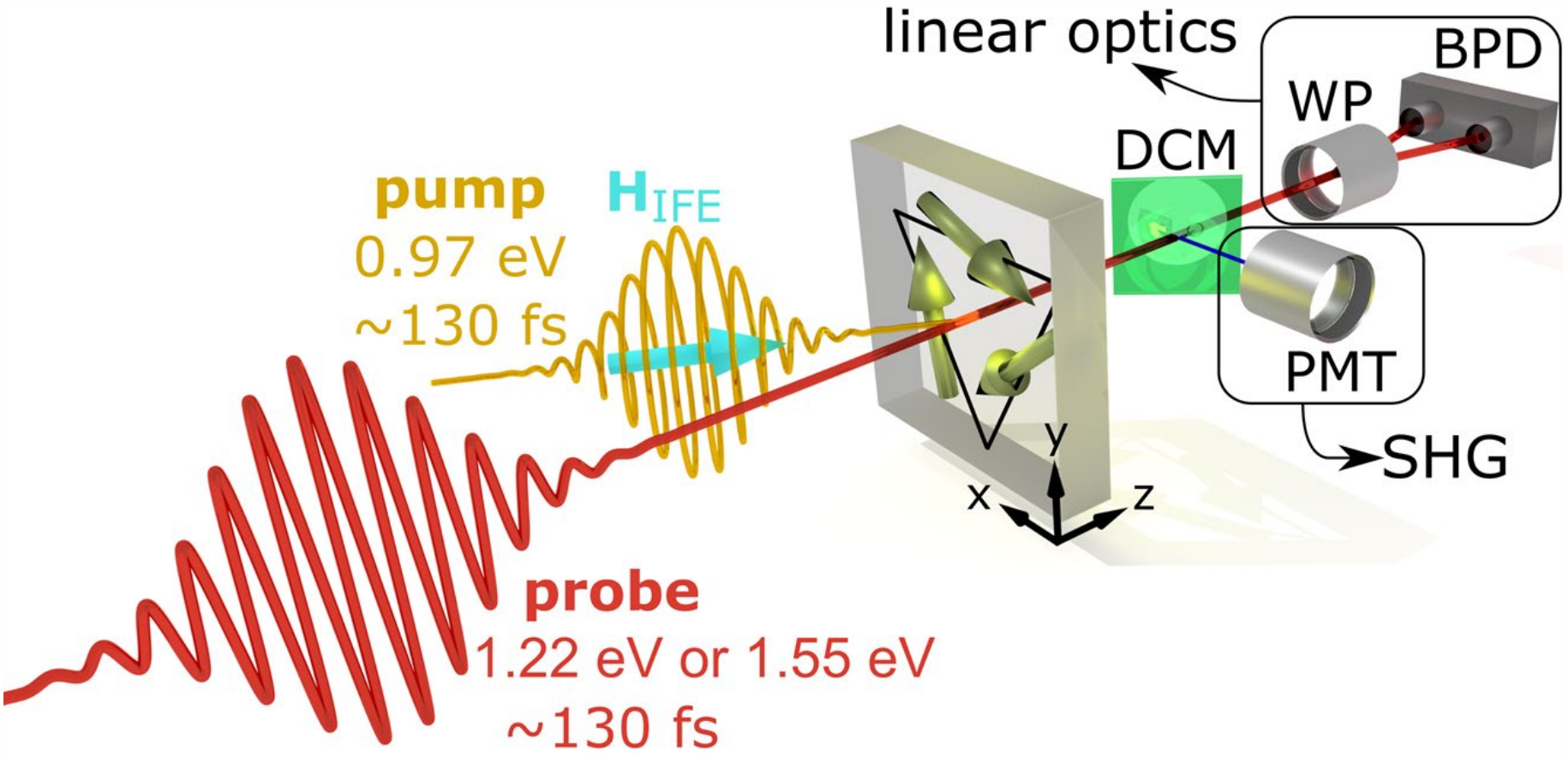
Coherent dynamics



Coherent dynamics



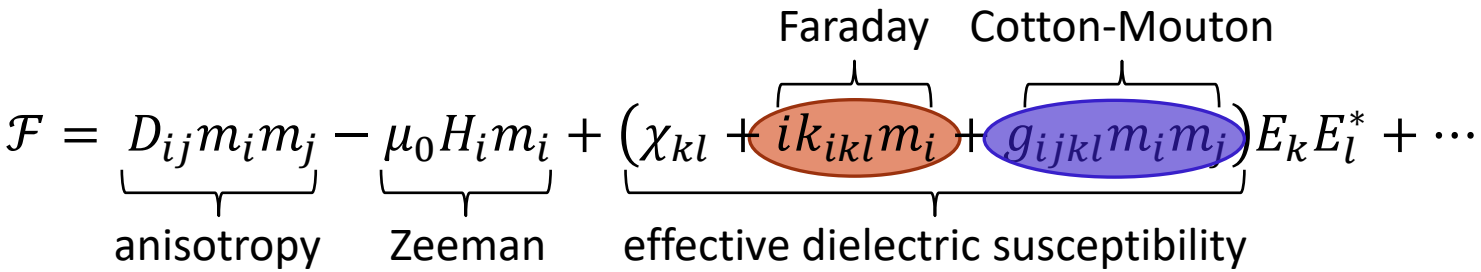
Setup


linear optics
BPD
WP
DCM
pump
H_{IFE}
0.97 eV
~130 fs
PMT
SHG
y
x
z
probe
1.22 eV or 1.55 eV
~130 fs

Inverse magneto-optical effects

$$\mathcal{F} = \underbrace{D_{ij} m_i m_j}_{\text{anisotropy}} - \underbrace{\mu_0 H_i m_i}_{\text{Zeeman}} + \underbrace{\left(\chi_{kl} + i k_{ikl} m_i + g_{ijkl} m_i m_j\right)}_{\text{effective dielectric susceptibility}} E_k E_l^* + \cdots$$

Inverse magneto-optical effects



Inverse magneto-optical effects

$$\mathcal{F} = \underbrace{D_{ij} m_i m_j}_{\text{anisotropy}} - \underbrace{\mu_0 H_i m_i}_{\text{Zeeman}} + \underbrace{\left(\chi_{kl} + \overbrace{ik_{ikl} m_i}^{\text{Faraday}} + \overbrace{g_{ijkl} m_i m_j}^{\text{Cotton-Mouton}}\right)}_{\text{effective dielectric susceptibility}} E_k E_l^* + \cdots$$

$$\mathcal{F} = \underbrace{\left(D_{ij} + \overbrace{g_{ijkl} E_k E_l^*}^{\text{Inverse CME}}\right)}_{\text{effective anisotropy}} m_i m_j - \underbrace{\left(\mu_0 H_i + \overbrace{ik_{ikl} E_k E_l^*}^{\text{Inverse FE}}\right)}_{\text{effective magnetic field}} m_i + \chi_{kl} E_k E_l^* + \cdots$$

Inverse magneto-optical effects

$$\mathcal{F} = \underbrace{D_{ij} m_i m_j}_{\text{anisotropy}} - \underbrace{\mu_0 H_i m_i}_{\text{Zeeman}} + \underbrace{\left(\chi_{kl} + \overbrace{ik_{ikl} m_i}^{\text{Faraday}} + \overbrace{g_{ijkl} m_i m_j}^{\text{Cotton-Mouton}}\right)}_{\text{effective dielectric susceptibility}} E_k E_l^* + \cdots$$

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$$\mu_0 \boldsymbol{H}_{eff} = -\frac{\partial \mathcal{F}}{\partial \boldsymbol{m}} \sim E_i E_j^*$$

Optomagnetic field pulse

Inverse magneto-optical effects

$$\mathcal{F} = \underbrace{D_{ij} m_i m_j}_{\text{anisotropy}} - \underbrace{\mu_0 H_i m_i}_{\text{Zeeman}} + \underbrace{\left(\chi_{kl} + \overbrace{ik_{ikl} m_i}^{\text{Faraday}} + \overbrace{g_{ijkl} m_i m_j}^{\text{Cotton-Mouton}}\right)}_{\text{effective dielectric susceptibility}} E_k E_l^* + \cdots$$

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$$\mu_0 \boldsymbol{H}_{eff} = -\frac{\partial \mathcal{F}}{\partial \boldsymbol{m}} \sim E_i E_j^*$$

Optomagnetic field pulse

$$\frac{\partial \boldsymbol{m}}{\partial t} = -\mu_0 \gamma \boldsymbol{m} \times \boldsymbol{H}_{eff} - \mu_0 \gamma \alpha \frac{\boldsymbol{m}}{|\boldsymbol{m}|} \times (\boldsymbol{m} \times \boldsymbol{H}_{eff})$$

Inverse magneto-optical effects

$$\mathcal{F} = \underbrace{D_{ij} m_i m_j}_{\text{anisotropy}} - \underbrace{\mu_0 H_i m_i}_{\text{Zeeman}} + \underbrace{\left(\chi_{kl} + \overbrace{ik_{ikl} m_i}^{\text{Faraday}} + \overbrace{g_{ijkl} m_i m_j}^{\text{Cotton-Mouton}}\right)}_{\text{effective dielectric susceptibility}} E_k E_l^* + \cdots$$

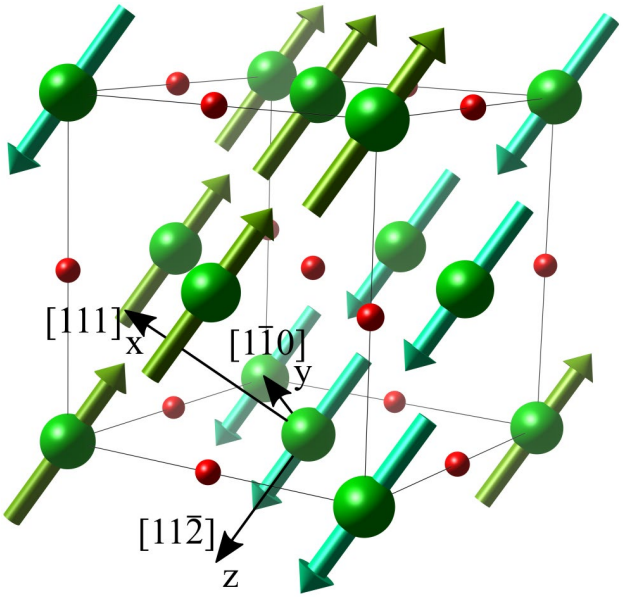
$$\mathcal{F} = \underbrace{\left(D_{ij} + \overbrace{g_{ijkl} E_k E_l^*}^{\text{Inverse CME}}\right)}_{\text{effective anisotropy}} m_i m_j - \underbrace{\left(\mu_0 H_i + \overbrace{ik_{ikl} E_k E_l^*}^{\text{Inverse FE}}\right)}_{\text{effective magnetic field}} m_i + \chi_{kl} E_k E_l^* + \cdots$$

$$\mu_0 \boldsymbol{H}_{eff} = -\frac{\partial \mathcal{F}}{\partial \boldsymbol{m}} \sim E_i E_j^*$$

Optomagnetic field pulse

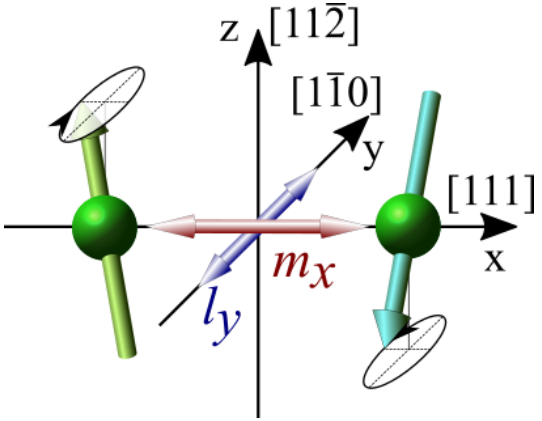
$$\frac{\partial \boldsymbol{m}}{\partial t} = -\mu_0 \gamma \boldsymbol{m} \times \boldsymbol{H}_{eff} - \mu_0 \gamma \alpha \frac{\boldsymbol{m}}{|\boldsymbol{m}|} \times (\boldsymbol{m} \times \boldsymbol{H}_{eff})$$

NiO



- Fully compensated AFM
- T_N = 523 K

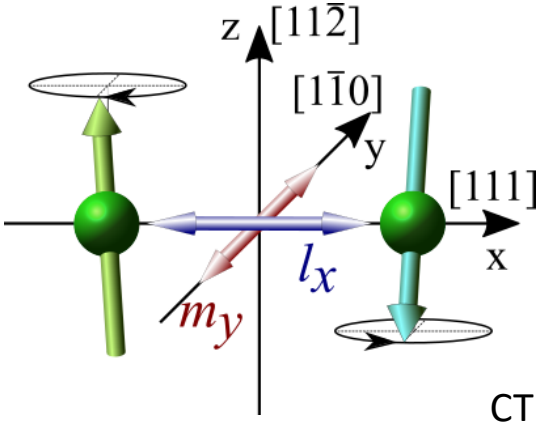
In-plane mode



$\sigma^{\pm}$ ⟷ IFE

lin ⟷ ICME

Out-of-plane mode



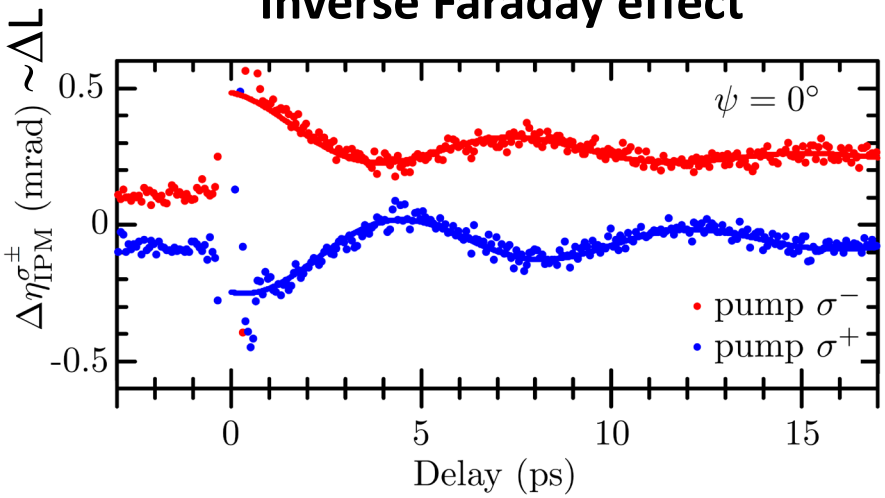
$\sigma^{\pm}$ ⟷ ICME

lin ⟷ ICME

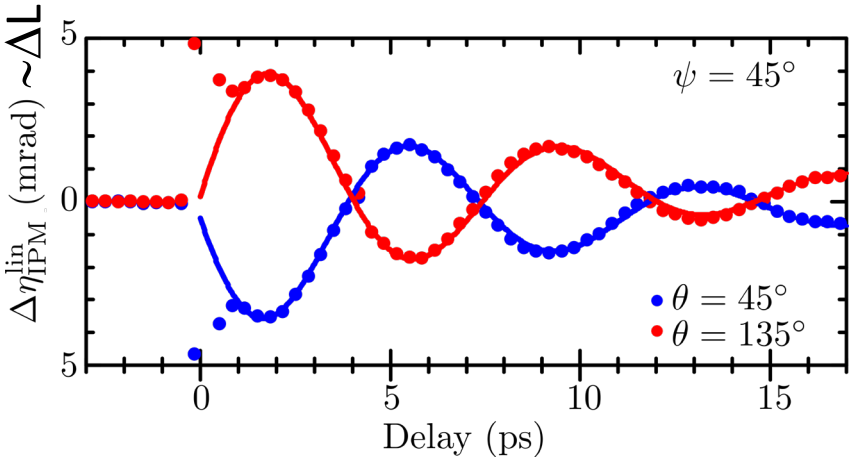
CT et al., PRB **95**, 174407 (2017)

Inverse magneto-optical effects

Inverse Faraday effect



Inverse Cotton-Mouton effect

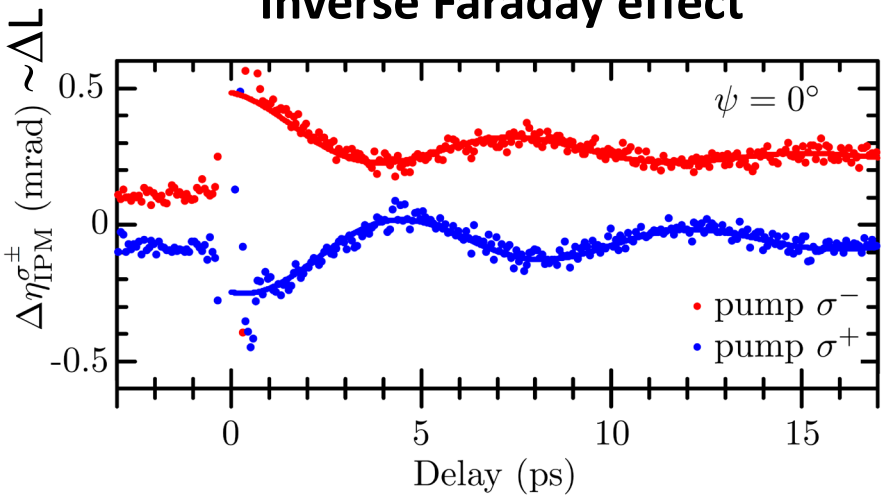


CT et al., PRB **95**, 174407 (2017)

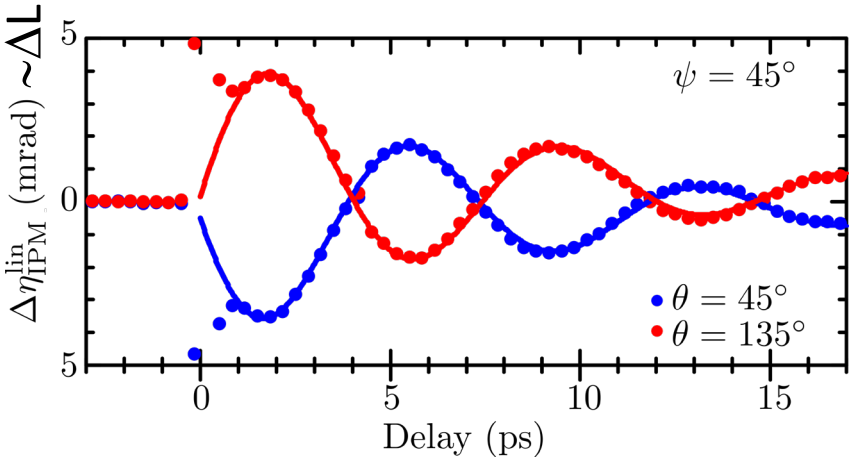
CT et al., PRB **95**, 174407 (2017)

Inverse magneto-optical effects

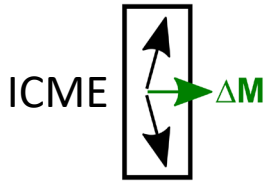
Inverse Faraday effect



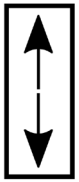
Inverse Cotton-Mouton effect

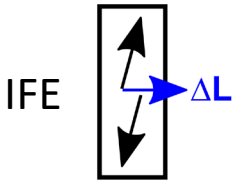


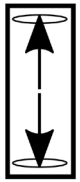
CT et al., PRB **95**, 174407 (2017)



equilibrium **excitation** **excited state**





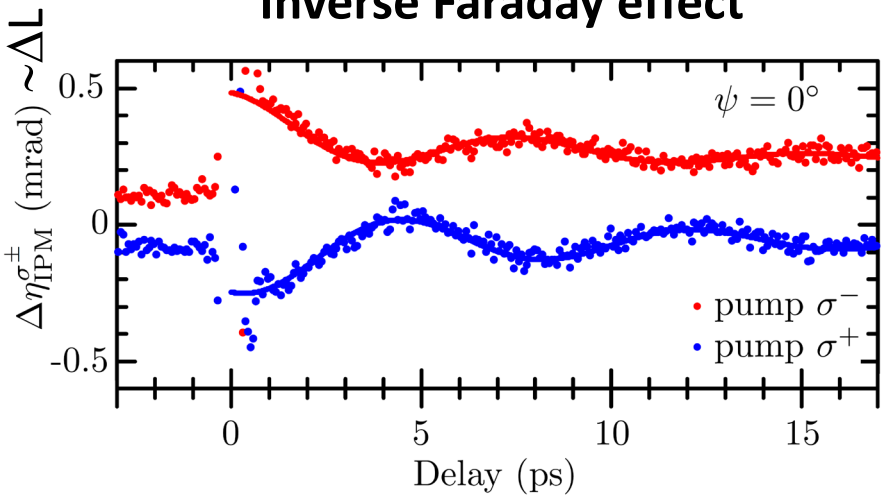


frequency ω
lifetime τ

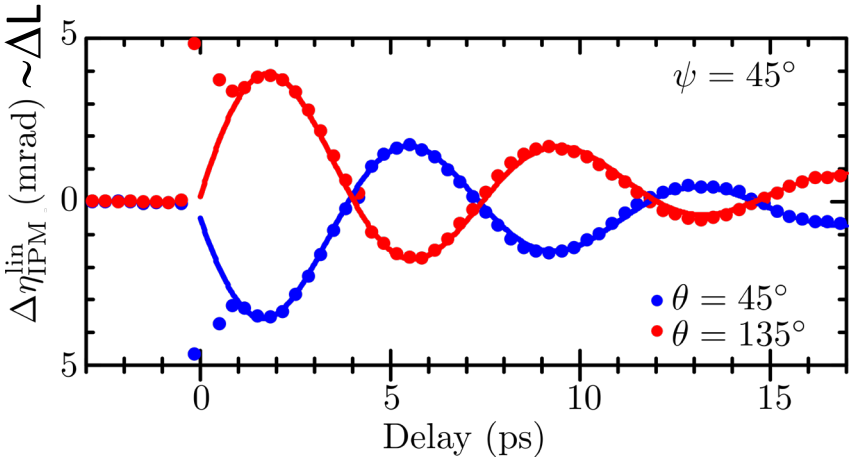
CT et al., PRB **95**, 174407 (2017)

Inverse magneto-optical effects

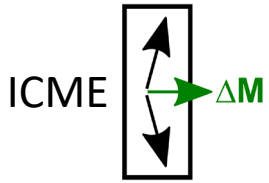
Inverse Faraday effect



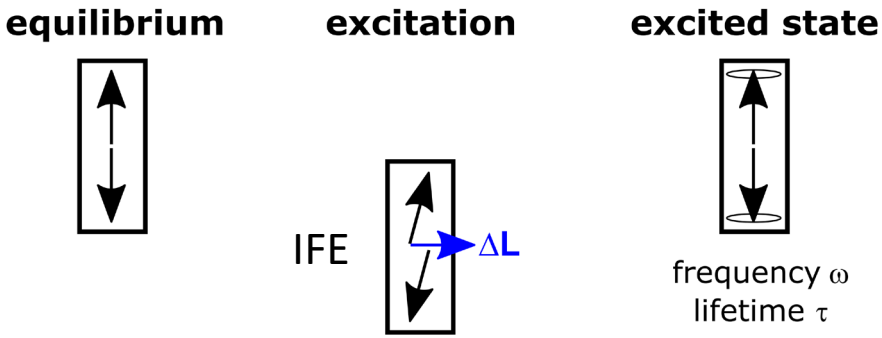
Inverse Cotton-Mouton effect



CT et al., PRB **95**, 174407 (2017)



ICME three orders of magnitude more efficient!

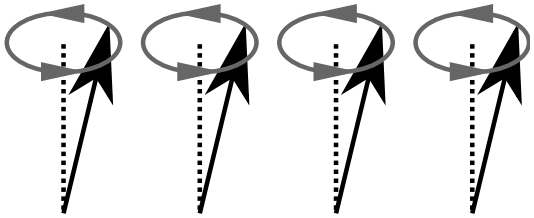


CT et al., PRB **95**, 174407 (2017)

AFM spin precessions

Ferromagnetic spin precession

$$\Delta\mathcal{F} = D(M_x^2 + M_y^2)$$



Magnetic moments parallel at all times

CT et al., PRB **95**, 174407 (2017)

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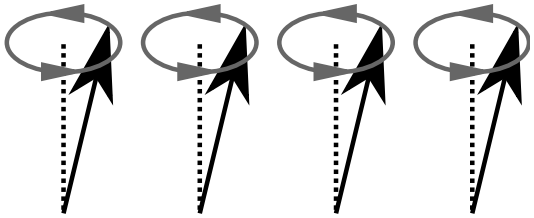
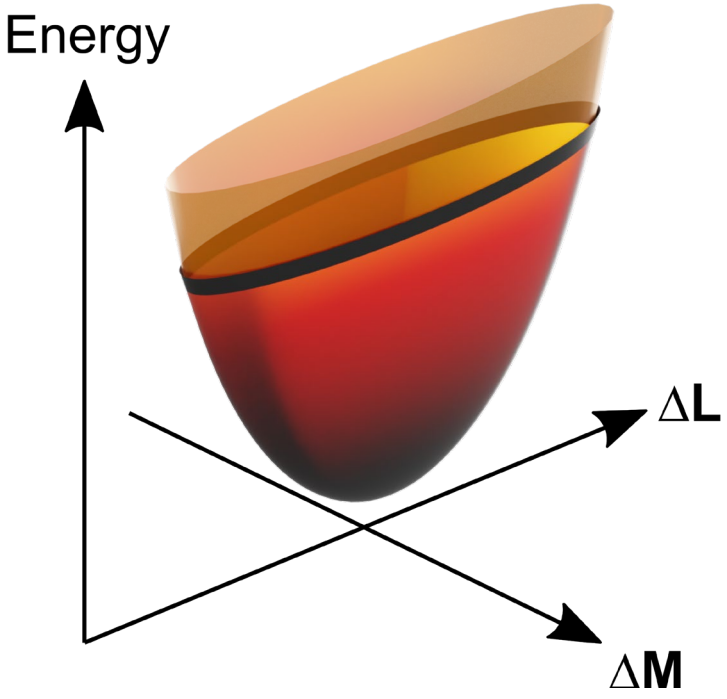


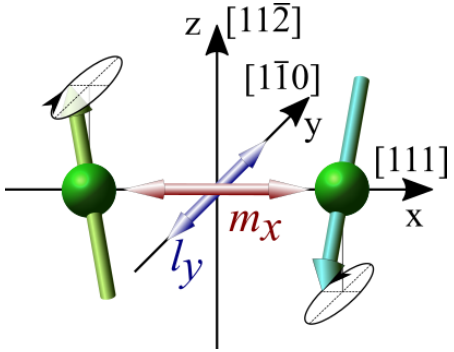
Magnetic moments parallel at all times

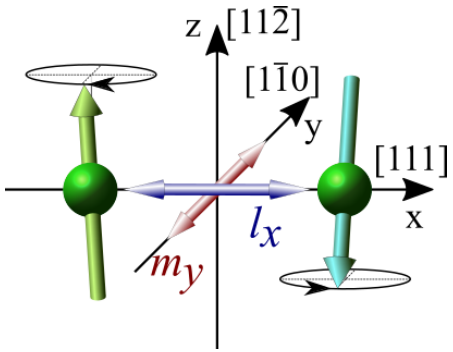
Antiferromagnetic spin precession

$$\Delta\mathcal{F} = \underbrace{J(\Delta \boldsymbol{M})^2}_{\text{Exchange}} + \underbrace{D((\Delta \boldsymbol{M})^2 + (\Delta \boldsymbol{L})^2)}_{\text{Anisotropy}}$$

Exchange >> Anisotropy



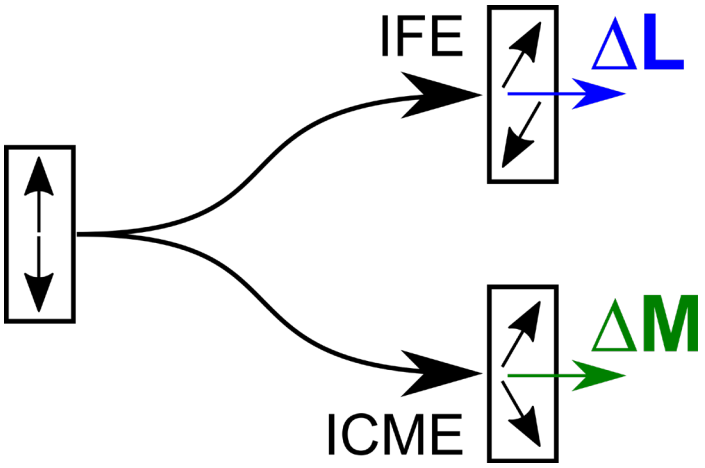


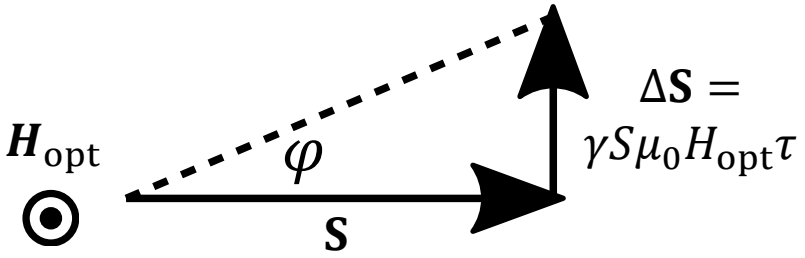


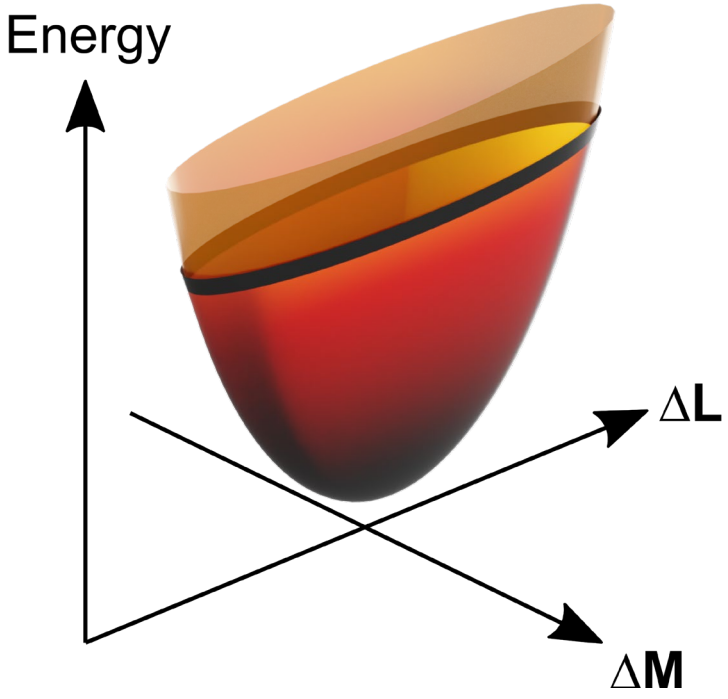
CT et al., PRB **95**, 174407 (2017)

AFM spin excitations

$$\Delta\mathcal{F} = \underbrace{J(\Delta \boldsymbol{M})^2}_{\text{Exchange}} + \underbrace{D((\Delta \boldsymbol{M})^2 + (\Delta \boldsymbol{L})^2)}_{\text{Anisotropy}} \quad (\text{Exchange} \gg \text{Anisotropy})$$



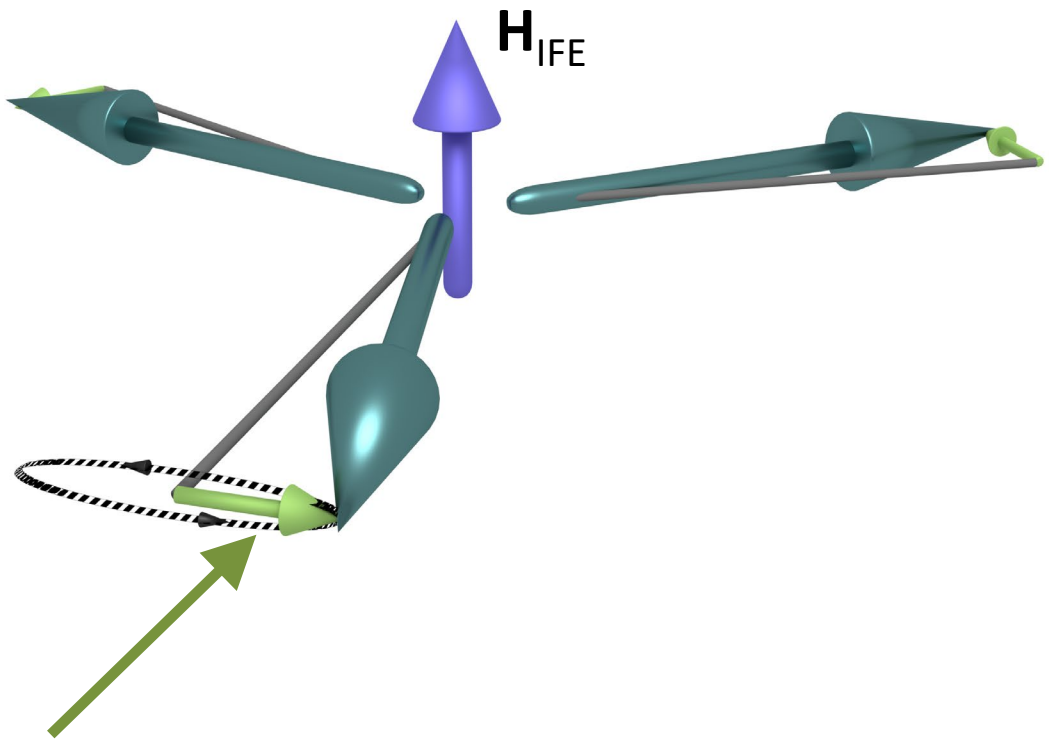




$\mu_0 H_{\text{opt}} = 1\ \text{T}$
$\tau = 100\ \text{fs}$
$\Rightarrow \varphi = 1°$

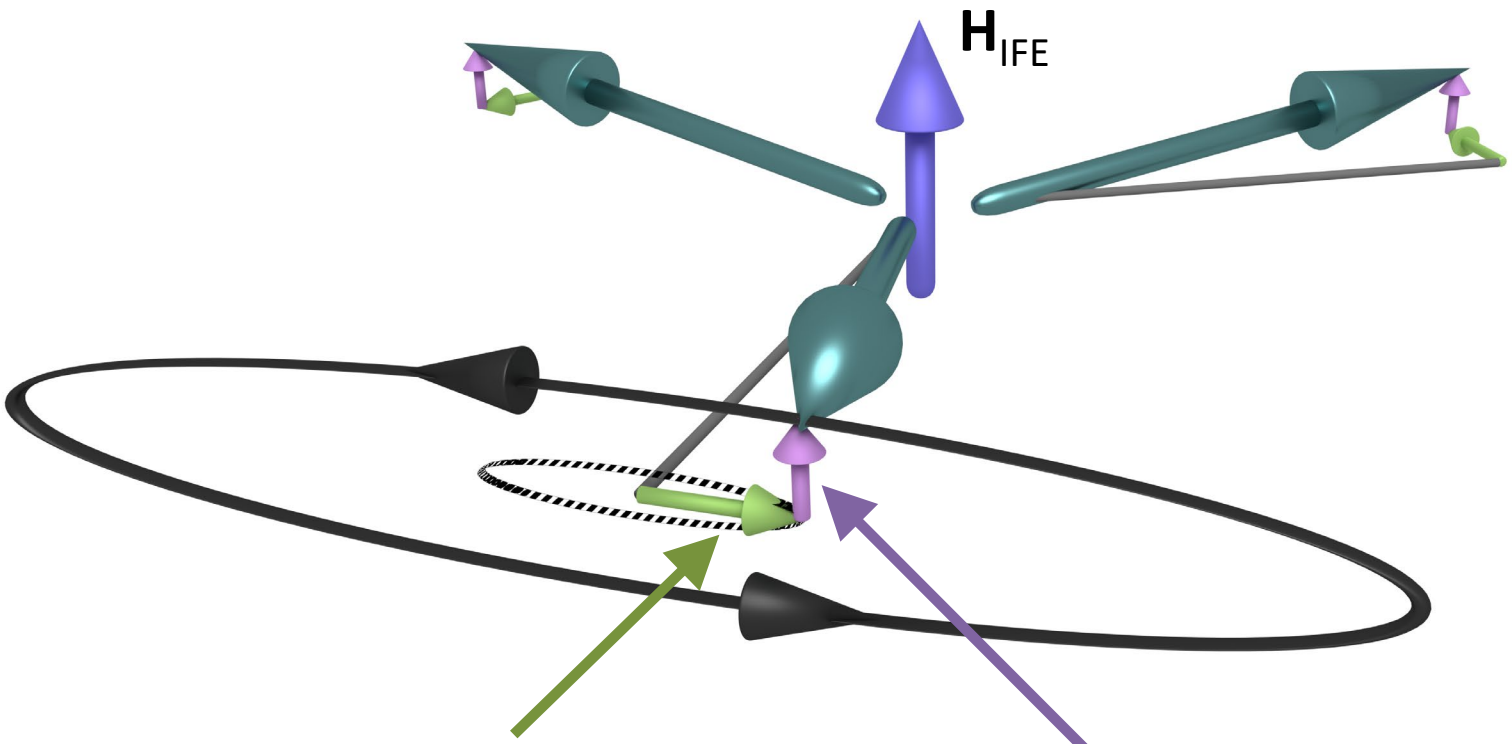
Independent of exchange/anisotropy constants

AFM spin precessions



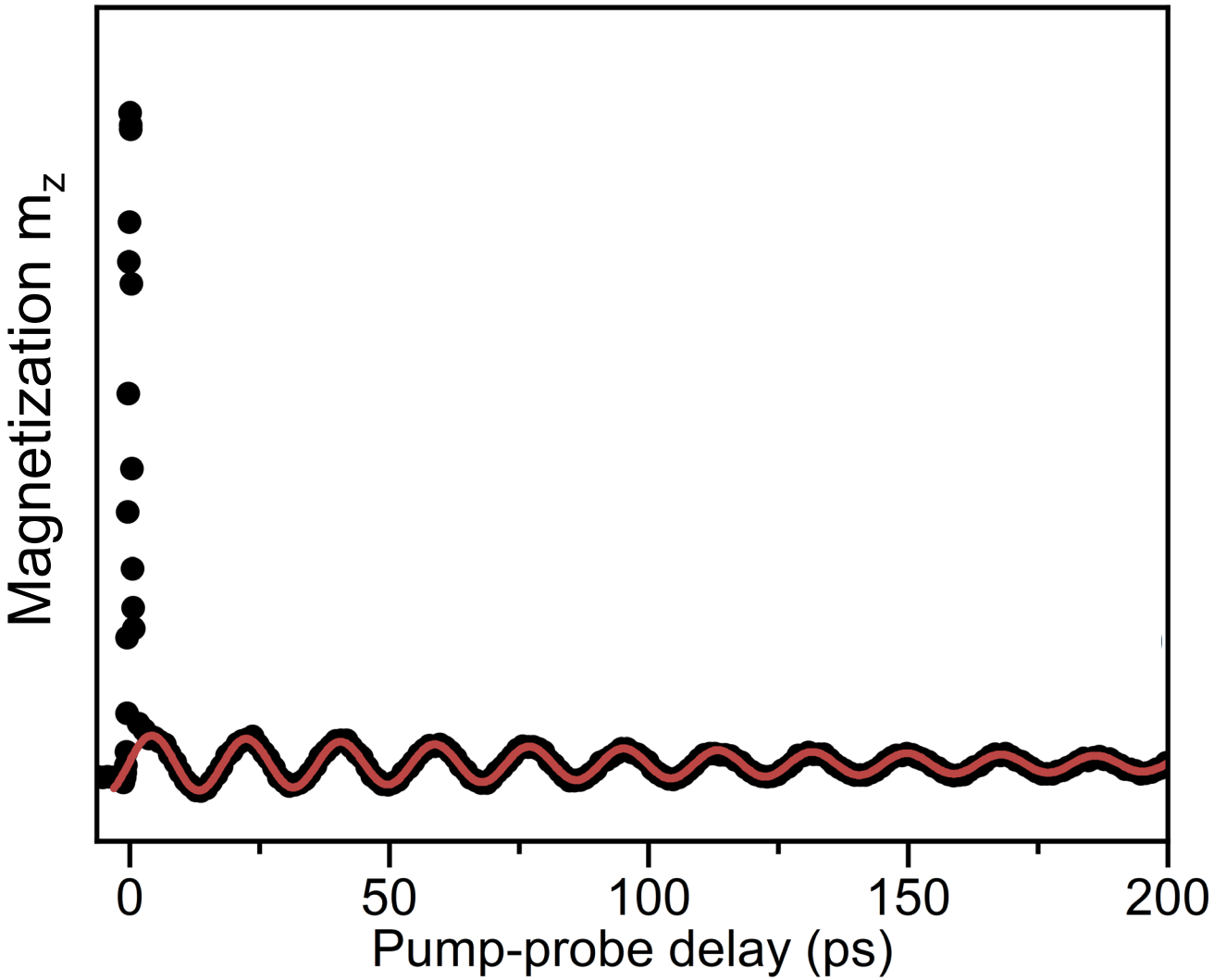
$$\dot{\boldsymbol{m}} = -\gamma_0 \boldsymbol{m} \times \boldsymbol{H}_{\mathrm{IFE}}$$

AFM spin precessions



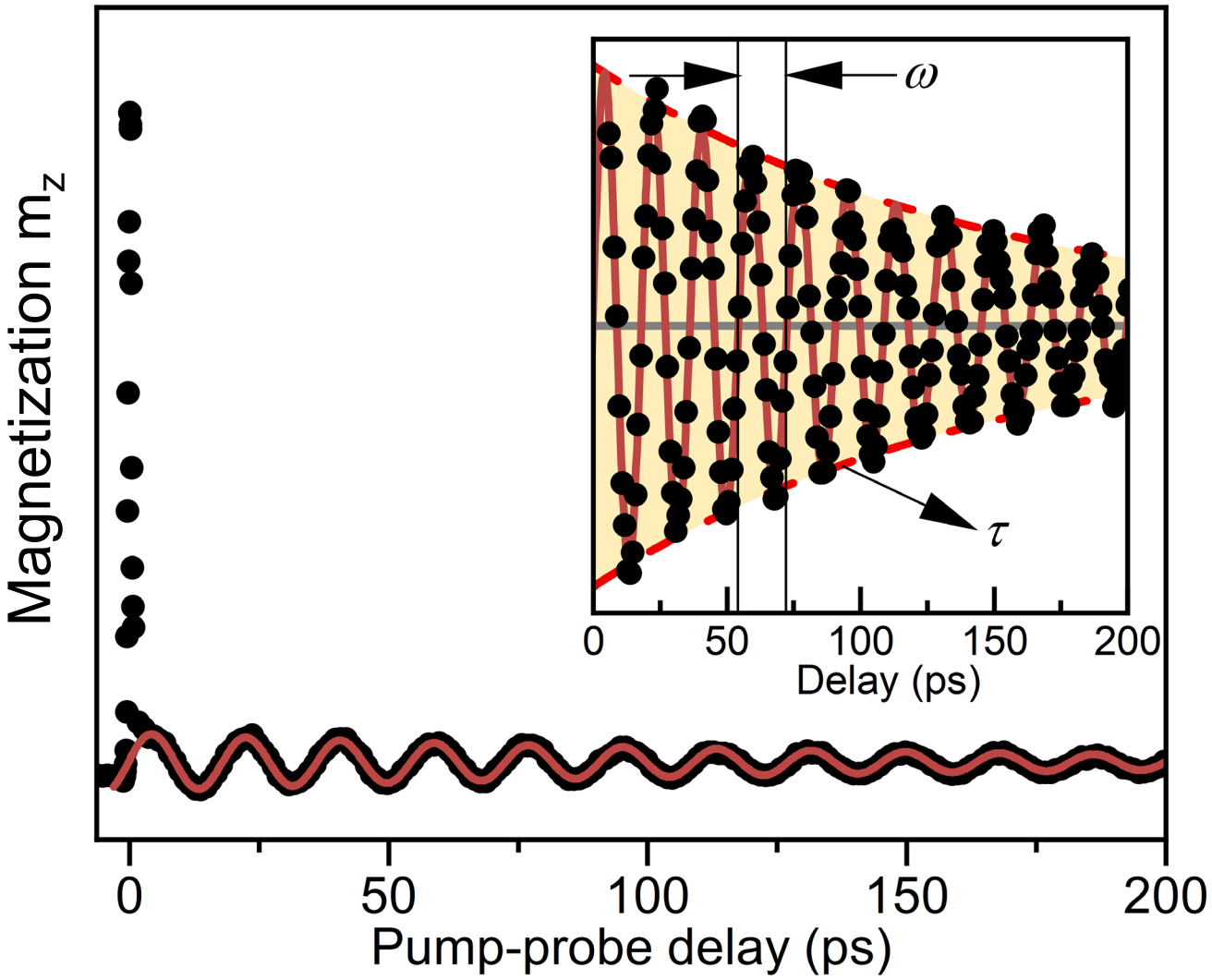
$$\dot{\boldsymbol{m}} = -\gamma_0 \boldsymbol{m} \times \boldsymbol{H}_{\mathrm{IFE}} - \gamma_0 \alpha \boldsymbol{m} \times (\boldsymbol{m} \times \boldsymbol{H}_{\mathrm{IFE}})$$

Ultrafast spin excitation via damping torques



CT et al., Nat. Commun. **11**, 6142 (2020)

Ultrafast spin excitation via damping torques



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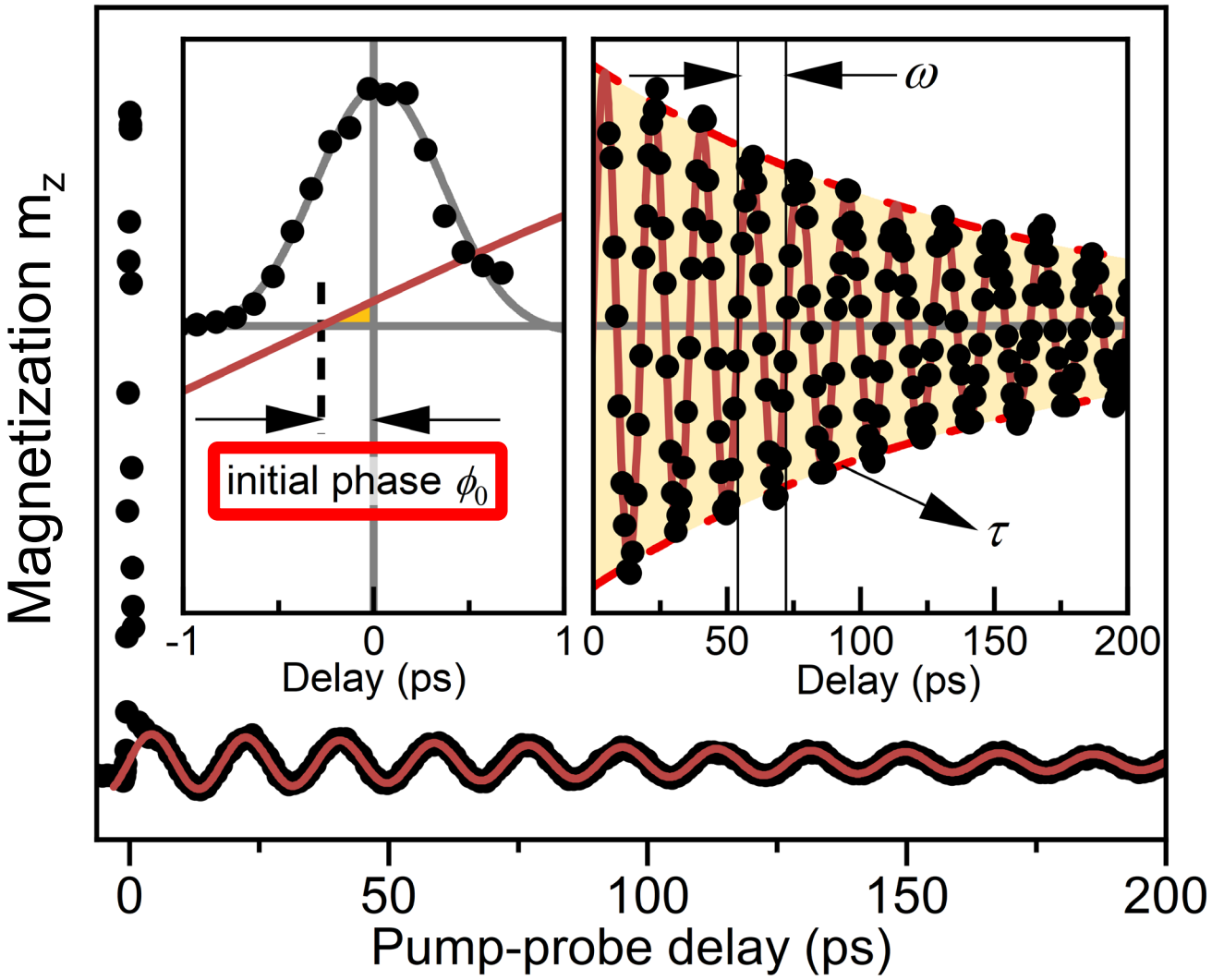

CT et al., Nat. Commun. **11**, 6142 (2020)

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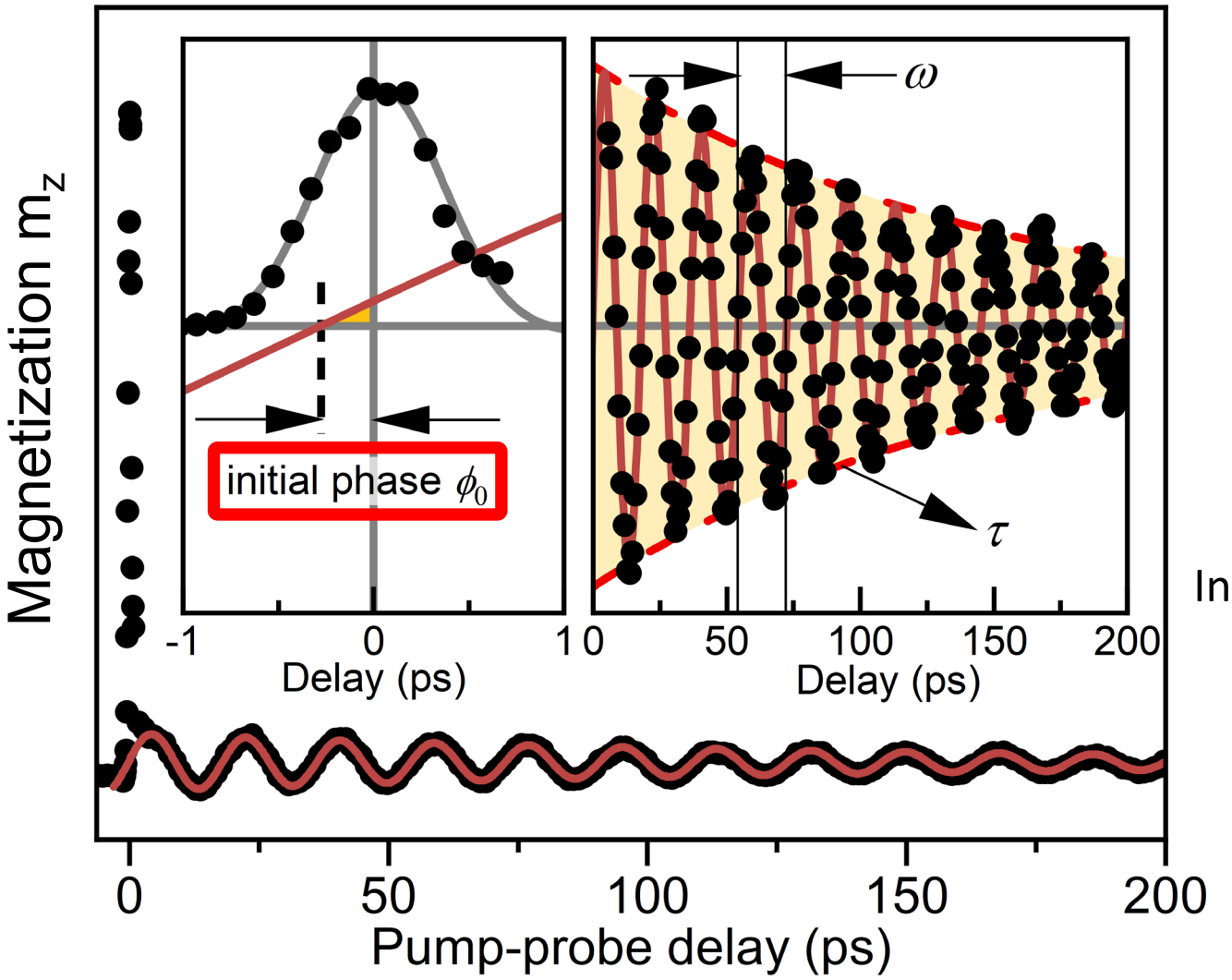


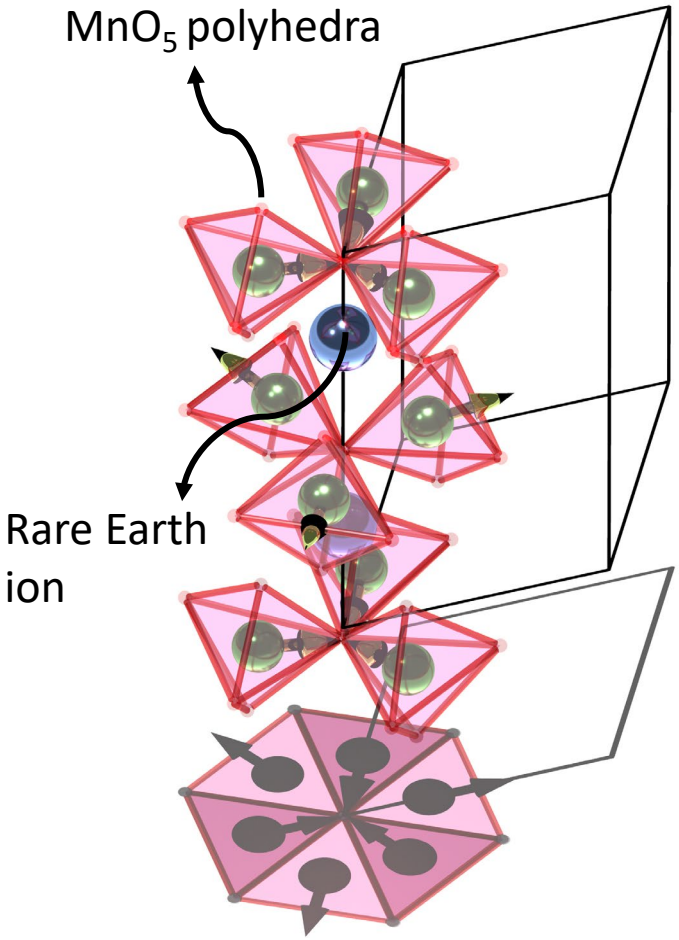
Two ways of measuring Gilbert damping α

Initial phase ϕ_0

Lifetime τ

CT et al., Nat. Commun. **11**, 6142 (2020)

h-RMnO$_3$



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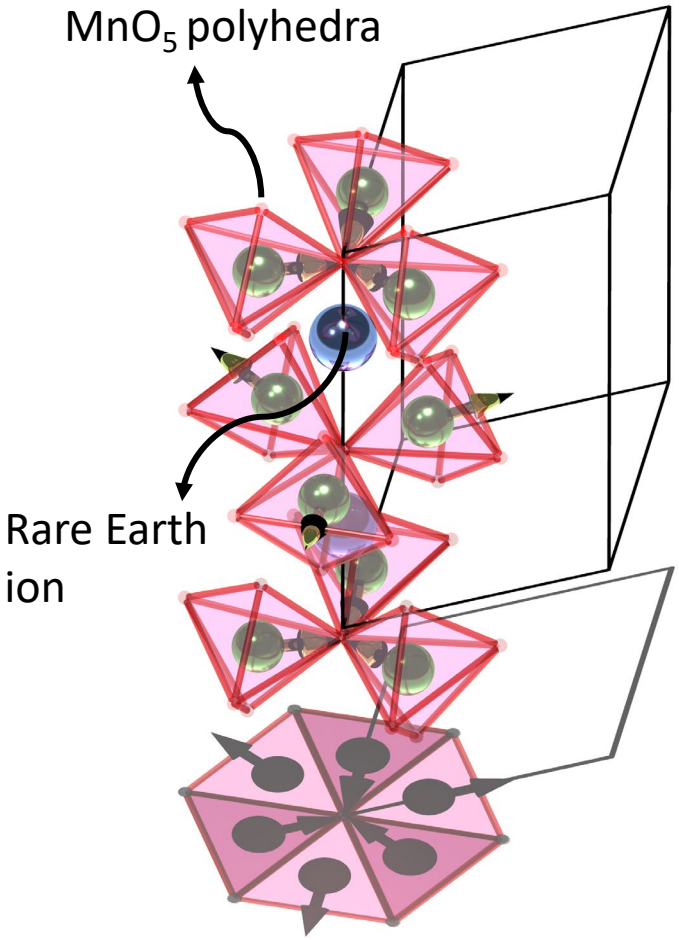

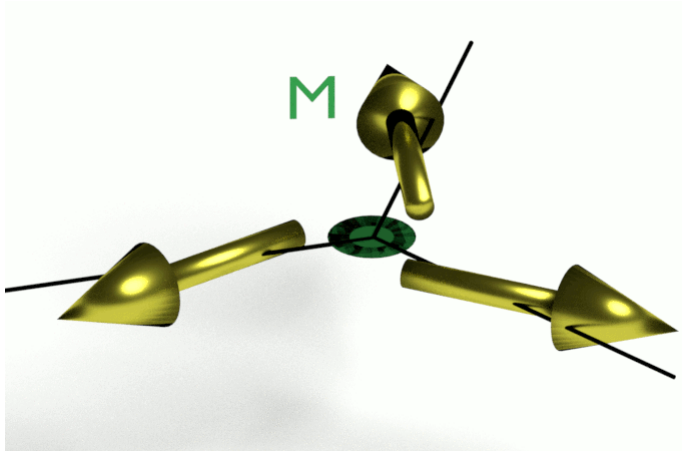

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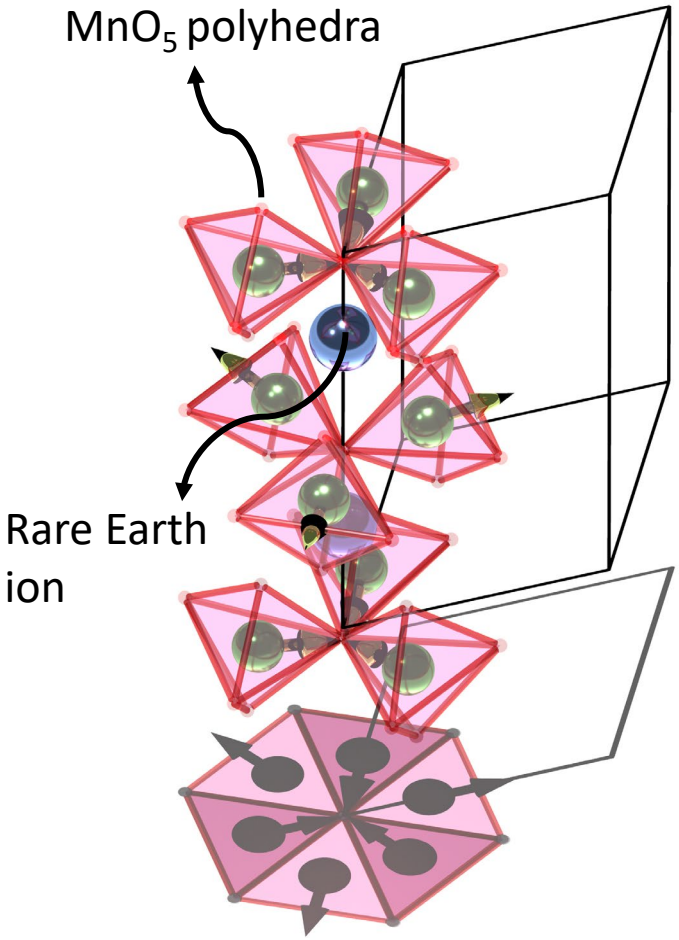
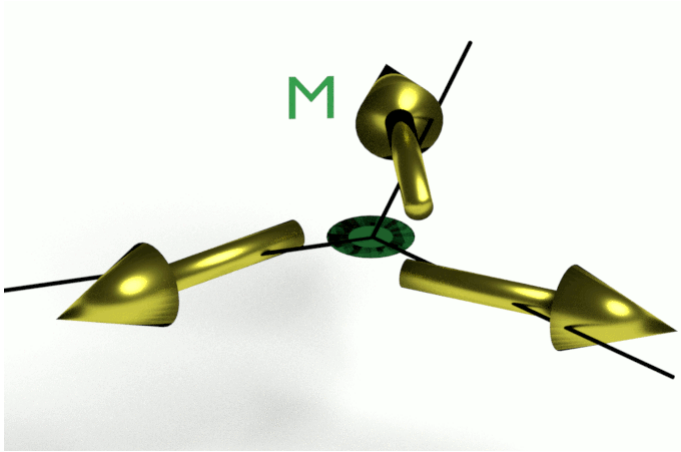
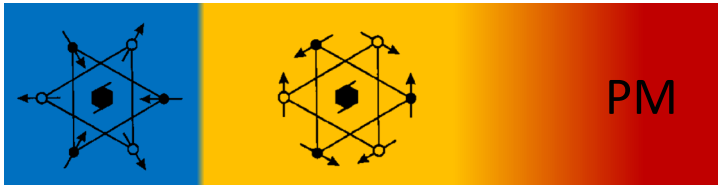




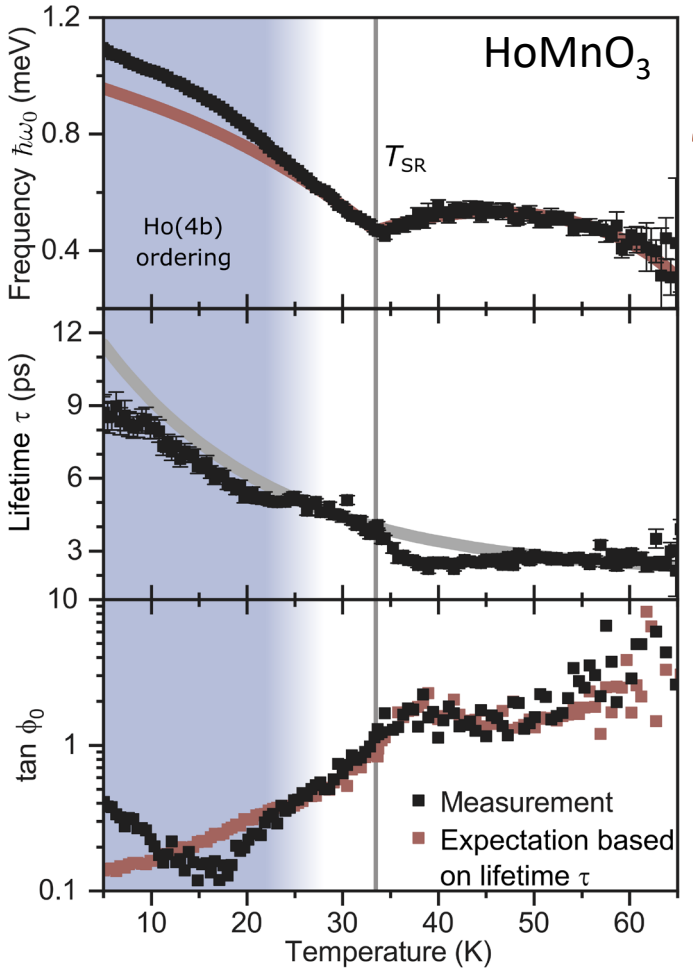
$HoMnO_3$



T_{SR} = 33 K T_N = 72 K

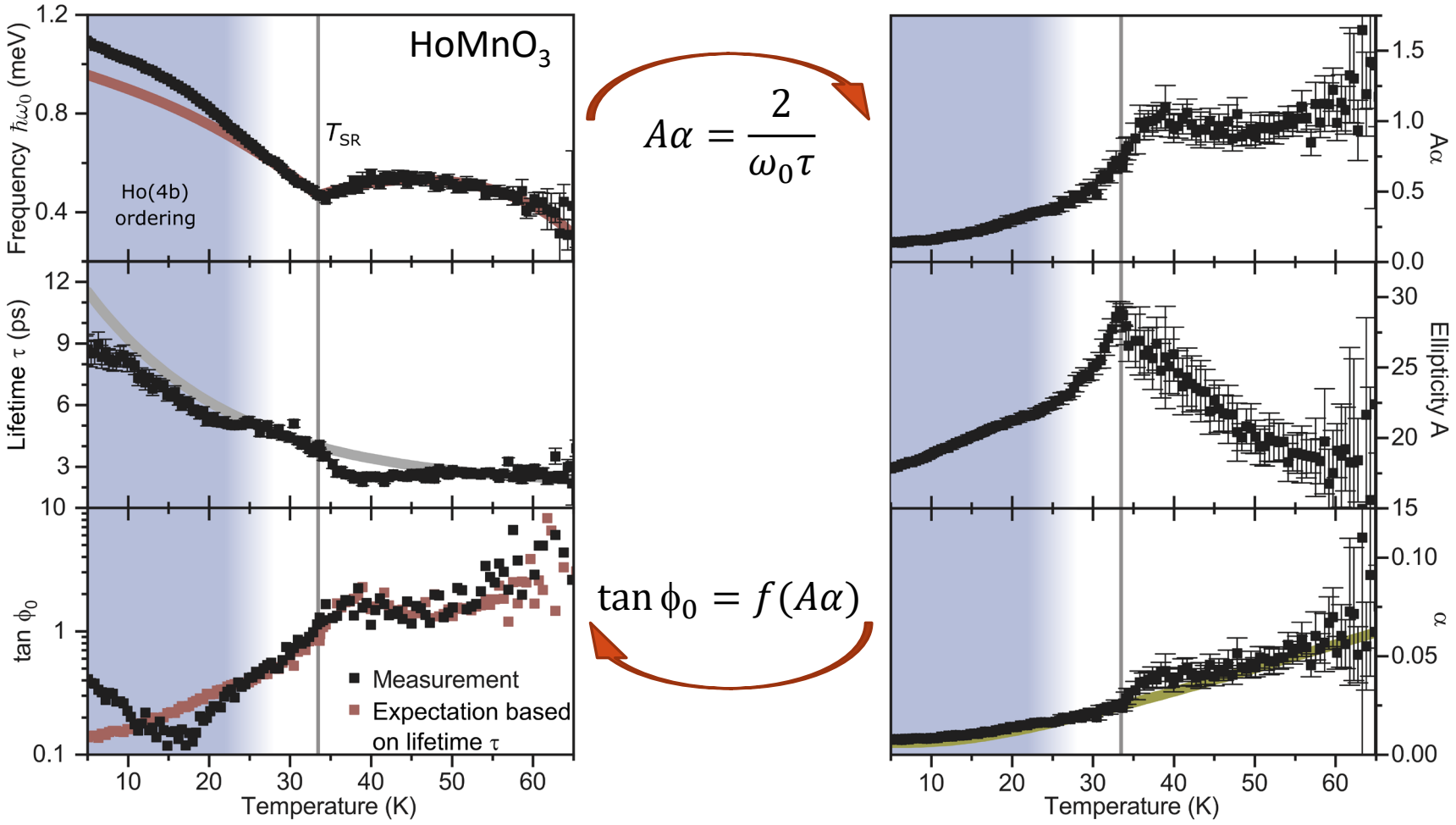
$YMnO_3$

Ultrafast spin excitation via damping torques



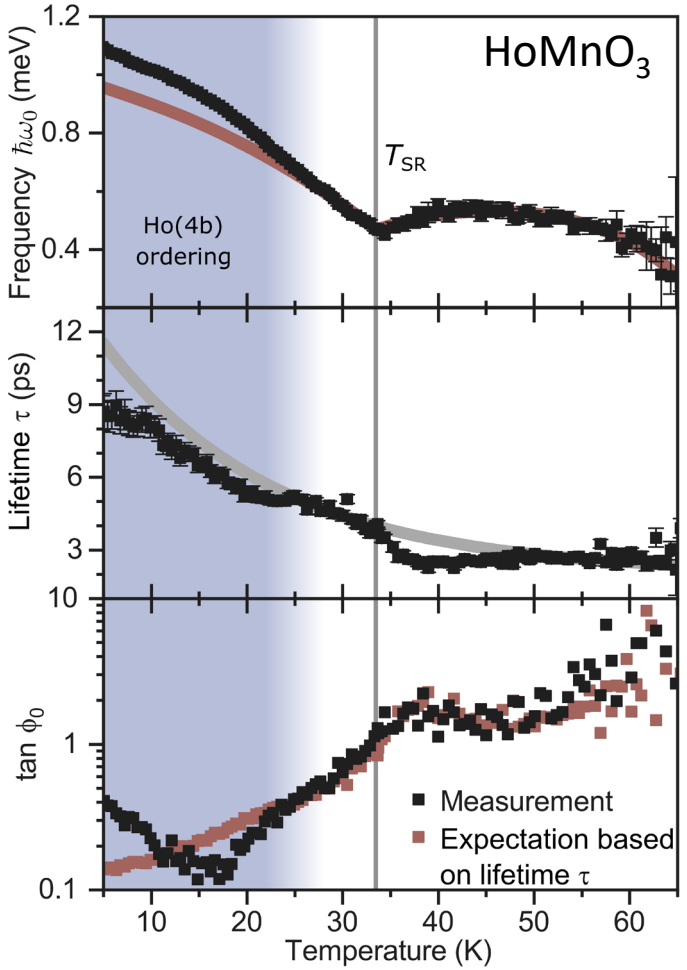
CT et al., Nat. Commun. **11**, 6142 (2020)

Ultrafast spin excitation via damping torques



CT et al., Nat. Commun. **11**, 6142 (2020)

Ultrafast spin excitation via damping torques



$\tan(\phi_0)$ ~ corresponds to energy ratio:

$$\frac{\textbf{spin excitation via damping torque of IFE}}{\textbf{spin excitation via field torque of IFE}}$$

→ $\tan(\phi_0) > 1$ implies: excitation via damping torque dominating despite $\alpha \ll 1$

Reason: strongly elliptical spin precession in antiferromagnets ($A \gg 1$)

CT et al., Nat. Commun. **11**, 6142 (2020)

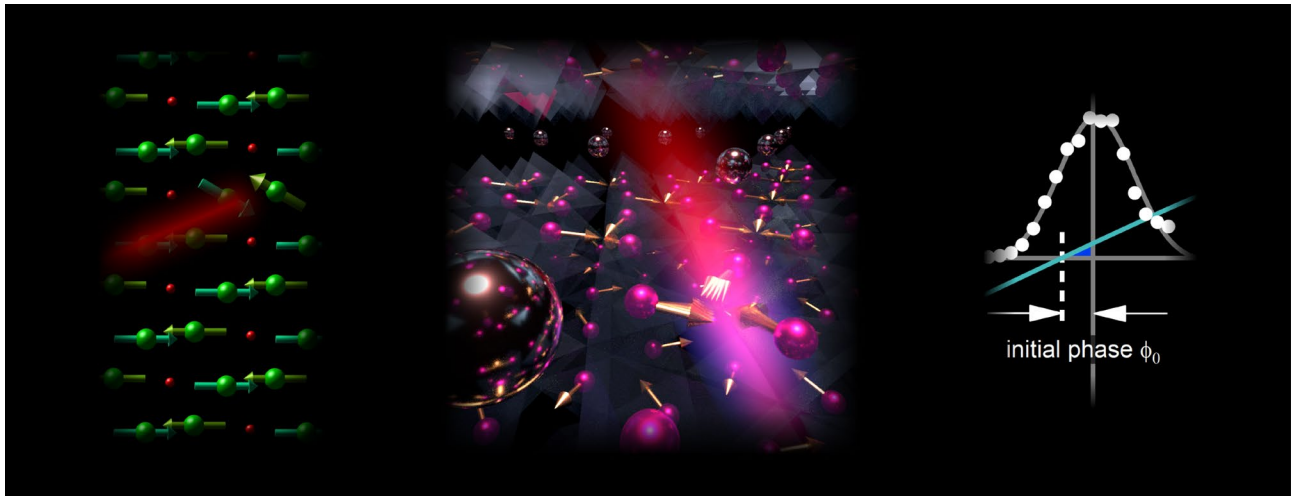
Three questions

- How can we excite antiferromagnets?

  → Inverse magneto-optical effects (PRB 95, 174407 (2017), Nat. Commun. **11**, 6142 (2020))

- How can we probe an AFM order parameter?

- What's next?



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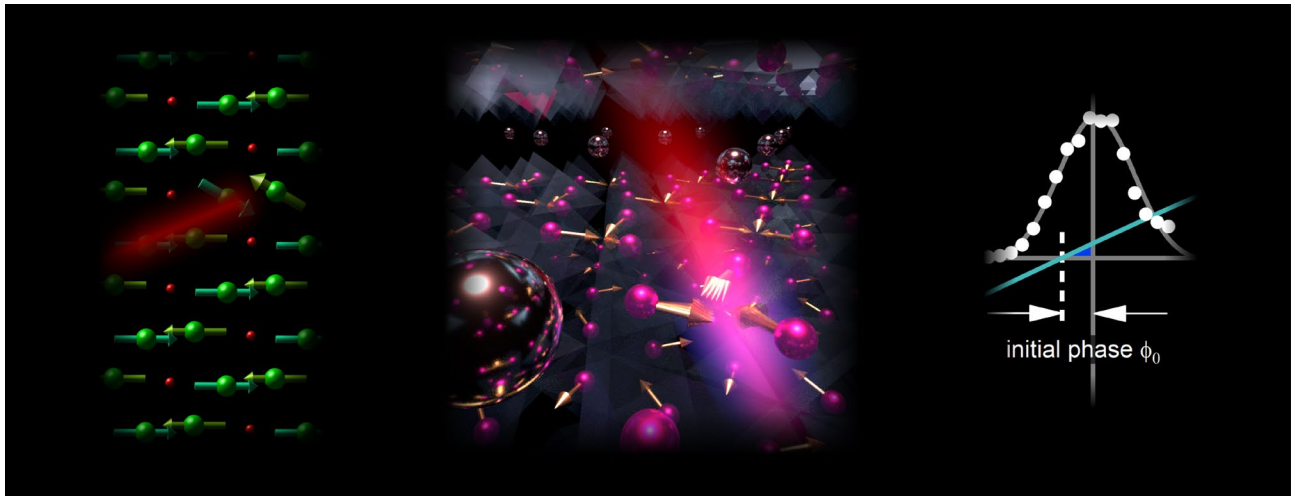
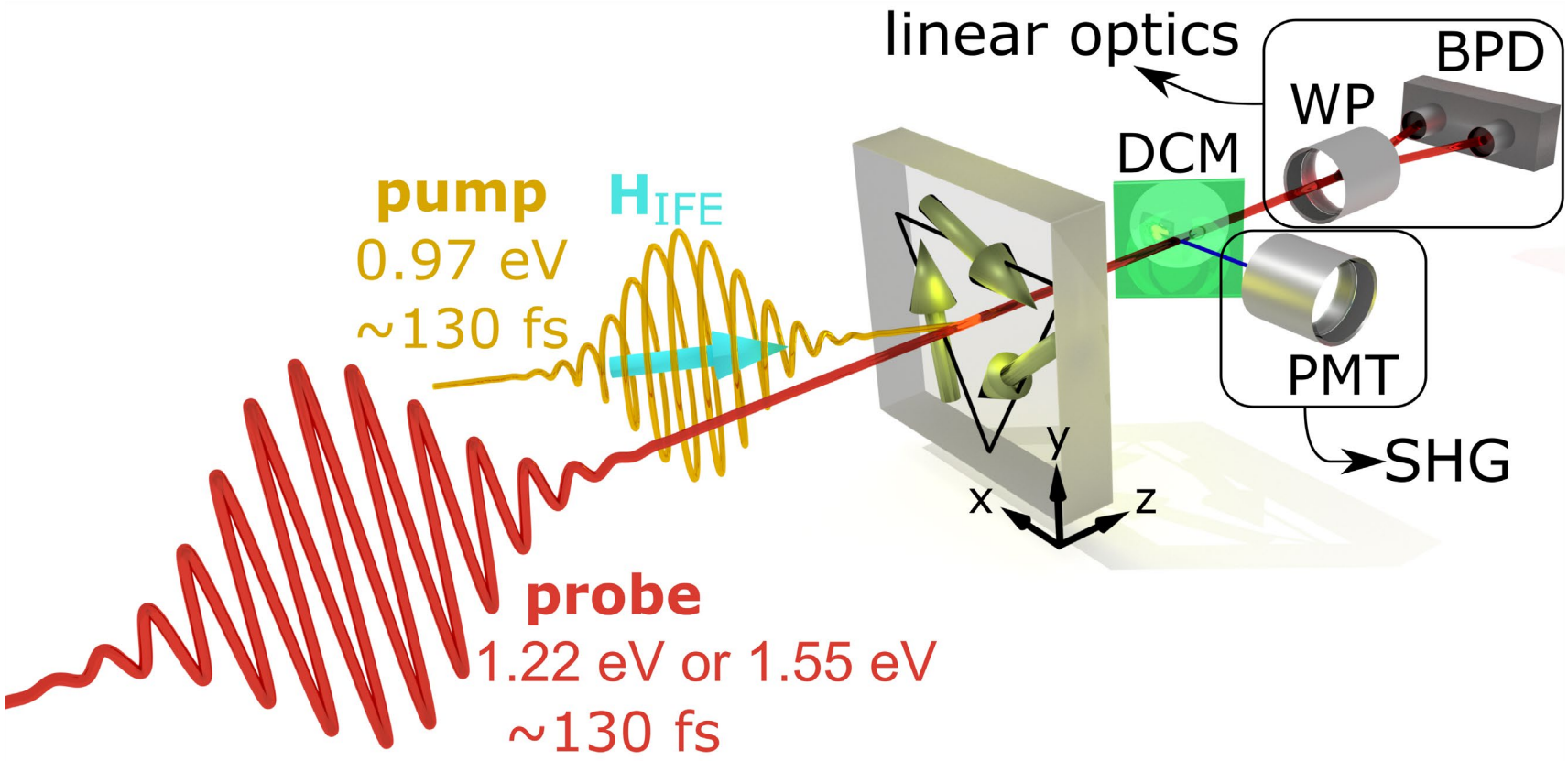


Setup

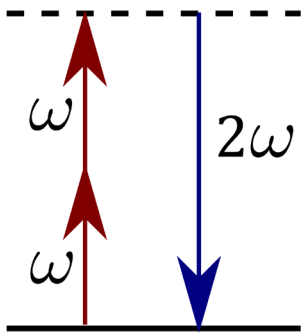


Independent/simultaneous detection of linear and nonlinear optics

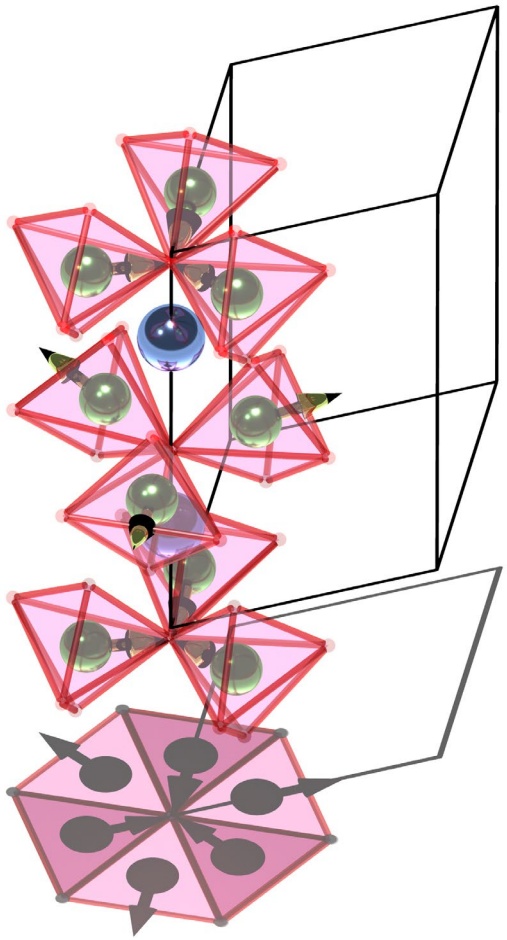
h-$YMnO_3$

Optical second-harmonic generation

$$P_i(2\omega) = \varepsilon_0 \chi_{ijk}(\boldsymbol{L}) E_j(\omega) E_k(\omega)$$



h-$YMnO_3$



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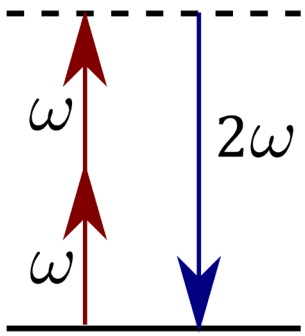

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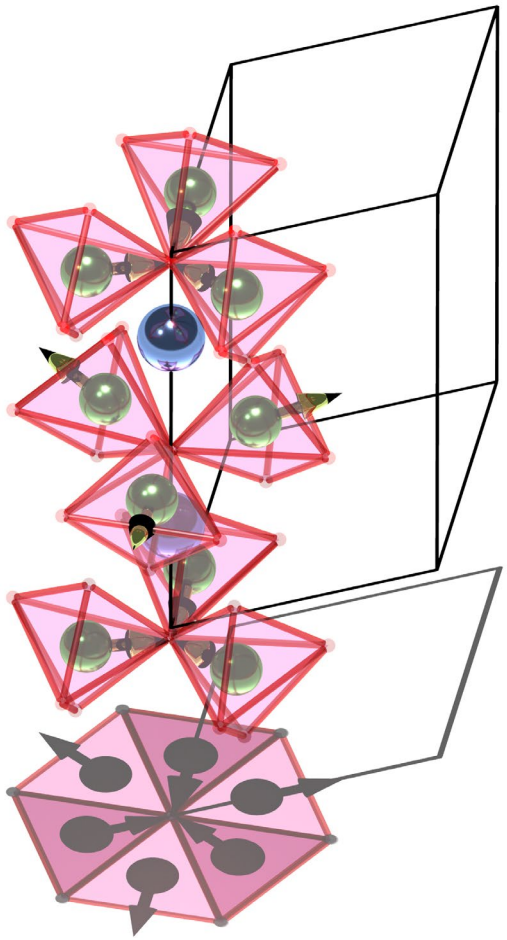

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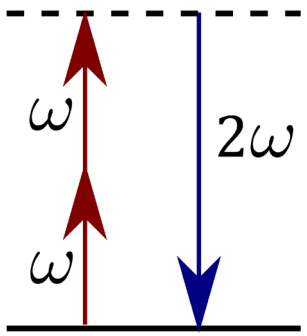
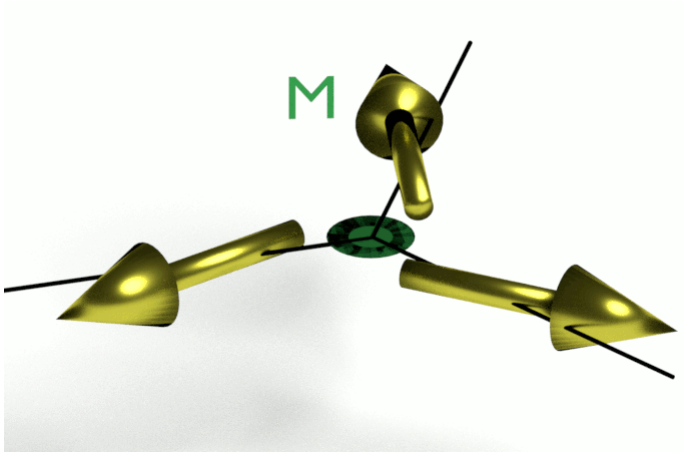
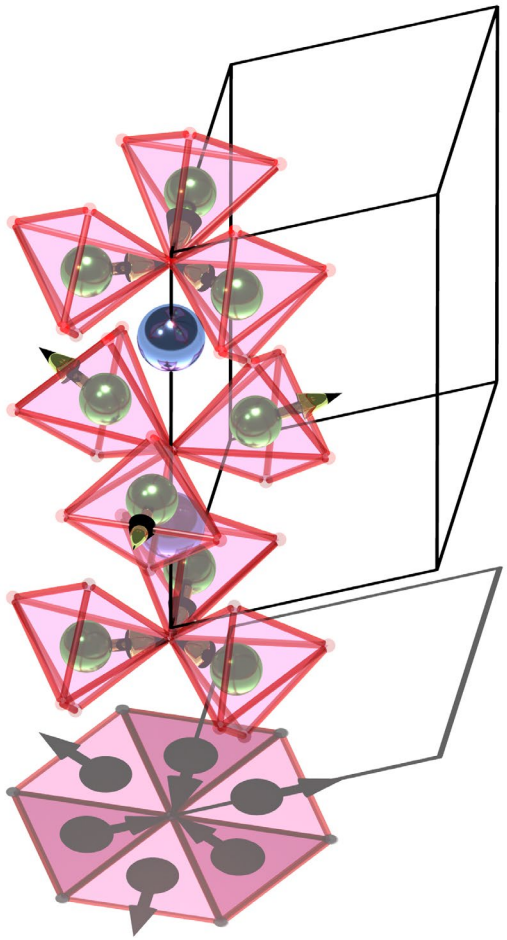




h-$YMnO_3$



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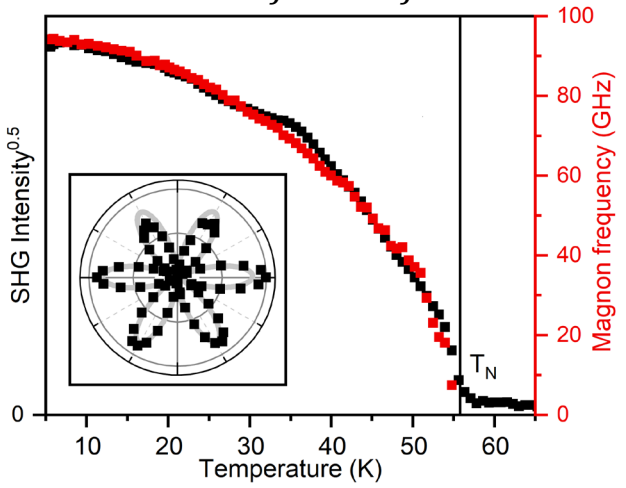
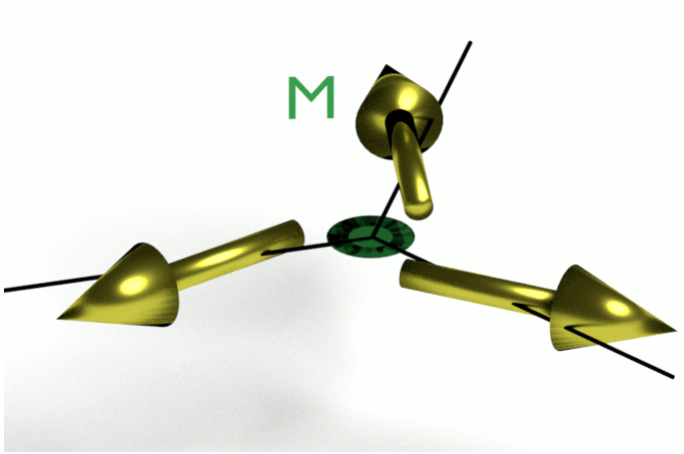
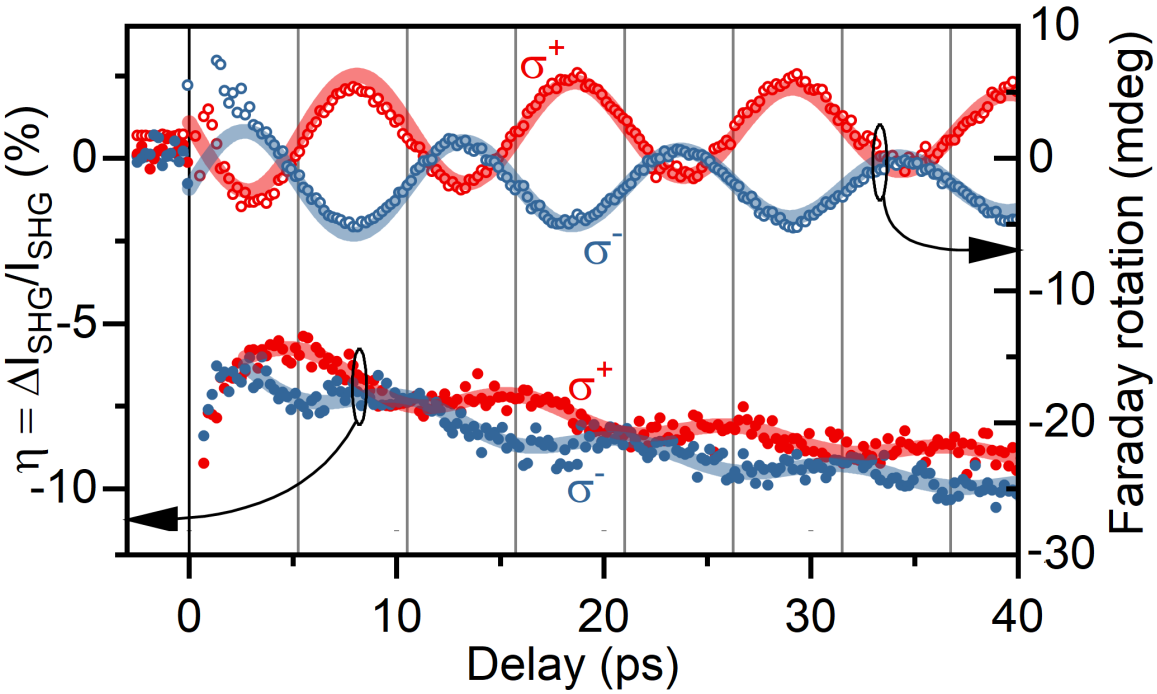




SHG modulation



- Sine-like Faraday response
- Helicity dependent
- Cosine-like SHG modulation

$$P_i(2\omega) = \varepsilon_0 \chi_{ijk}(\boldsymbol{L}) E_j(\omega) E_k(\omega)$$

modulation of $\chi(\boldsymbol{L})$

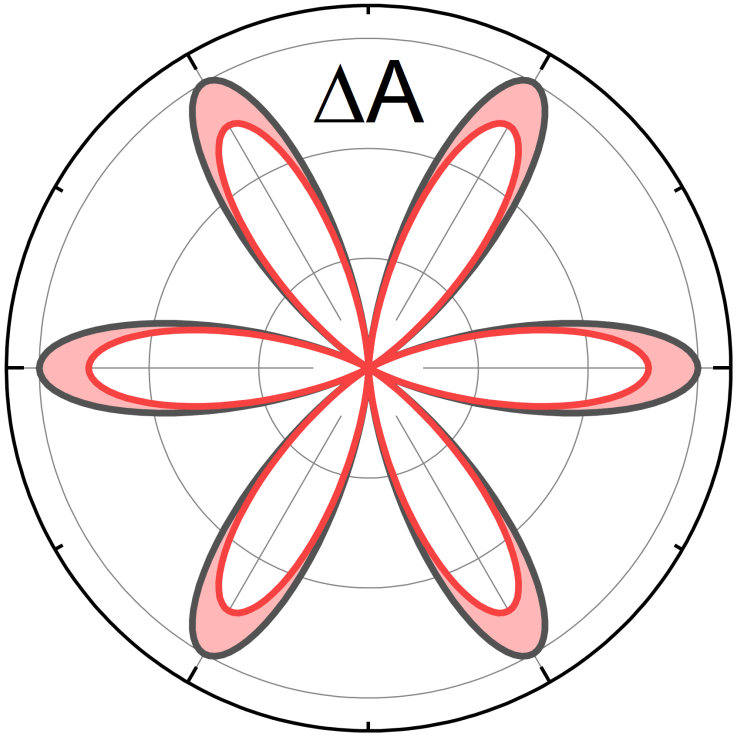
CT et al., Nat. Commun. **10**, 3995 (2019)

SHG modulation

2 scenarios, how **L** can affect χ

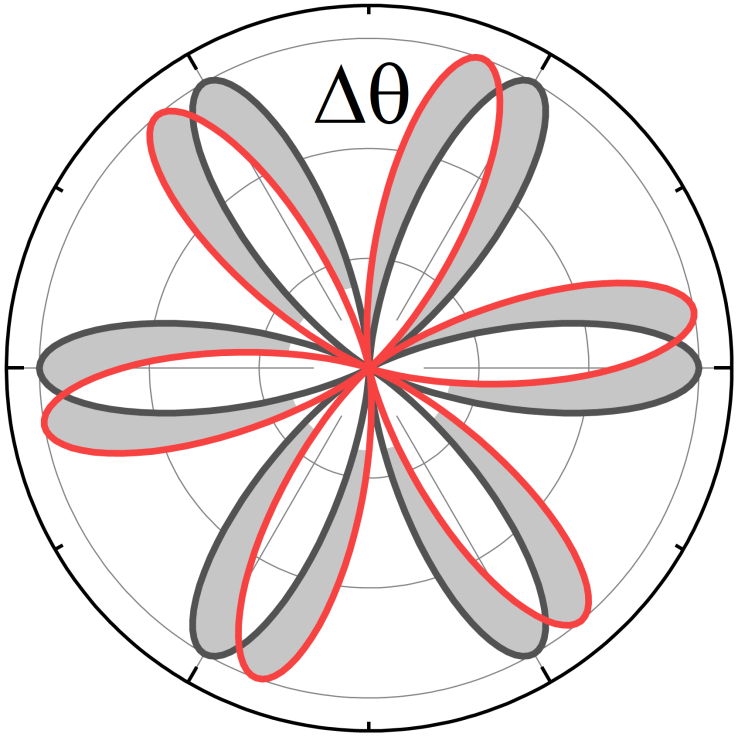
Symmetry-conserving dynamics

$|\chi|$ changes
→ amplitude change of SHG

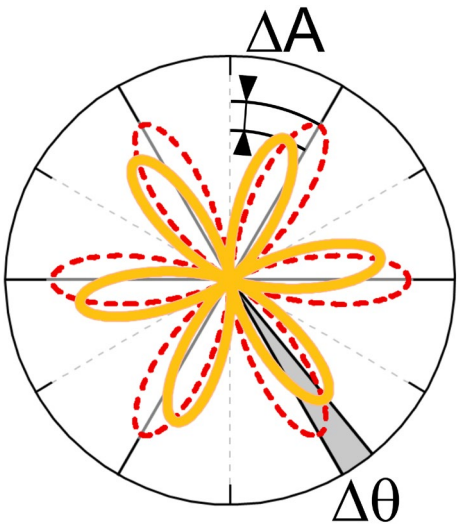


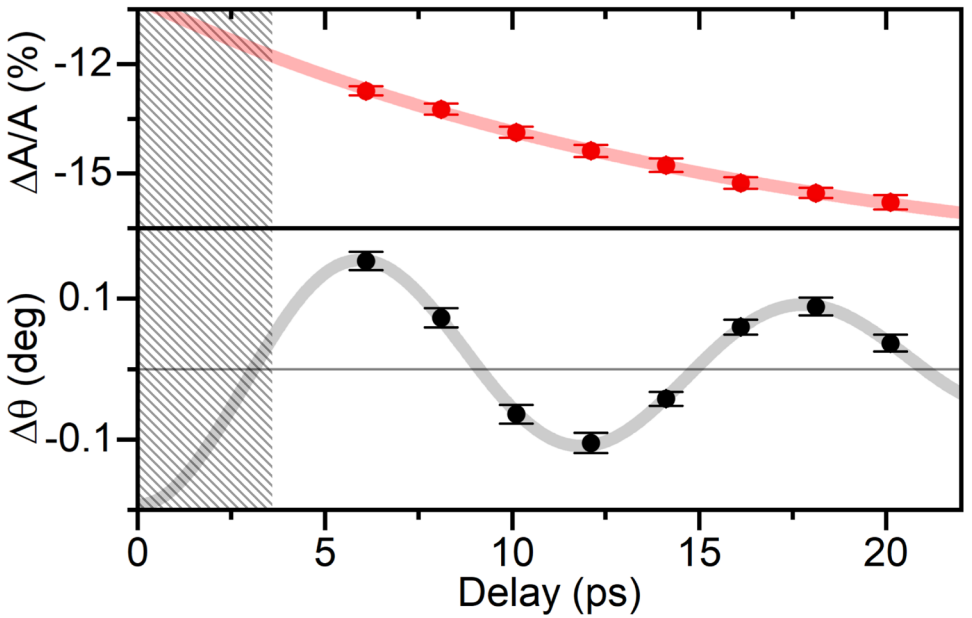
Symmetry-breaking dynamics

New components χ_{ijk}
→ Symmetry change of SHG



Anisotropy modulation

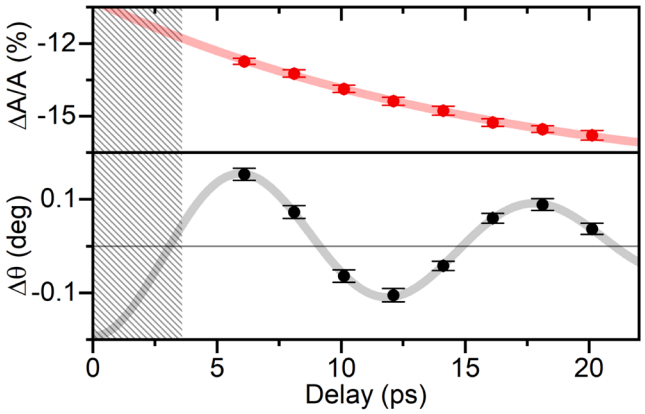




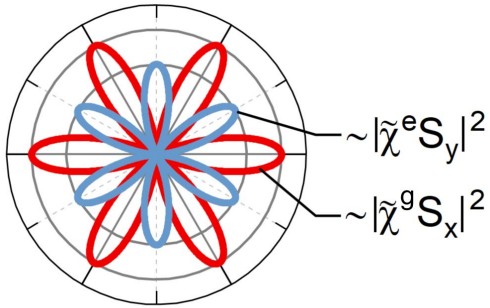
- Amplitude follows exponential decay (→ thermalization)
- Orientation follows cos-like behavior (background free)

CT et al., Nat. Commun. **10**, 3995 (2019)

Anisotropy modulation



Change of anisotropy is interference phenomenon

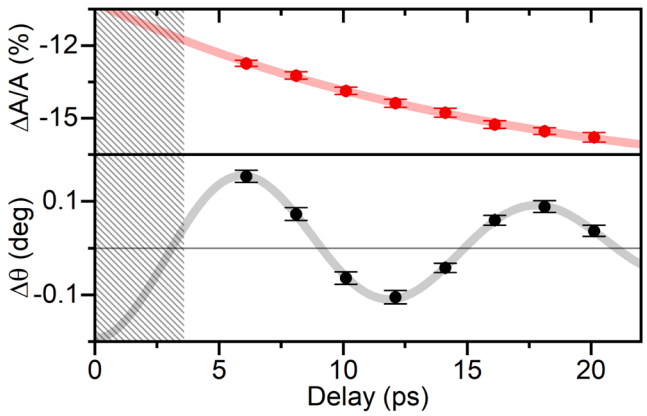


- $\Delta A \sim \Delta|\boldsymbol{S}|$
- $\Delta\theta \sim \alpha \approx S_y/S_x$

Separation of thermal and non-thermal dynamics by their symmetry response

CT et al., Nat. Commun. **10**, 3995 (2019)

Anisotropy modulation



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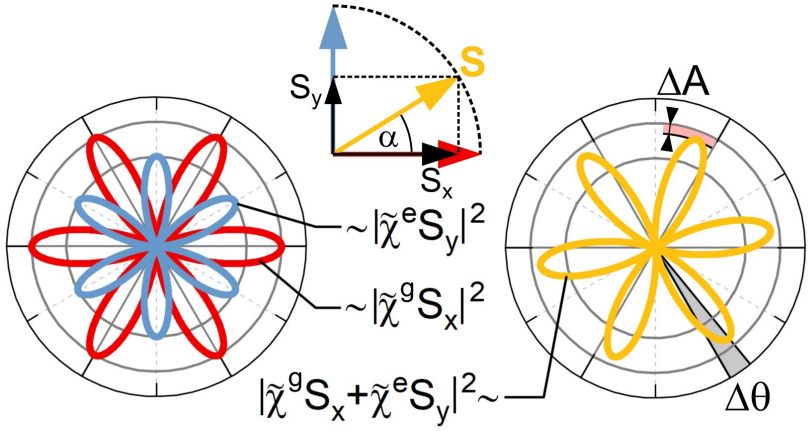

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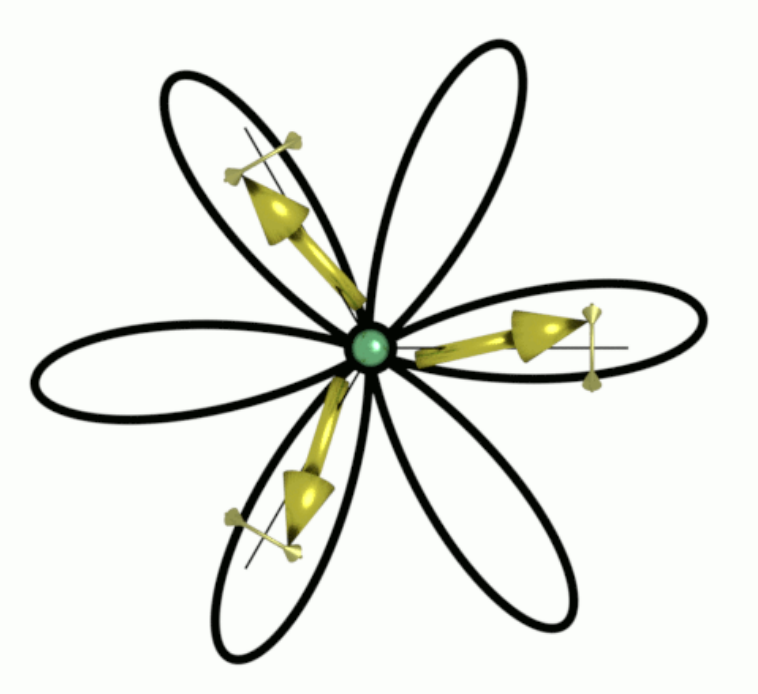
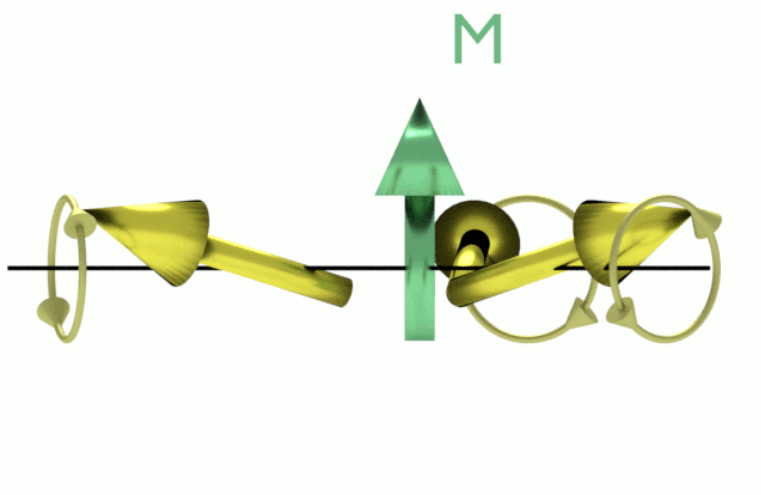
Z-mode reconstruction

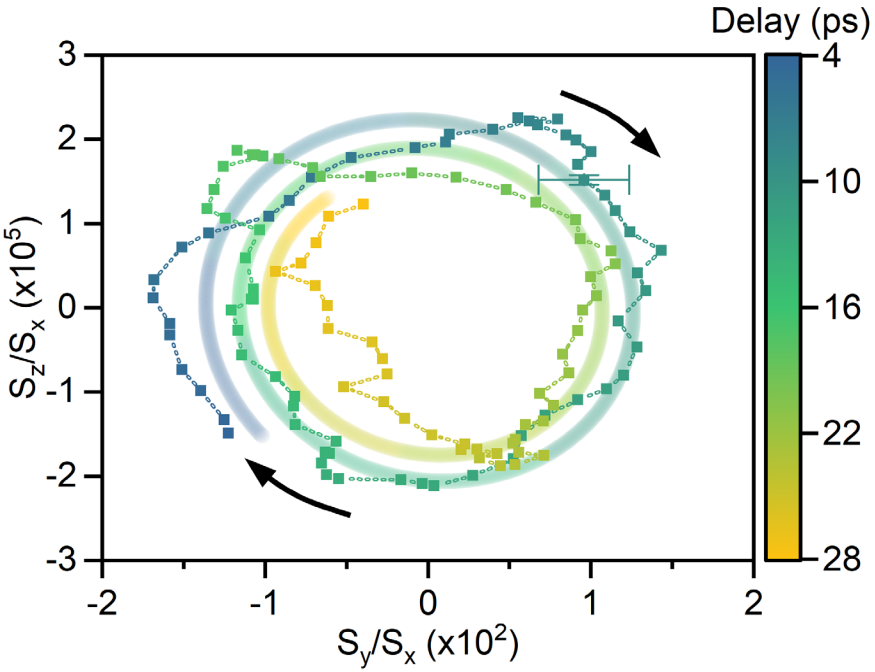
SHG



Faraday rotation



Z-mode reconstruction



- In-plane canting $\alpha \approx 1°$
- Out-of-plane canting 2.4 mdeg
- Strong ellipticity is general property of AFMs

CT et al., Nat. Commun. **10**, 3995 (2019)

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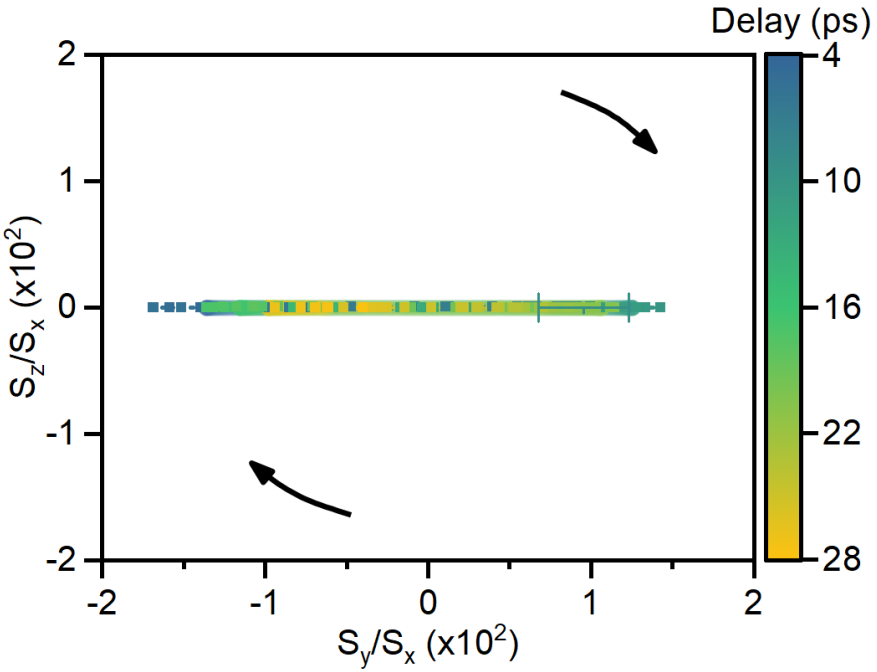

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Three questions

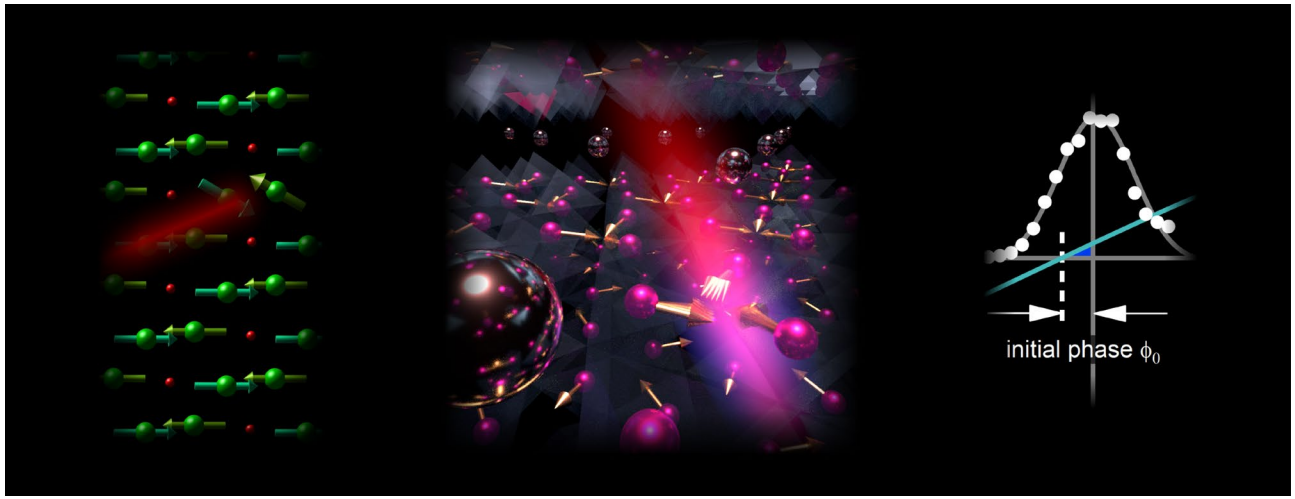
- How can we excite antiferromagnets?

  → Inverse magneto-optical effects (PRB 95, 174407 (2017), Nat. Commun. **11**, 6142 (2020))

- How can we probe an AFM order parameter?

  → Time-resolved SHG (CT et al., Nat Commun. 10, 3995 (2019))

- What's next?



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A perspective on nonlinearities in coherent magnetization dynamics

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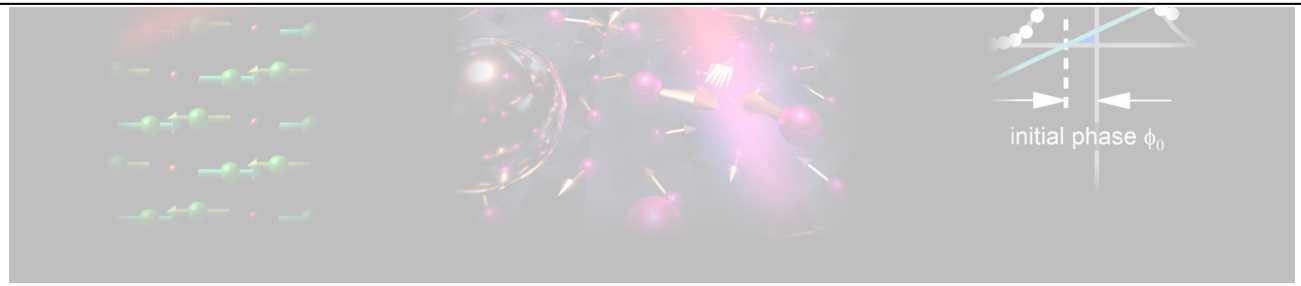
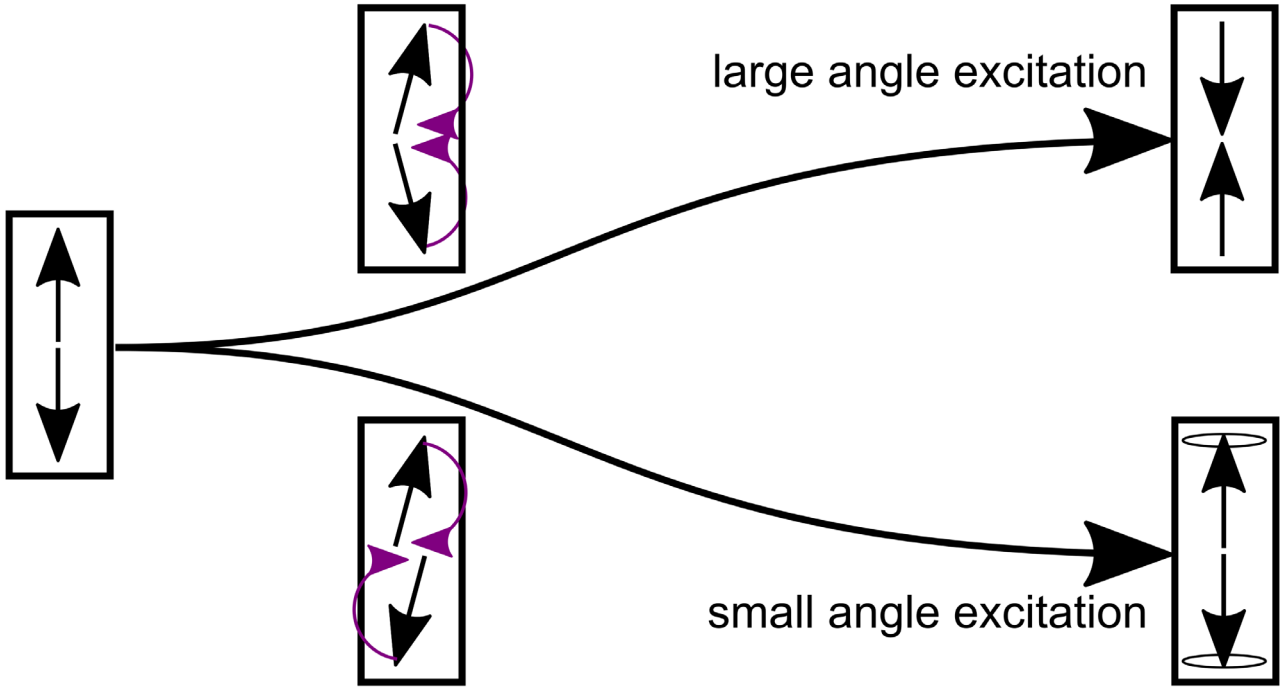
Jingwen Li,[1] Chia-Jung Yang,[1] Ritwik Mondal,[2,3,a)] Christian Tzschaschel,[4,a)] and Shovon Pal[5,a)]



AFM switching



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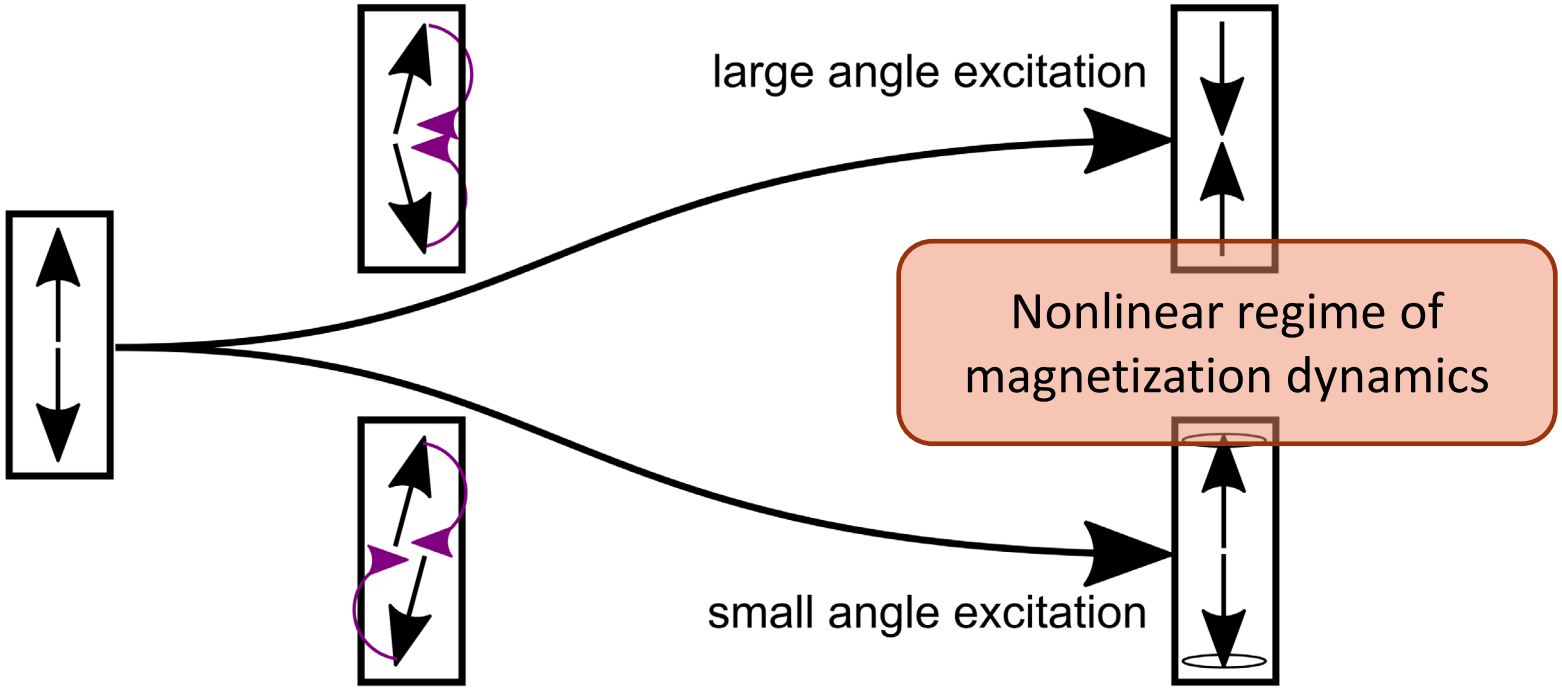
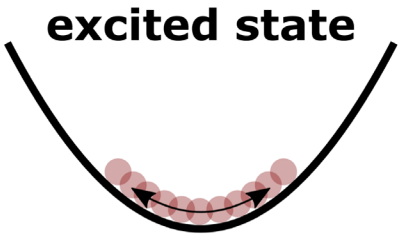


Nonlinear Magnetization Dynamics



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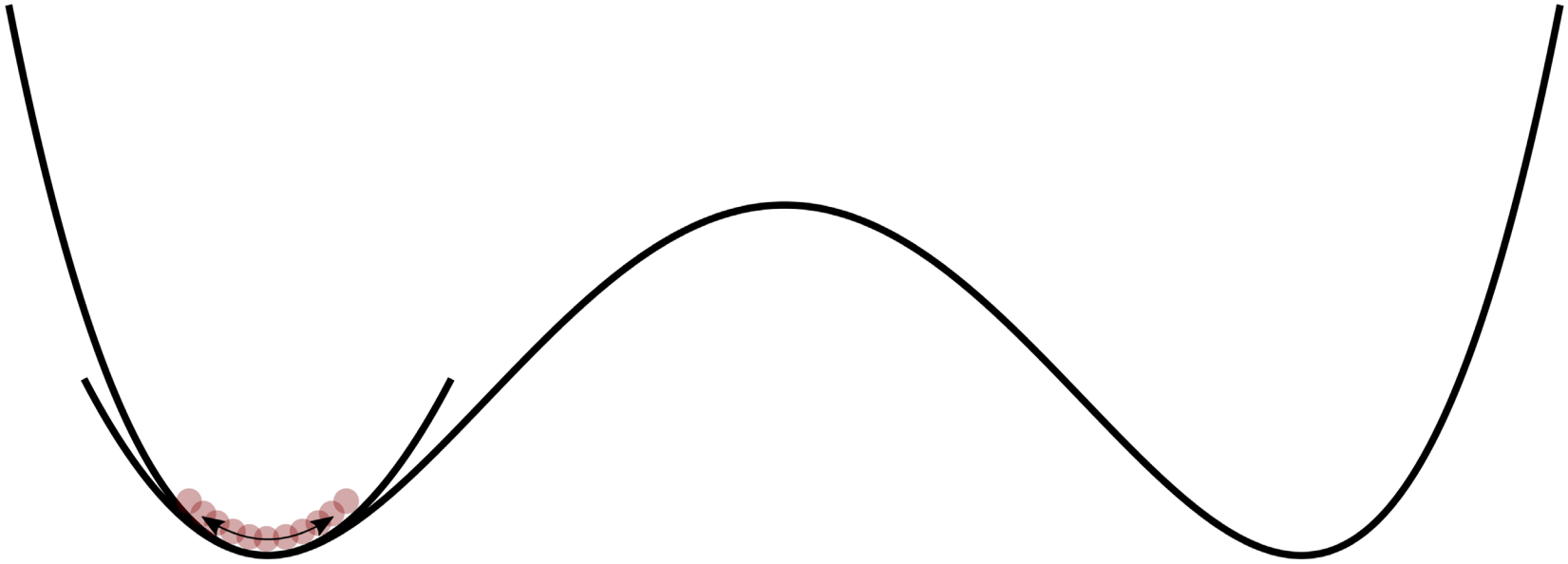

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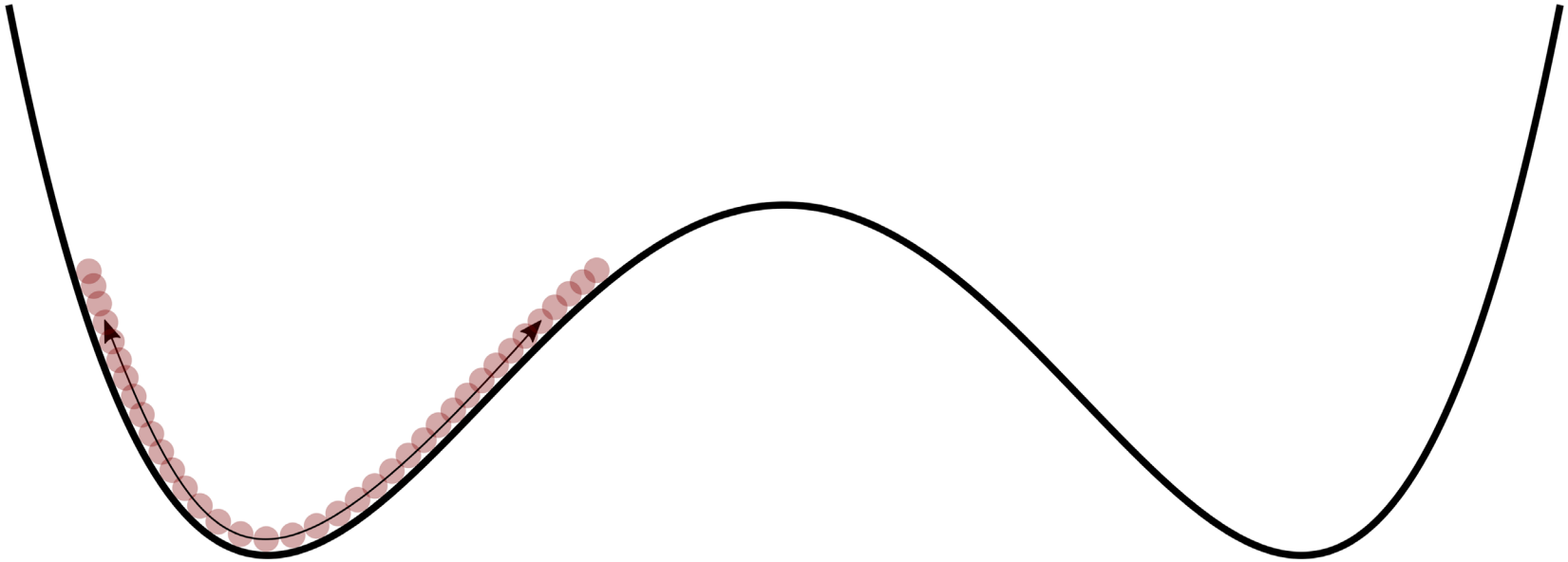

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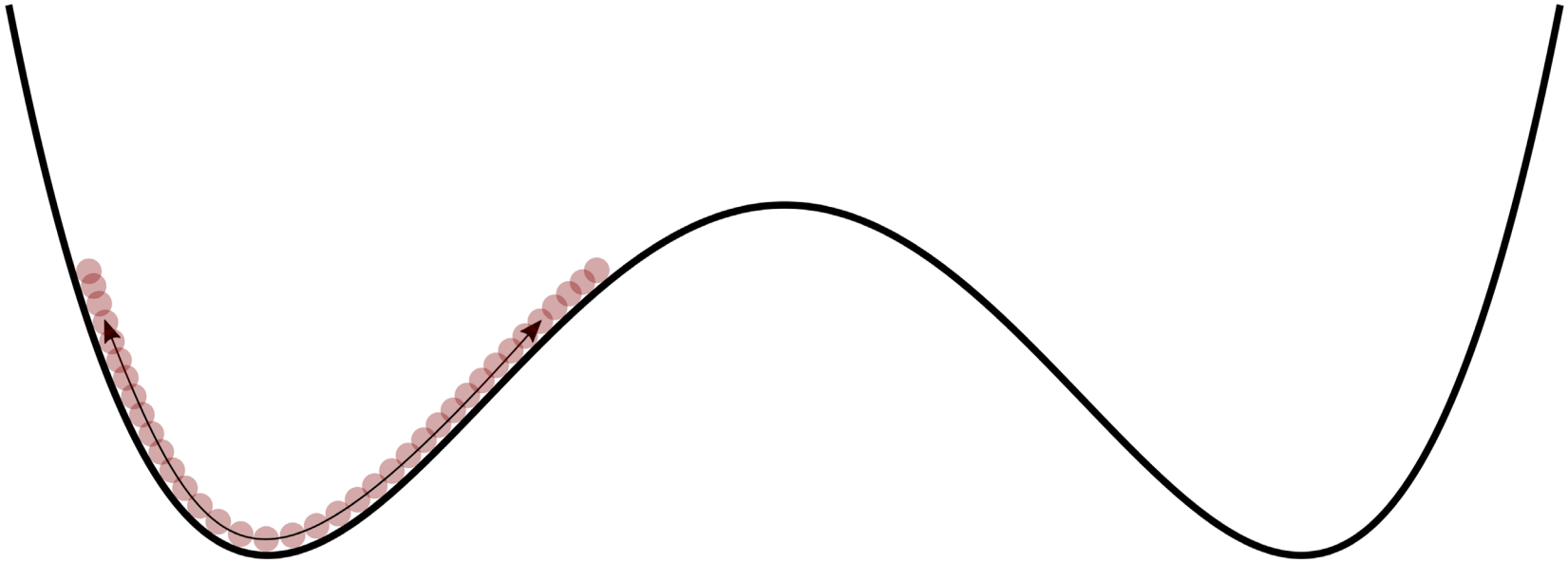
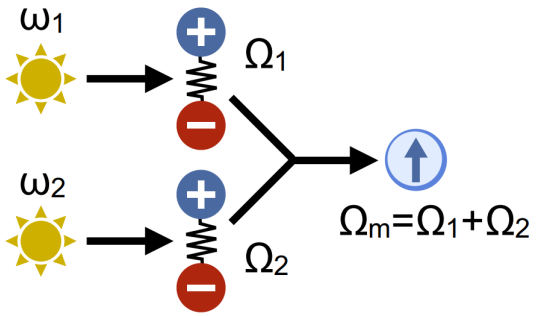


Understanding nonlinear magnetization dynamics advances and accelerates the development of ways for switching magnetization

Jingwen Li, CT et al., Appl. Phys. Lett. **120,** 050501 (2022)

Nonlinear Antiferromagnetic Dynamics

Stronger



Phonomagnetism

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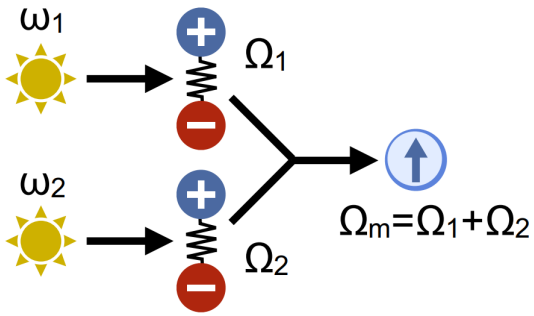

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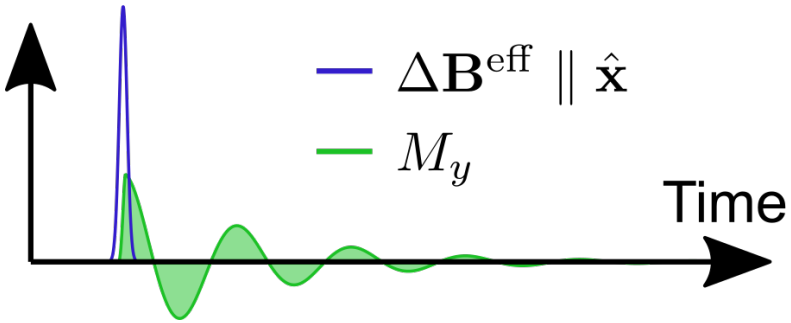

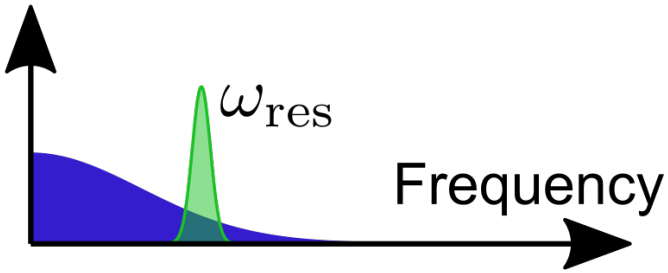

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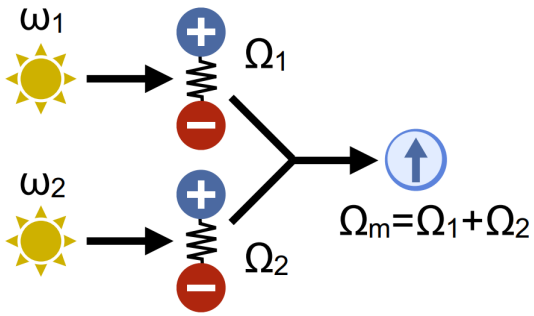

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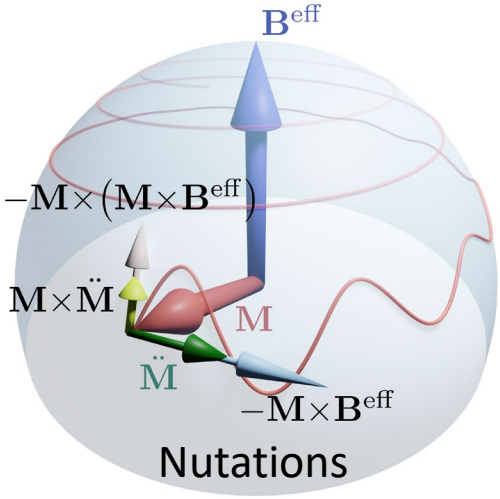
Faster

$$\frac{\partial \boldsymbol{M}}{\partial t} = -\gamma \boldsymbol{M} \times \boldsymbol{B}_{eff} - \gamma\alpha \frac{\boldsymbol{M}}{|\boldsymbol{M}|} \times (\boldsymbol{M} \times \boldsymbol{B}_{eff}) - \gamma \frac{\eta}{|\boldsymbol{M}|} (\boldsymbol{M} \times \ddot{\boldsymbol{M}})$$



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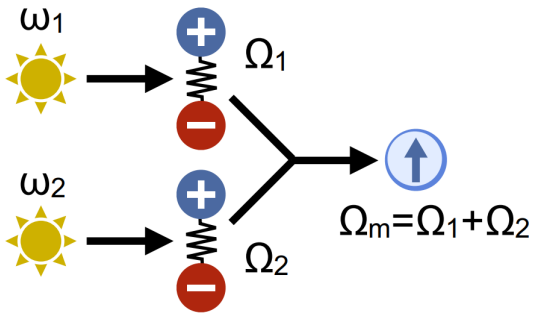

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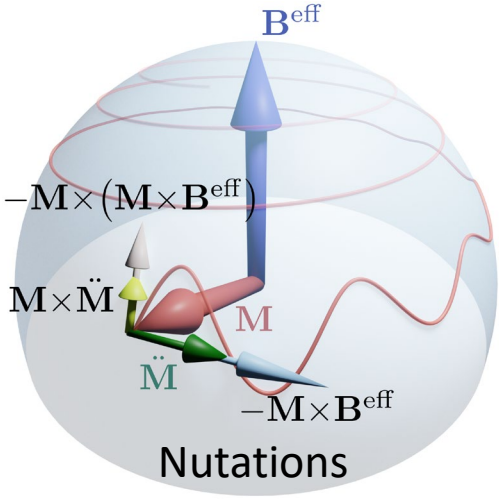
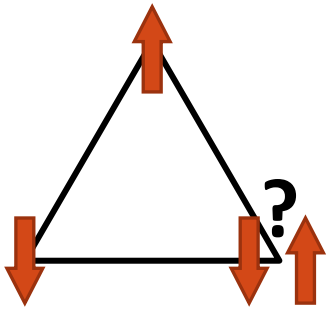
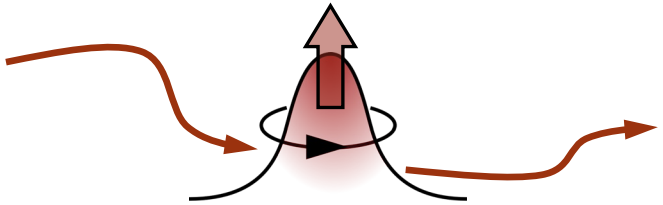


Weirder?



Frustrated magnetism



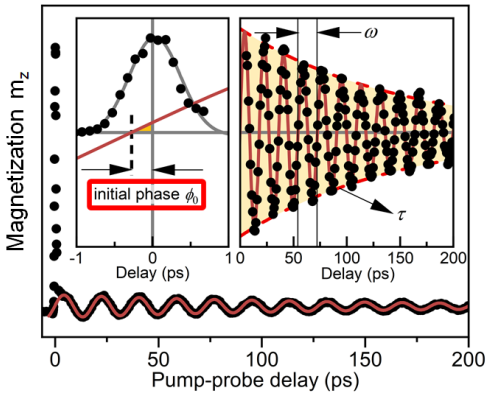
Berry curvature

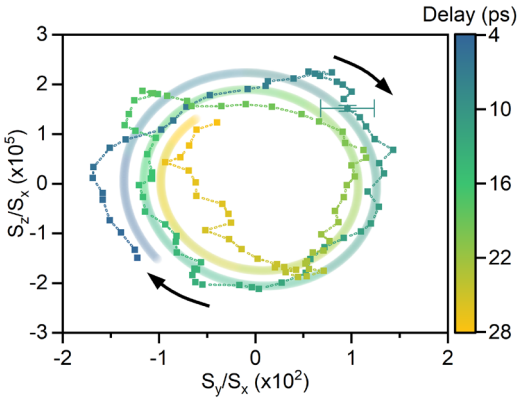
Jingwen Li, CT et al., Appl. Phys. Lett. **120,** 050501 (2022)

Conclusion

Ultrafast laser pulses can efficiently excite spin dynamics in AFMs

- Initial phase contains valuable information about excitation mechanism
- Strong ellipticity of spin precession facilitates new excitation pathways unavailable to FMs



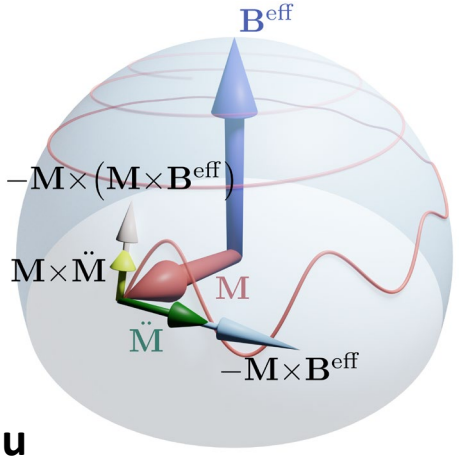


Nonlinear optics provide powerful probe for AFM spin dynamics

- Combination of linear and nonlinear optics enables full reconstruction and tracking of AFM order parameter

Fundamentally new magnetization dynamics

- Nonlinear antiferromagnetic dynamics
- Emerging concepts such as Topology/Frustration



email: ctzschaschel@fas.harvard.edu