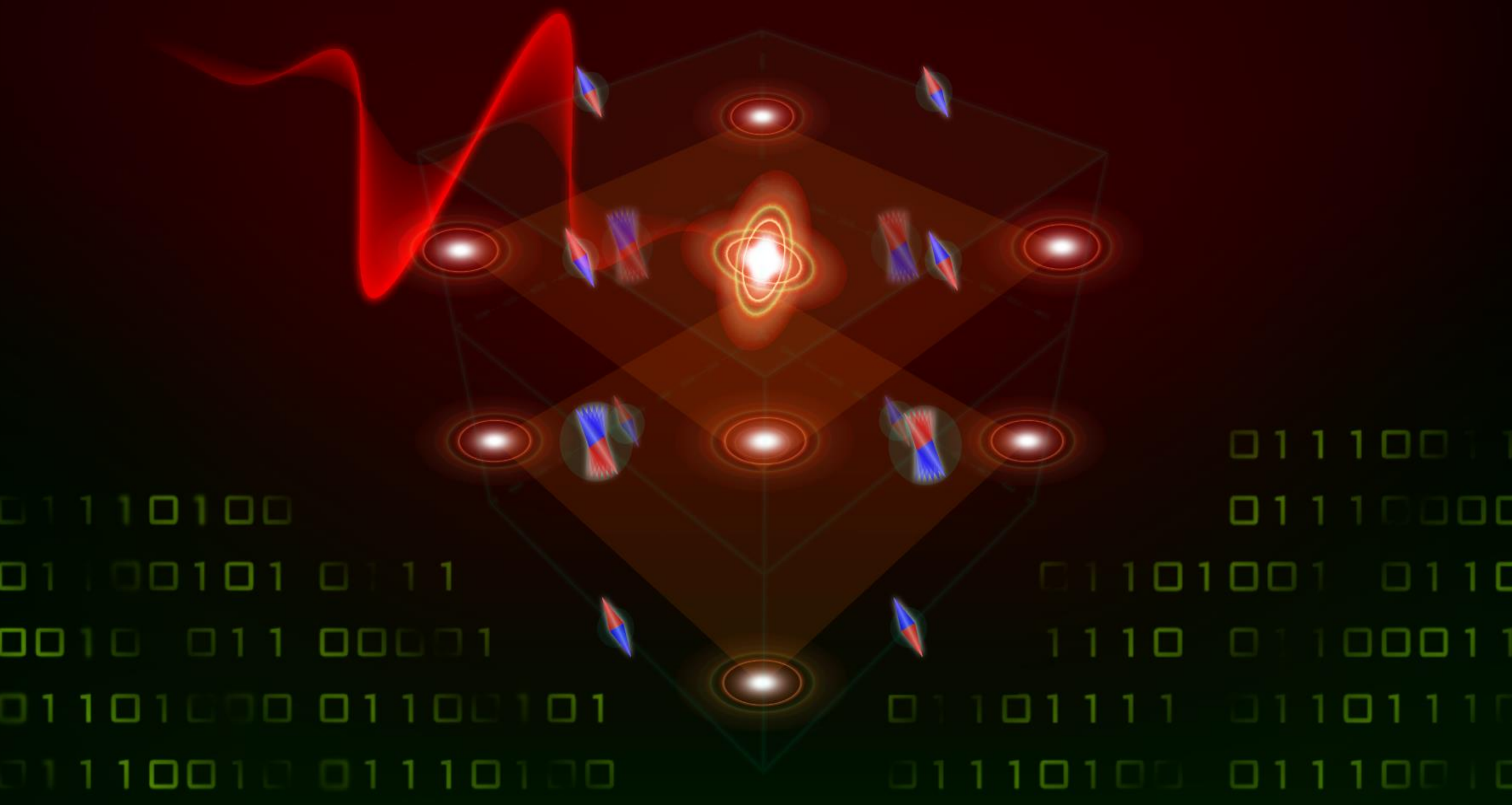
Terahertz Nonlinear Spin Control

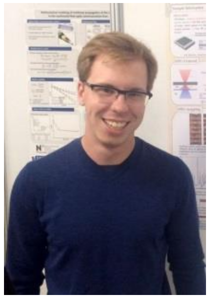
Rostislav Mikhaylovskiy



Acknowledgements

Radboud University, Netherlands





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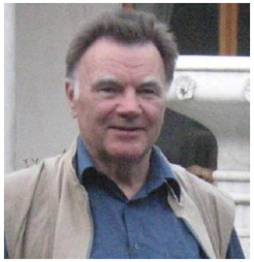
Moscow Physical-Technical Institute



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erc

SEVENTH FRAMEWORK PROGRAMME

FOM

Outline

- Introduction: why THz?

- THz linear spin control
  - Magnon-polaritons
  - Internal resonance

- THz nonlinear spin control
  - THz-driven anisotropy fields
  - Towards THz spin switching

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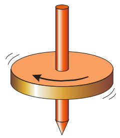
Magnetism is fast

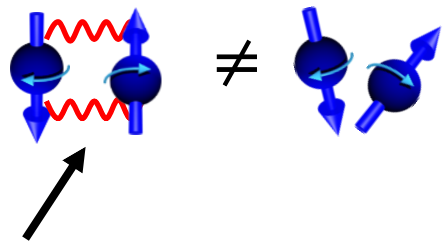
Magnetism – strongest quantum effect

$$W_{ex} = J\mathbf{S}_i\mathbf{S}_j \sim 10\,\mathrm{meV}$$



Spin

$$S_z = \pm\hbar/2$$



exchange interaction

$$\mathbf{B}_{ex} = \frac{\partial W_{ex}}{\partial \mathbf{S}_i} \sim 10^2 - 10^3\,\mathrm{T}$$

Femtosecond intrinsic scale

$$\tau_{ex} = \frac{eB_{ex}}{m_e c} \sim 100\,\mathrm{fs}$$

Magnetism is fast

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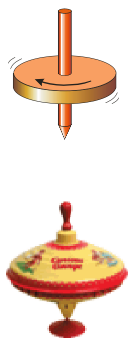

Spin

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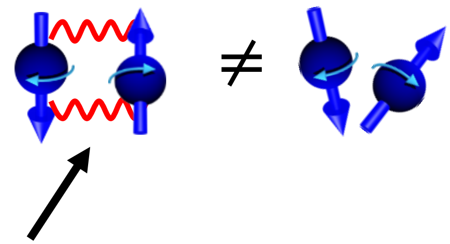

exchange interaction

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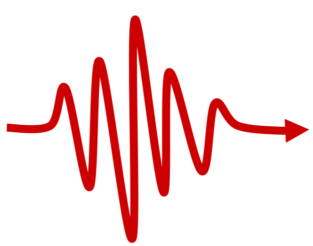
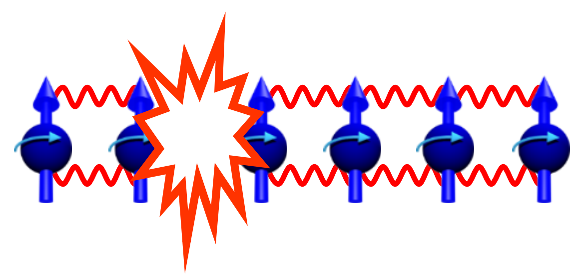
Femtosecond intrinsic scale

$$\tau_{\text{ex}} = \frac{eB_{\text{ex}}}{m_e c} \sim 100\,\text{fs}$$

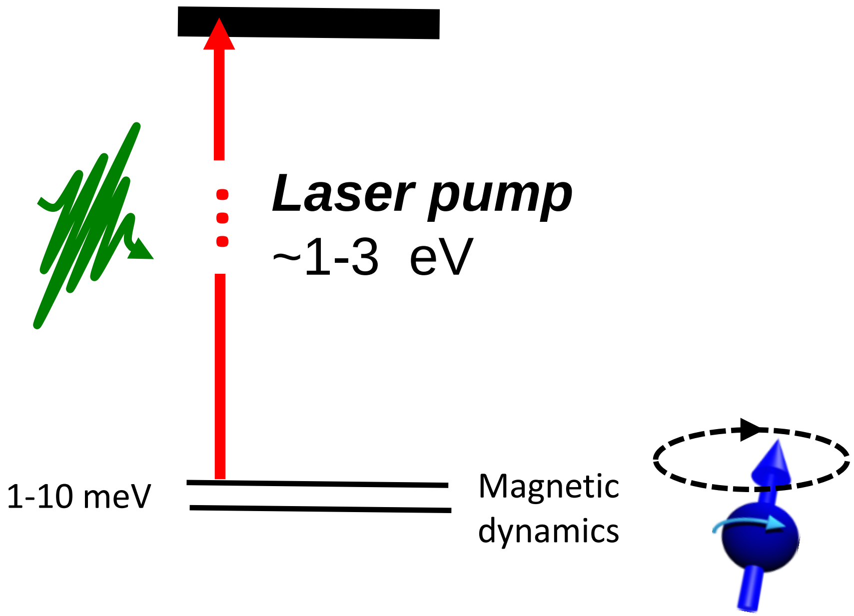
Ultrashort laser stimulus

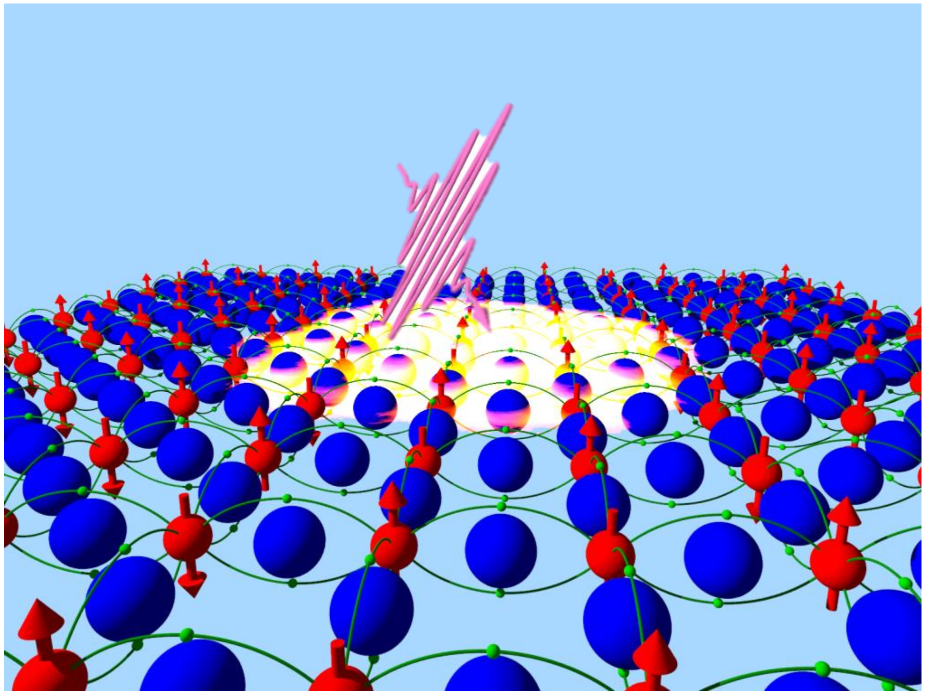
100 fs ⟷ 10 THz = 10^{13} Hz





Too much energy for optical excitation?



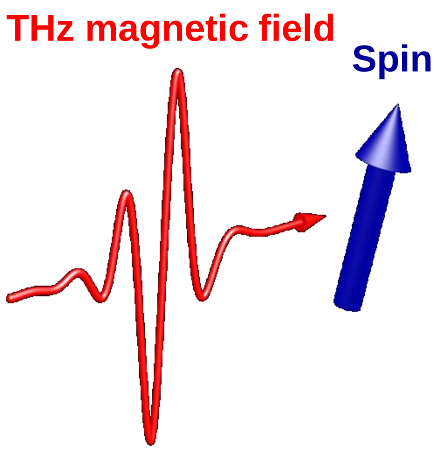


Can we excite magnetic order on its own energy scale and ultrafast?

Problem with laser excitation:

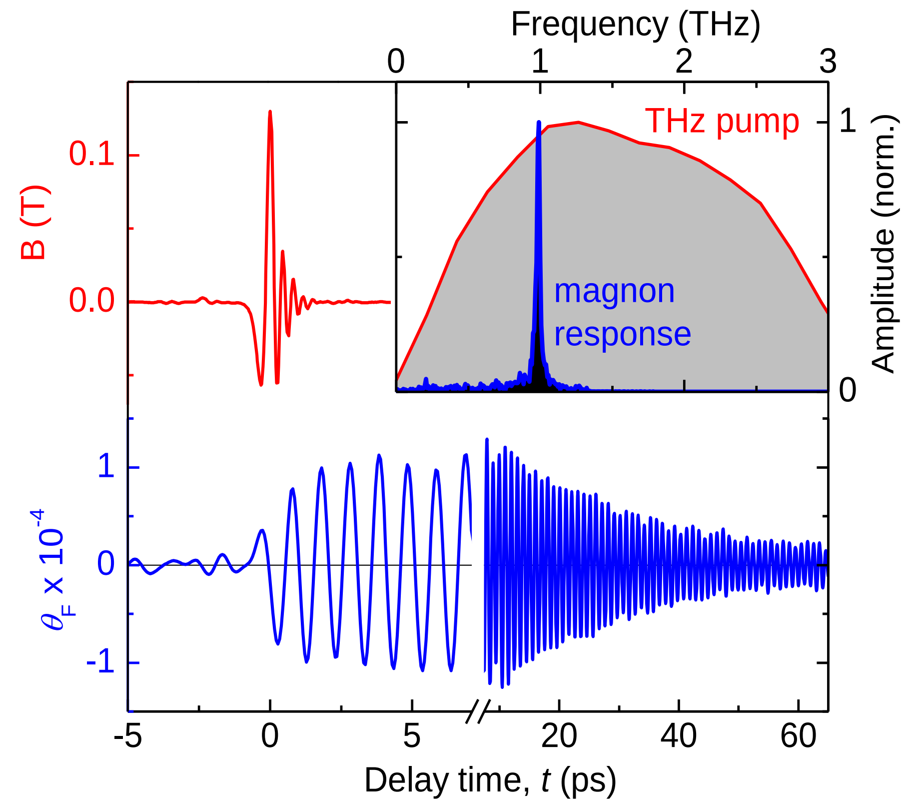
Photon energy exceeds magnetic energy scale

THz-field control of spins



$$\frac{\partial \mathbf{S}}{\partial t} = \gamma[\mathbf{S} \times \mathbf{B}_{\mathrm{THz}}]$$

Direct ultrafast interface to spins



T. Kampfrath, et al., Nature Photonics ***5****, 31 (2011)*

Only linear response

Small spin deflection

Outline

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- THz nonlinear spin control
  - THz magneto-phonics
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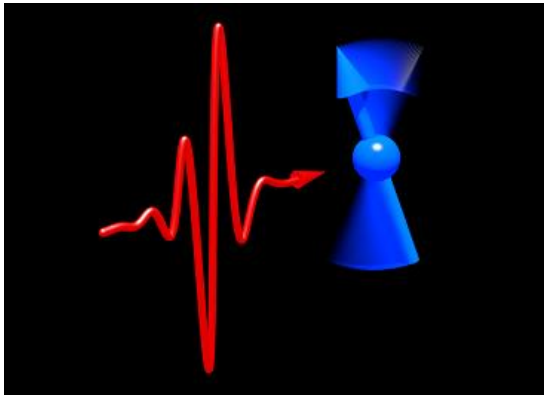
Magnonics or polaritonics?

THz frequency dipole active excitation

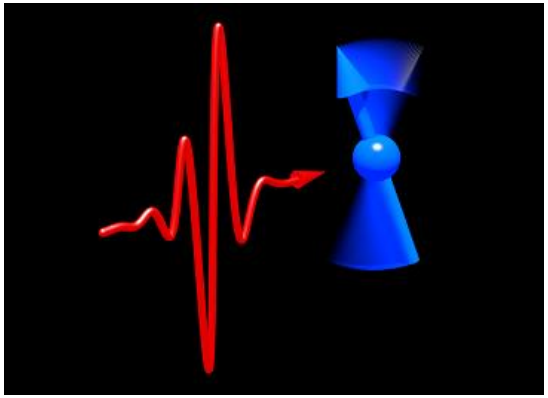


Emission power grows with dipole frequecy ~ω^2

THz magnons or …?

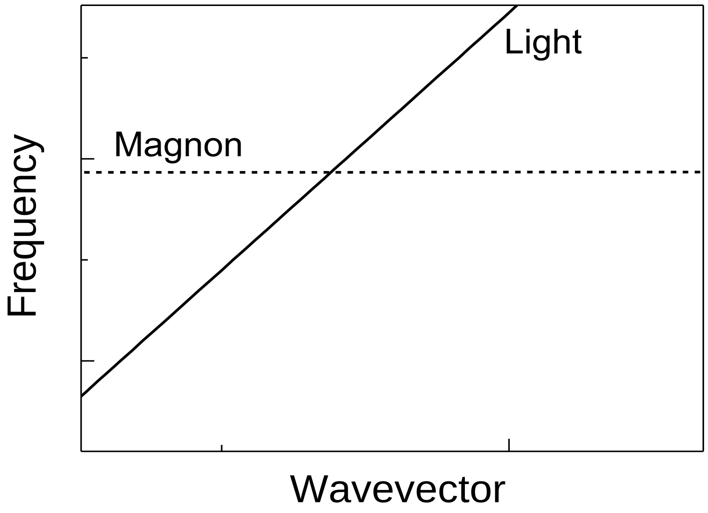
Magnonics or polaritonics?

THz frequency dipole active excitation



Emission power grows with dipole frequecy ~ω^2

Strong light-magnon coupling



THz magnons … - polaritons

Magnonics or polaritonics?

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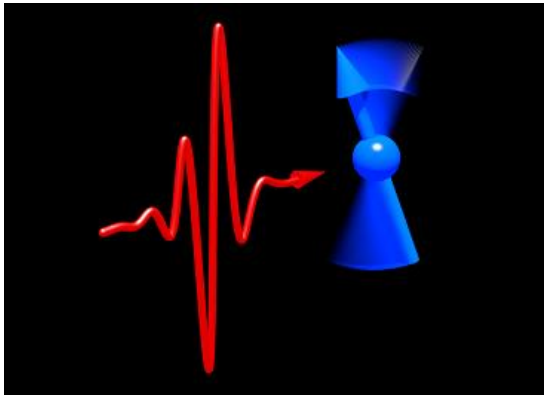

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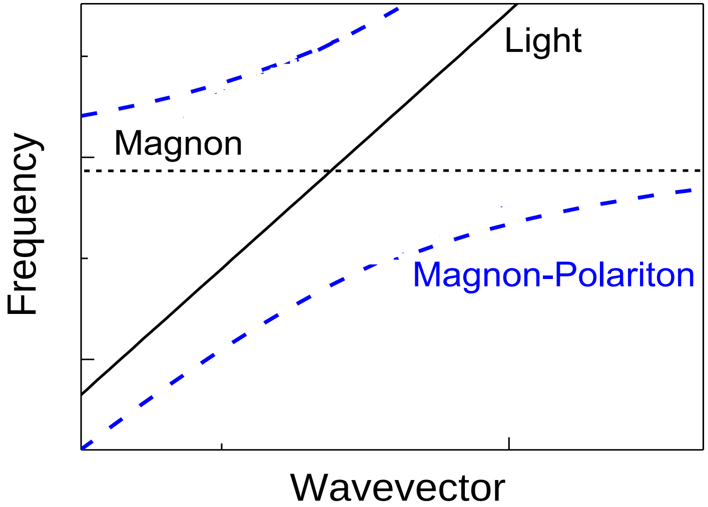

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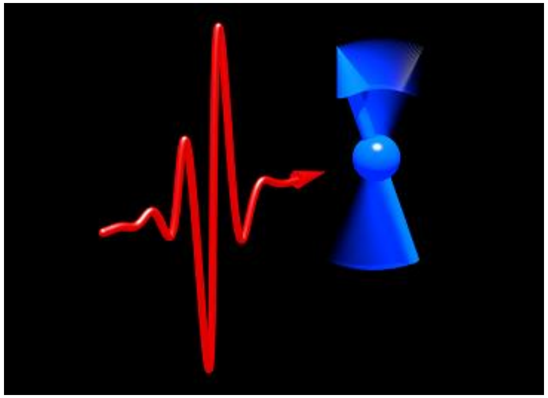

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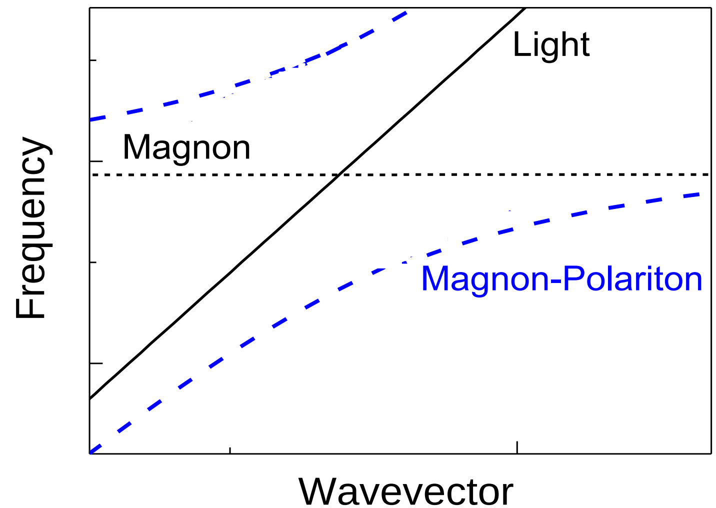
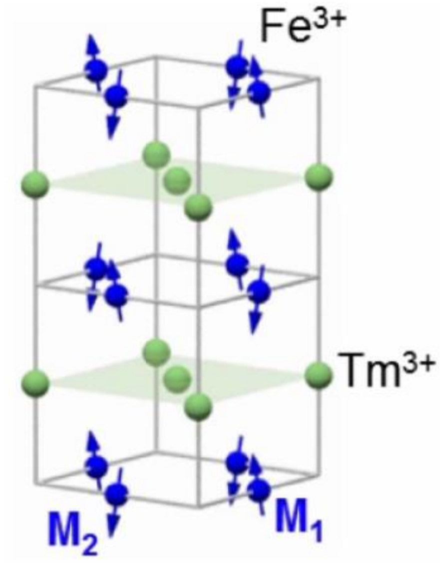


THz magnons … - polaritons

Do we see magnon-polariton signatures in experiment?

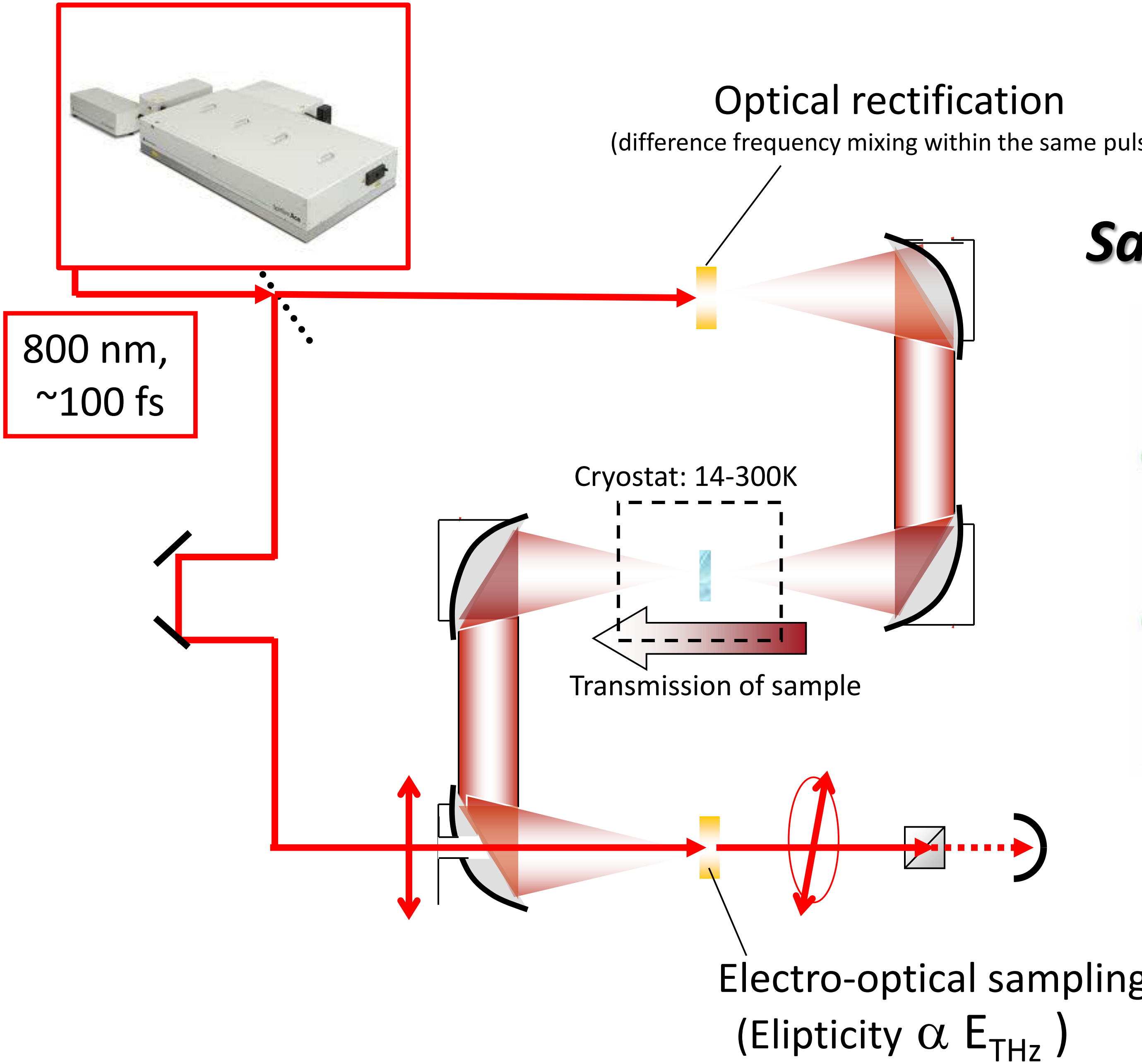
THz time-domain spectroscopy

Sample $TmFeO_3$

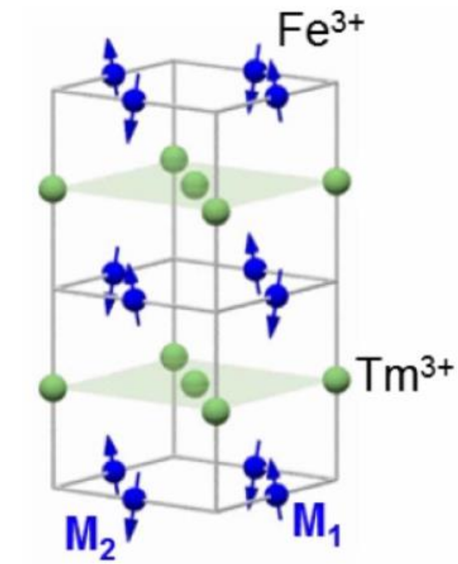


Magnon 0.8 THz

THz time-domain spectroscopy

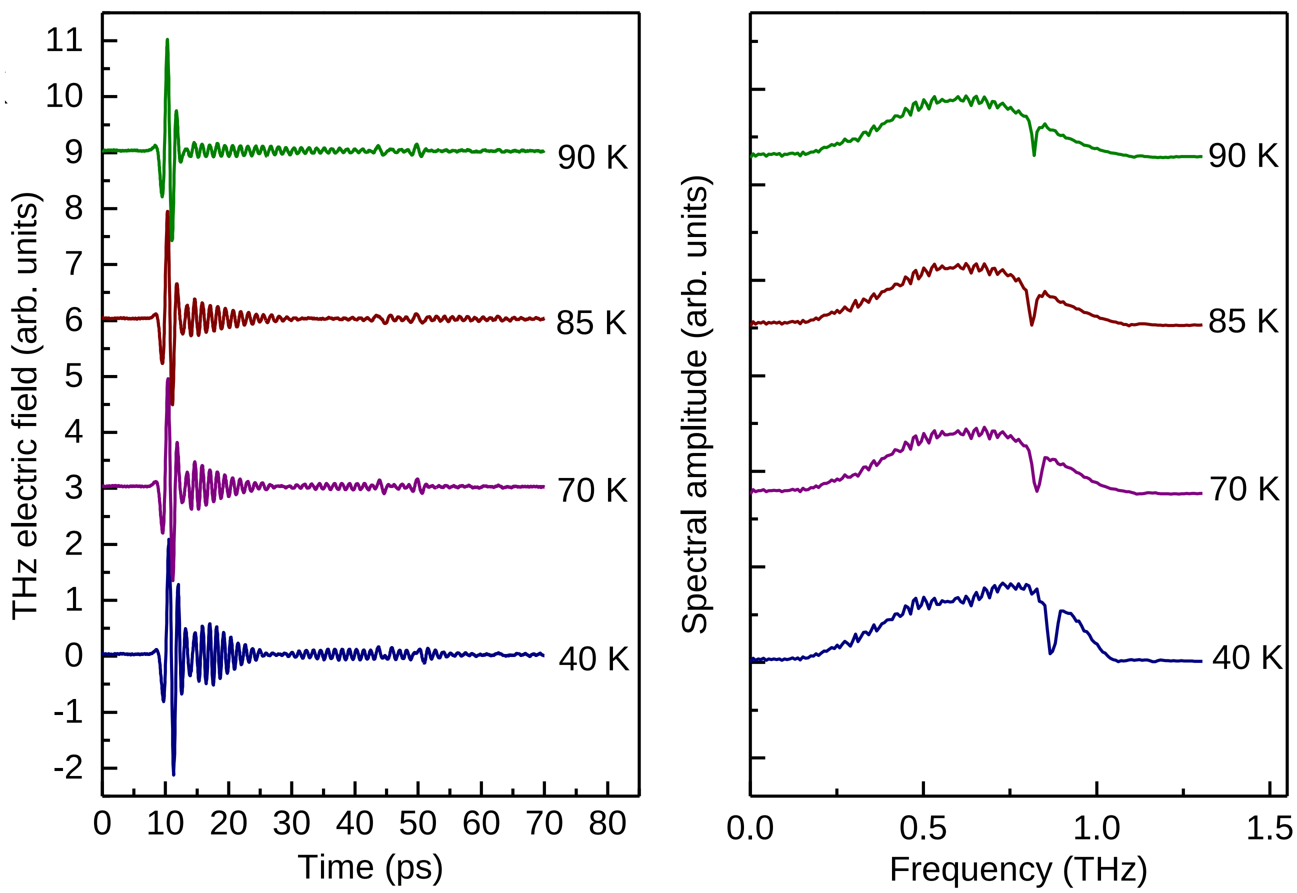


Sample $TmFeO_3$



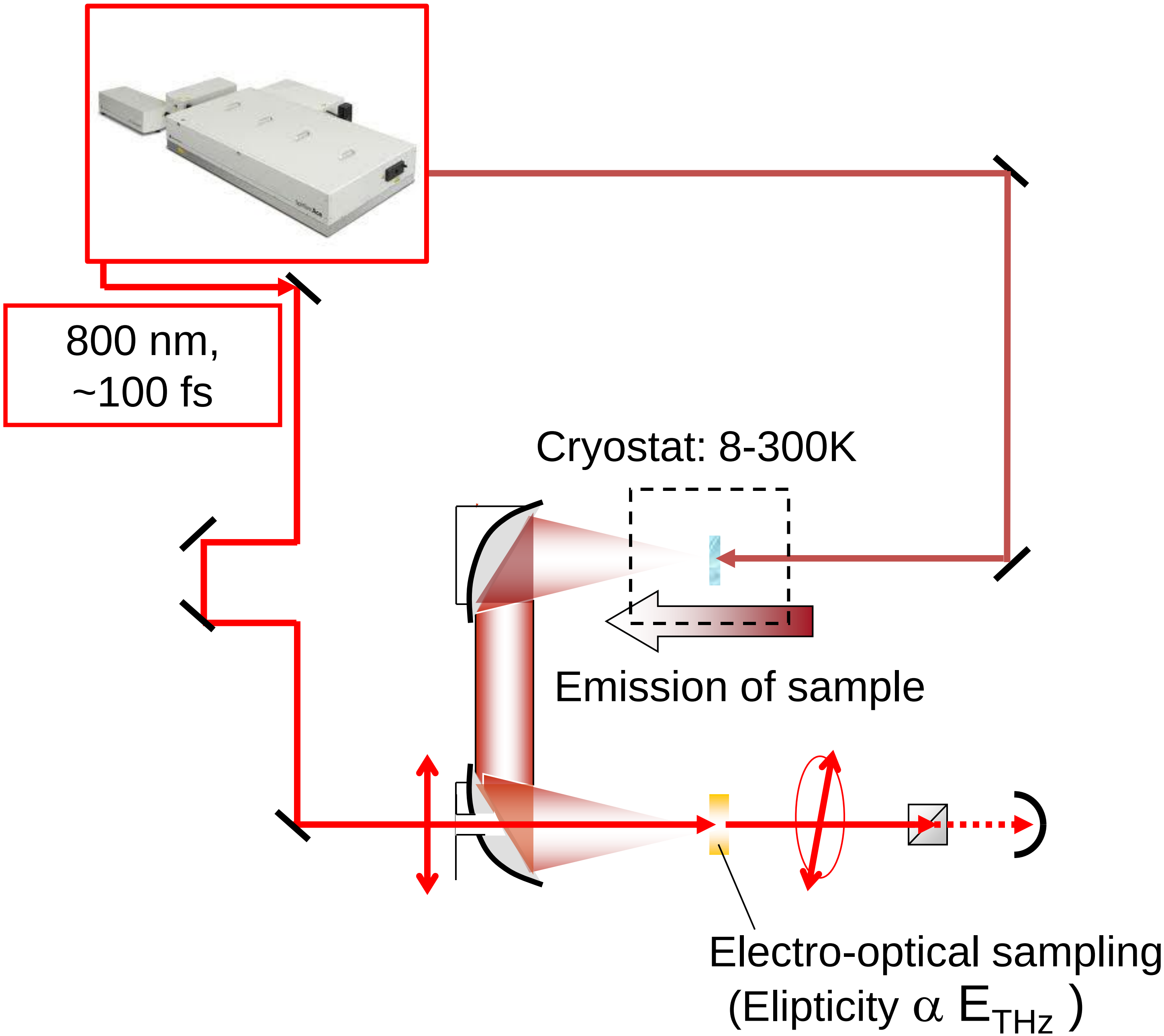
Magnon 0.8 THz

THz transmission through $TmFeO_3$

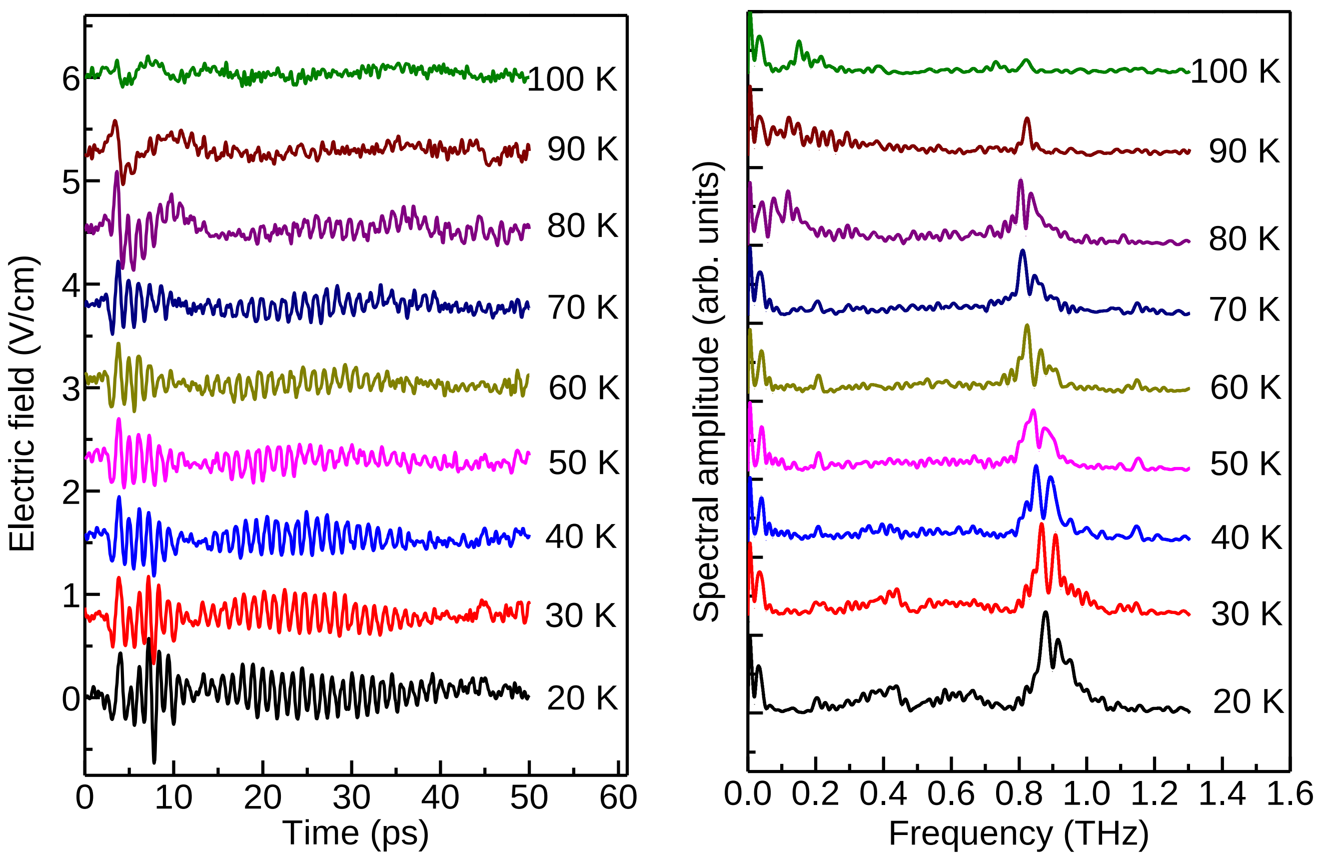


Clear beating in time domain waveforms

THz emission spectroscopy



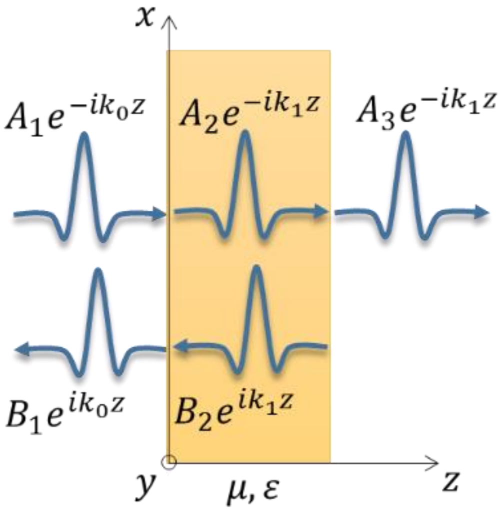
THz emission from $TmFeO_3$

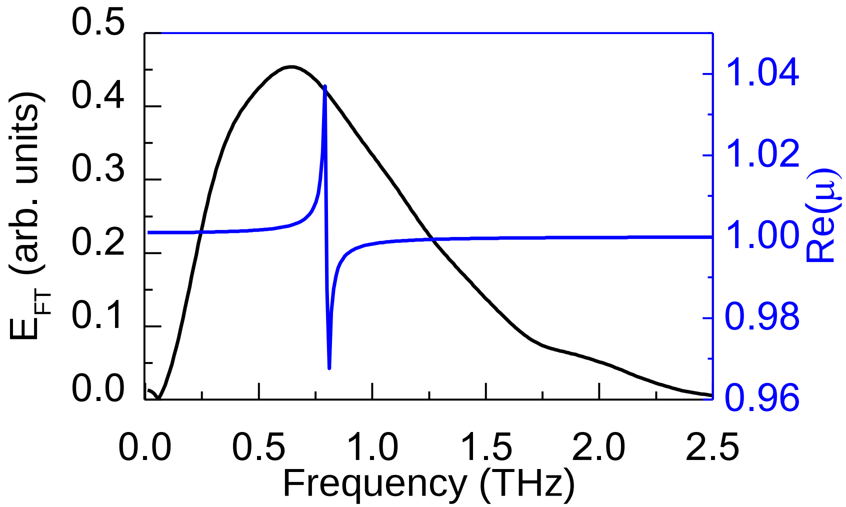


Again beating instead of single magnon frequency

Terahertz transmission analysis

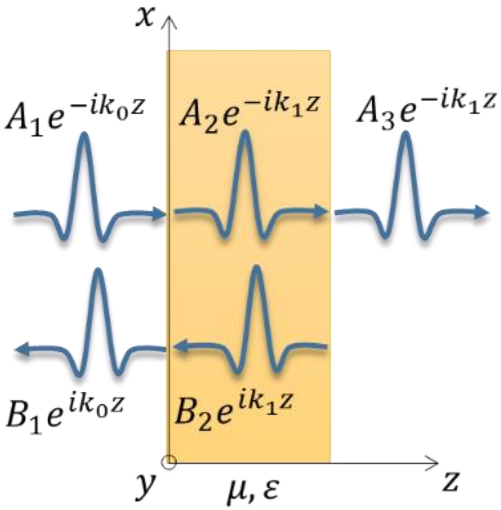
Solve Maxwell equations



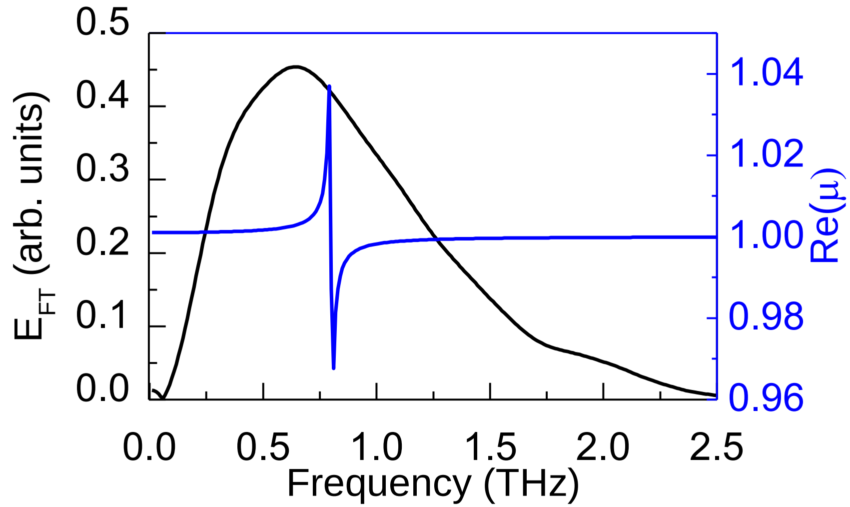


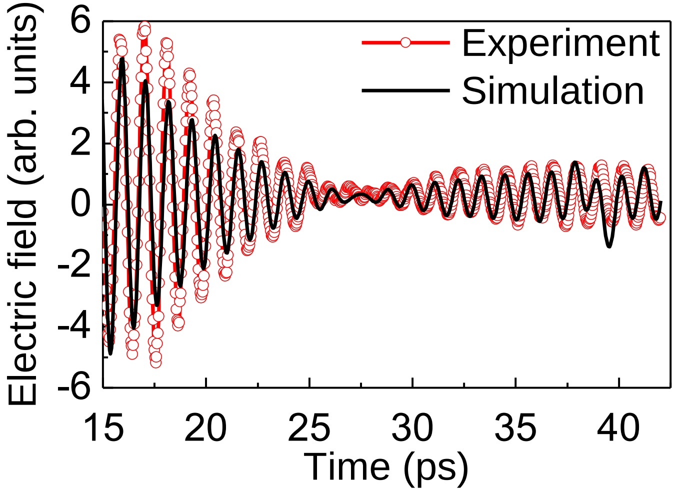
Terahertz transmission analysis

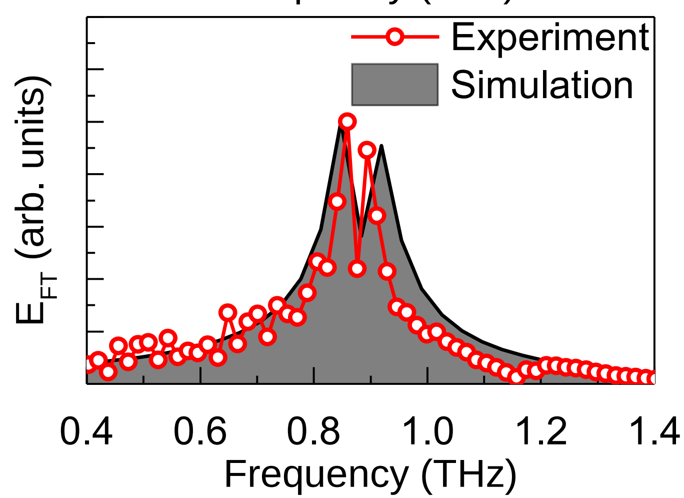
Solve Maxwell equations



No fitting parameters!





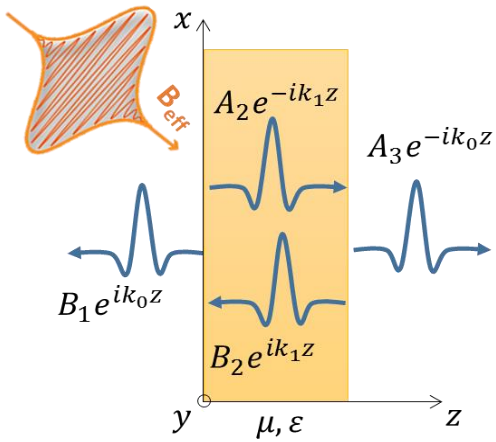


Terahertz emission analysis

Solve Maxwell equations

Light acts as effective magnetic field

R. Mikhaylovskiy, et al. Nature Comm. 6, 8190 (2015)



Terahertz emission analysis

Solve Maxwell equations

Fit effective field

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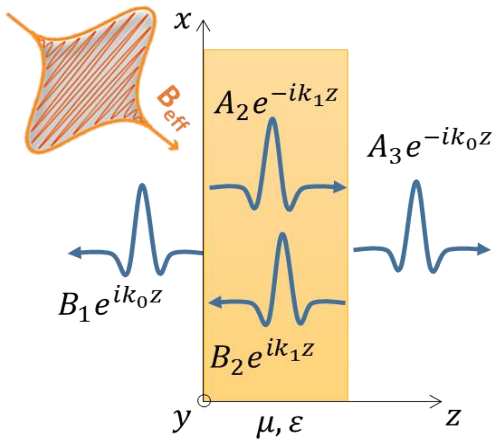
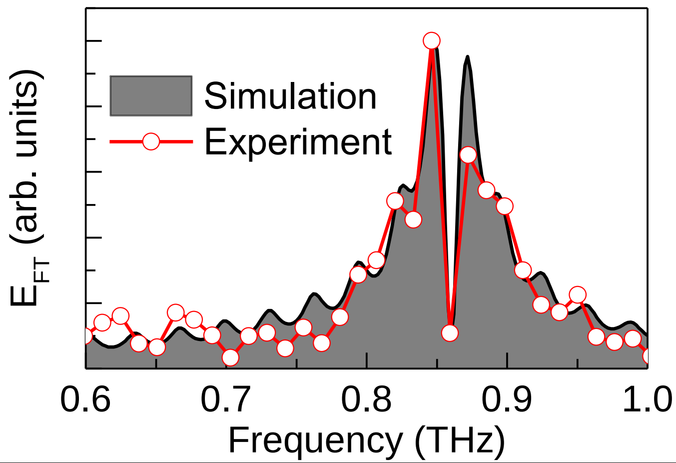
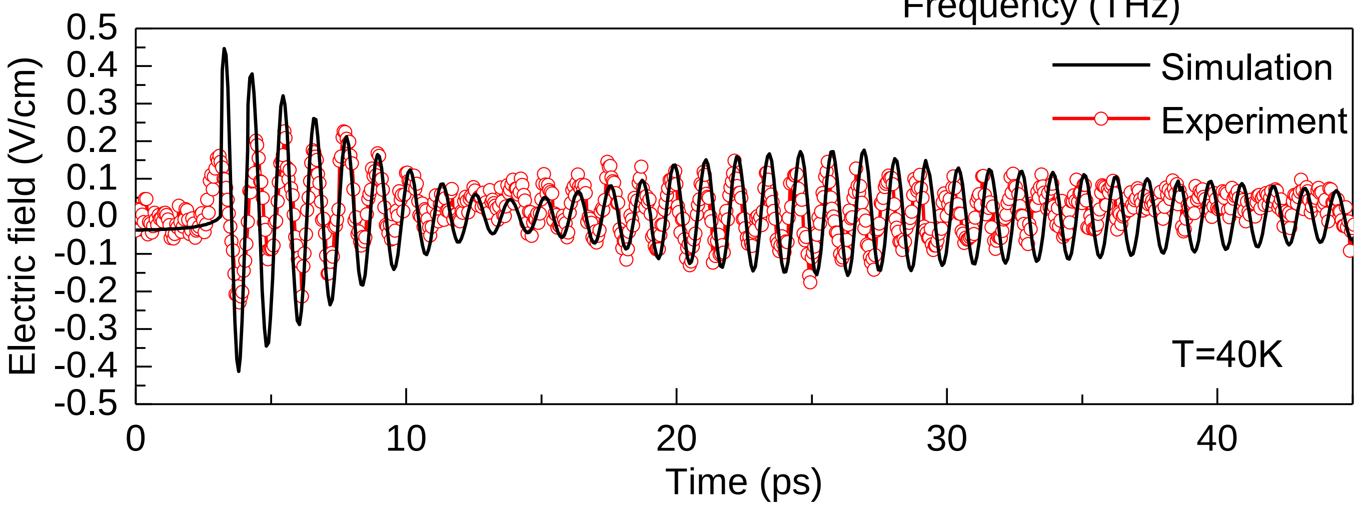
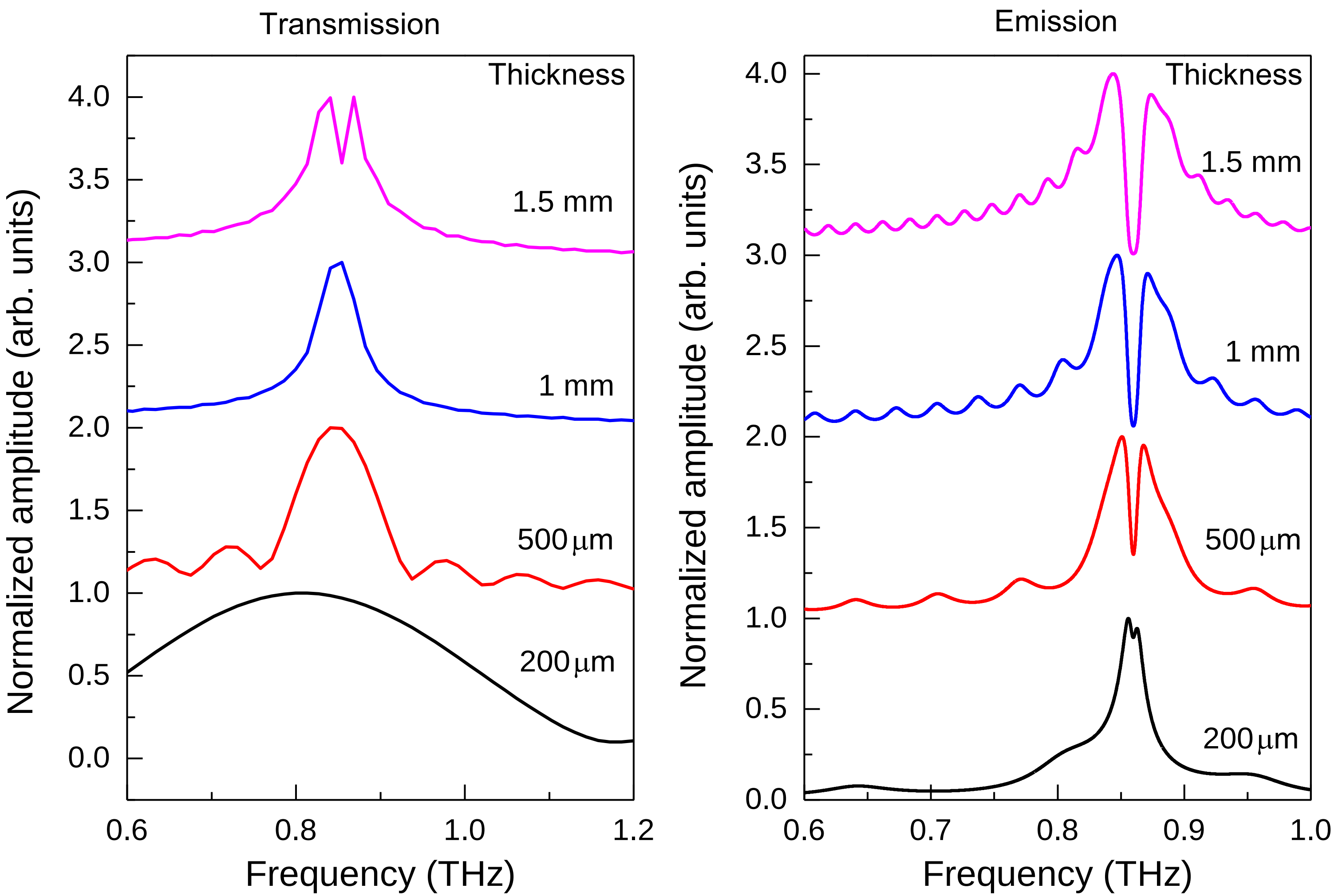
R. Mikhaylovskiy, et al. Nature Comm. 6, 8190 (2015)





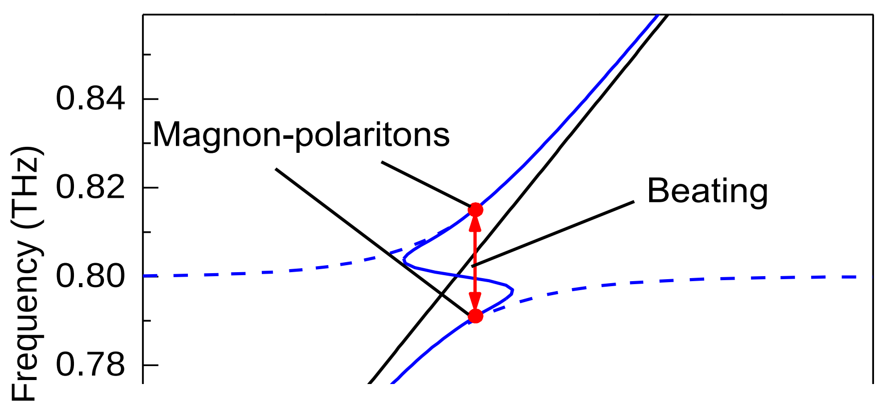


Thickness dependence



Origin of beating

Beating between polariton branches



Propagation effects are important for THz spin excitations!

Does polaritonics matter for THz spin control?

Wavevector (μm^{-1})

Terahertz Magnon-Polaritons in $TmFeO_3$

Kirill Grishunin,[†] Thomas Huisman,[‡] Guanqiao Li,[‡] Elena Mishina,[†] Theo Rasing,[‡] Alexey V. Kimel,[†,‡] Kailing Zhang,[§] Zuanming Jin,[§] Shixun Cao,[§] Wei Ren,[§] Guo-Hong Ma,[*,§] and Rostislav V. Mikhaylovskiy[*,‡]

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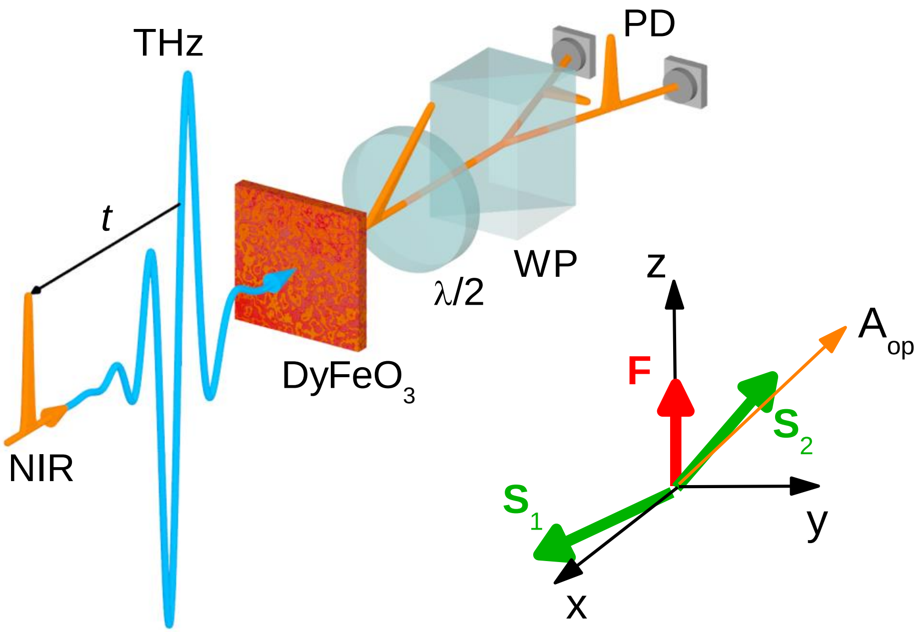
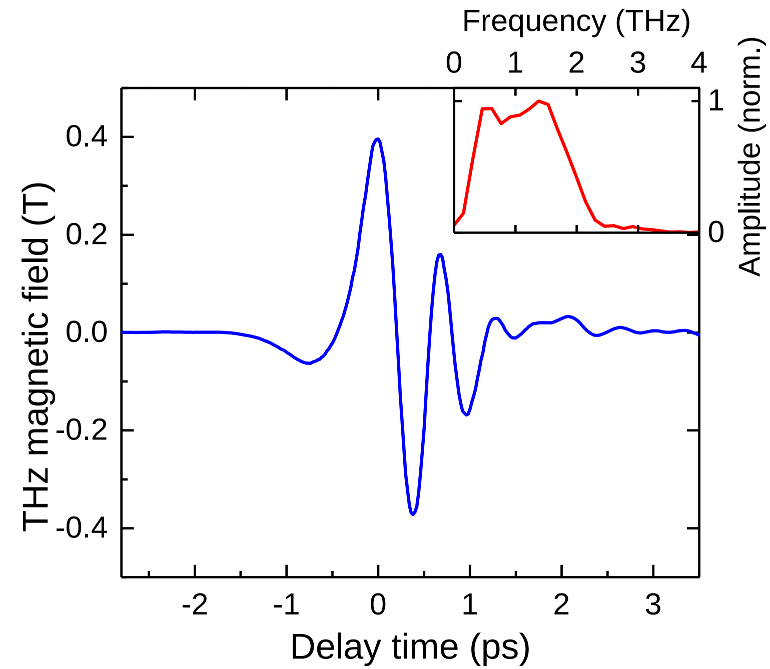
THz control in $DyFeO_3$

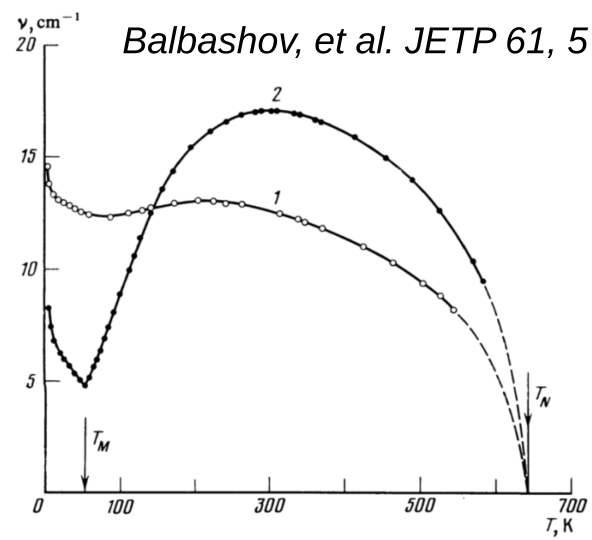




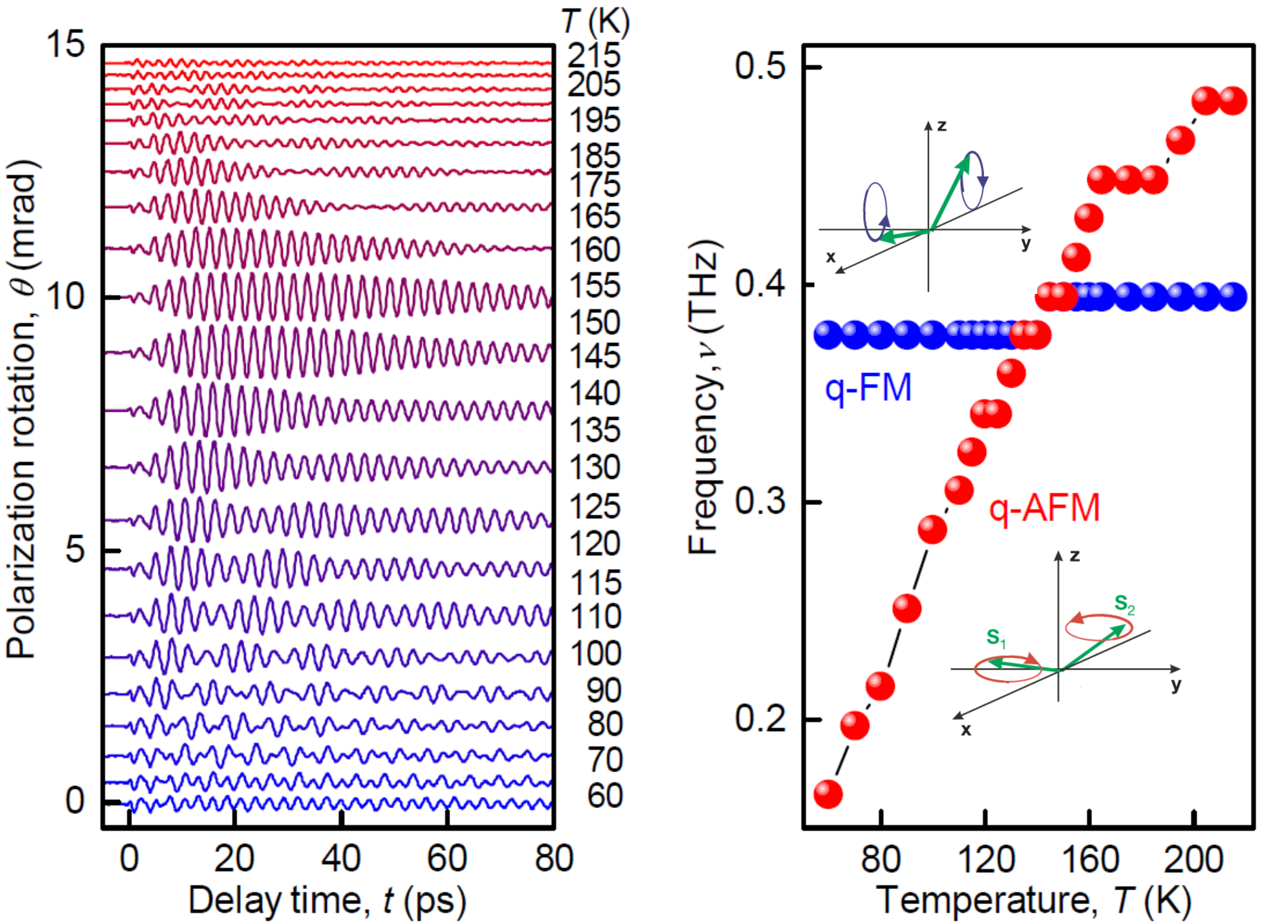
Propagation along optical axis

Antiferromagnetic modes cross each other in $DyFeO_3$

Balbashov, et al. JETP 61, 573 (1985)

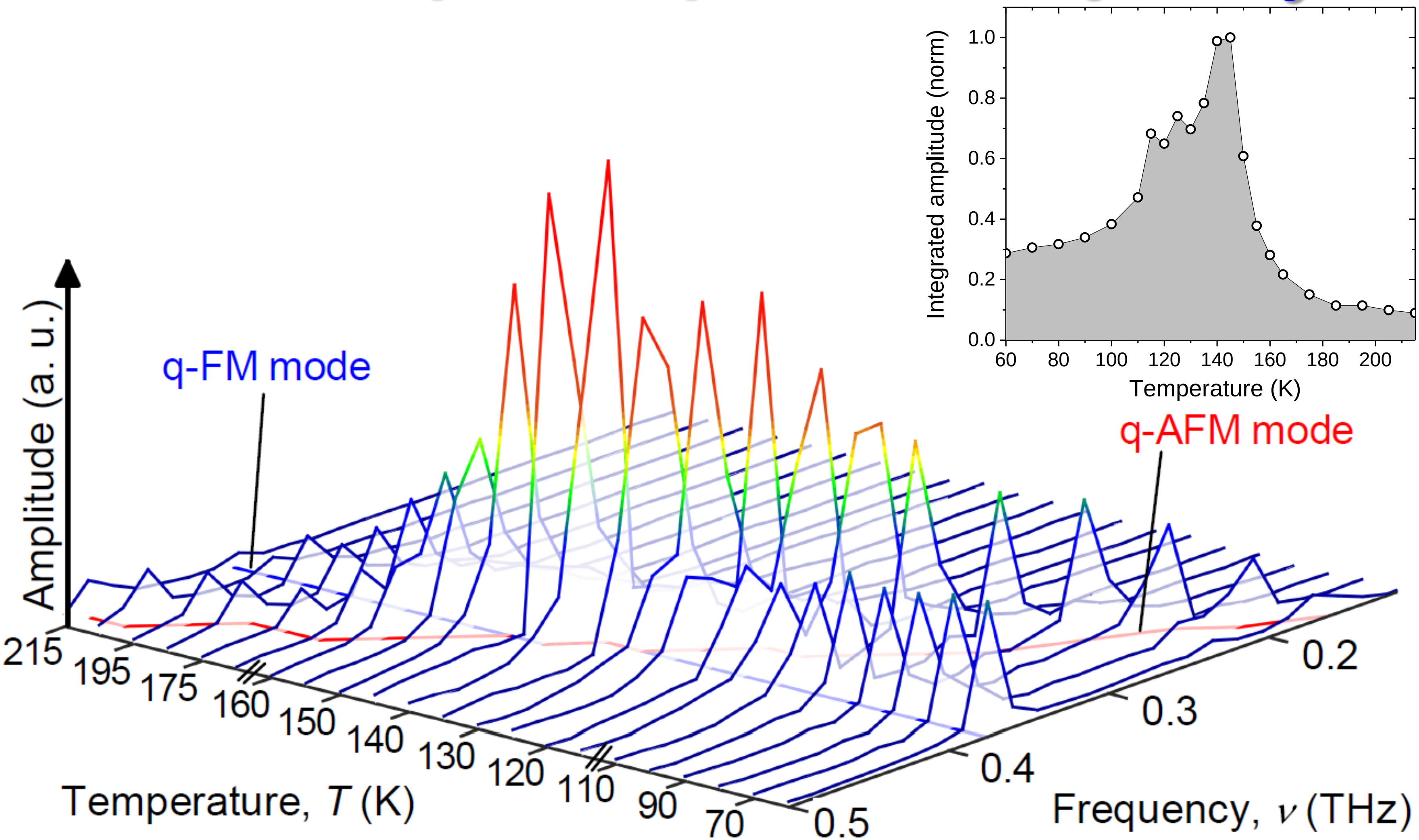


THz spin response of $DyFeO_3$



Enhancement at crossing point?

THz spin response of $DyFeO_3$



Enhancement of spectral weight

Internal resonance

Assume coupling between magnon modes

$$\ddot{l}_z + \gamma_{\mathrm{qFM}} \dot{l}_z + \omega^2_{\mathrm{qFM}} l_z = b(t)$$

$$\ddot{l}_y + \gamma_{\mathrm{qAFM}} \dot{l}_y + \omega^2_{\mathrm{qAFM}} l_y = b(t) + \boxed{\alpha l_z}$$

Internal resonance

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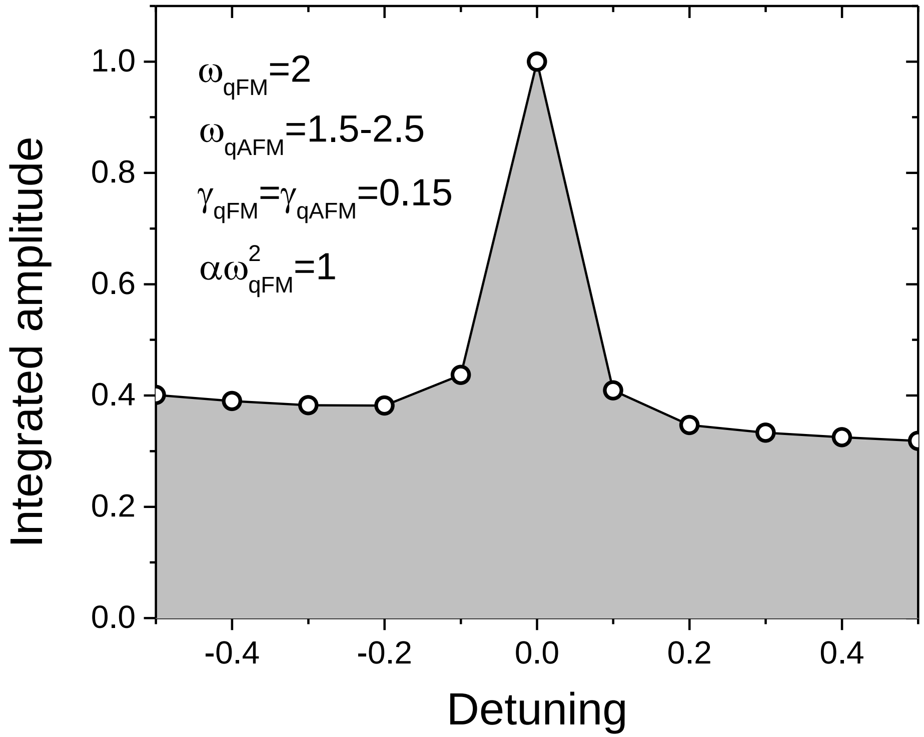


Additional resonant term

$$\tilde{l}_z(\omega) = \frac{\tilde{b}(\omega)}{\left(\omega^2_{\mathrm{qFM}} + i\gamma_{\mathrm{qFM}}\omega - \omega^2\right)}$$

$$\tilde{l}_y(\omega) = \frac{\tilde{b}(\omega)}{\left(\omega^2_{\mathrm{qAFM}} + i\gamma_{\mathrm{qAFM}}\omega - \omega^2\right)} + \boxed{\frac{\alpha\tilde{b}(\omega)}{\left(\omega^2_{\mathrm{qAFM}} + i\gamma_{\mathrm{qAFM}}\omega - \omega^2\right)\left(\omega^2_{\mathrm{qFM}} + i\gamma_{\mathrm{qFM}}\omega - \omega^2\right)}}$$

Coupling origin?

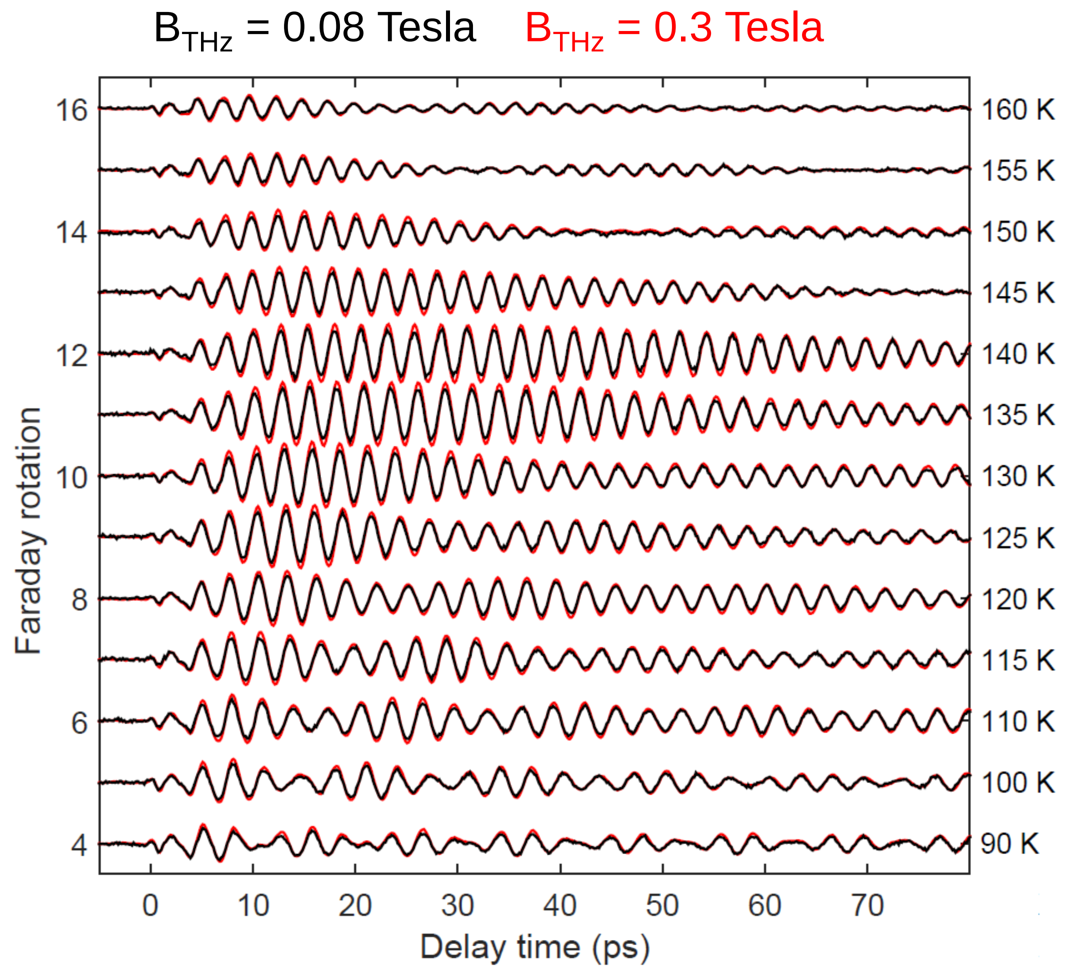
Coupling origin

Possibilities

- ~~*Nonlinear coupling*~~

Ruled out by experiment

Propagation!



No nonlinearity

Magnon-polaritons in $DyFeO_3$

Modes are orthogonal (?)

How do they propagate?

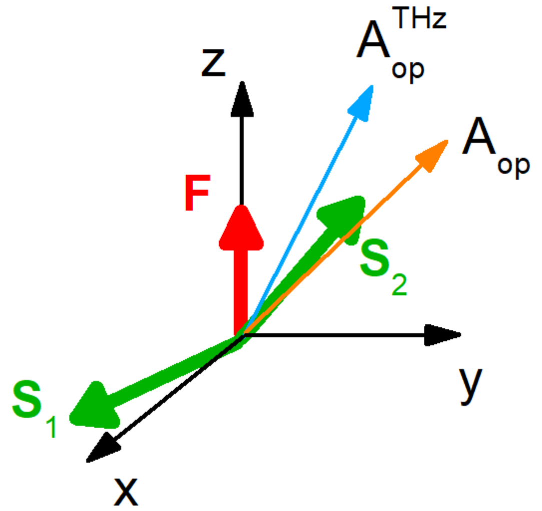
Orthoferrite is highly birefringent crystal at THz!

$$\hat{\varepsilon} = \begin{pmatrix} \varepsilon_x = 23.6 & 0 & 0 \\ 0 & \varepsilon_y = 21.3 & 0 \\ 0 & 0 & \varepsilon_z = 25.6 \end{pmatrix}$$

$$\Delta\mu < 0.1$$

Anisotropy from dielectric properties

THz optical axis is not sample normal



qFM $m_{x,y} \Leftrightarrow h_{x,y}$

qAFM $m_z \Leftrightarrow h_z$

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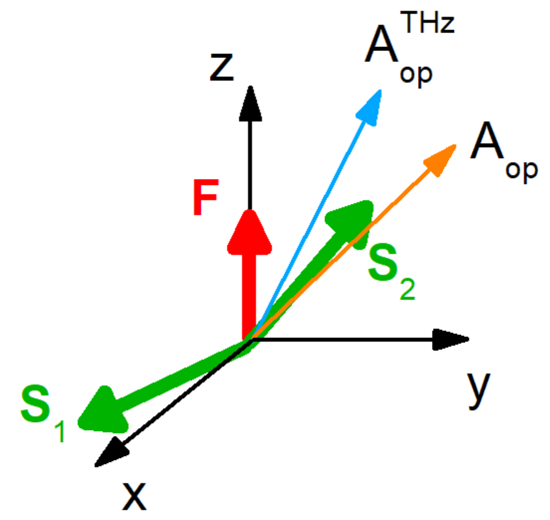

qFM $\cancel{m_{x,y} \Leftrightarrow h_{x,y}}$ $\quad m_{x,y} \Leftrightarrow h_o, h_e$

qAFM $\cancel{m_z \Leftrightarrow h_z}$ $\quad m_z \Leftrightarrow h_o, h_e$

Not normal modes of $DyFeO_3$

Coupling mediated by lattice/dielectric properties

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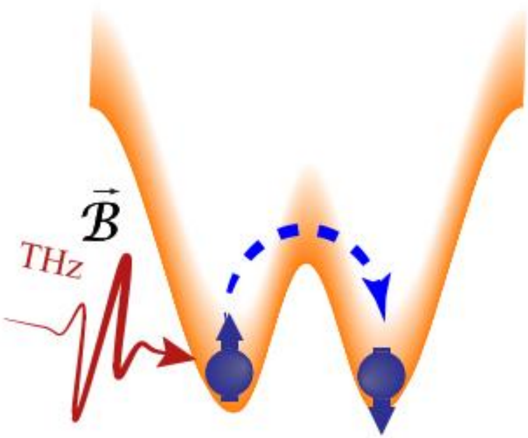
Electric field driven terahertz control

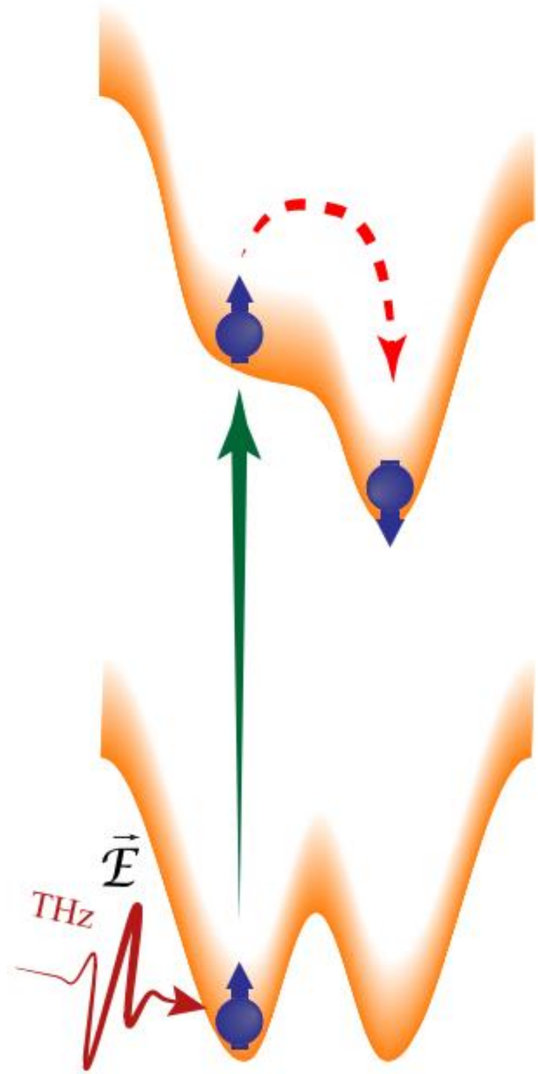
Magnetic nonlinearity?



Magnetic field works against high potential barrier

10 Tesla are required

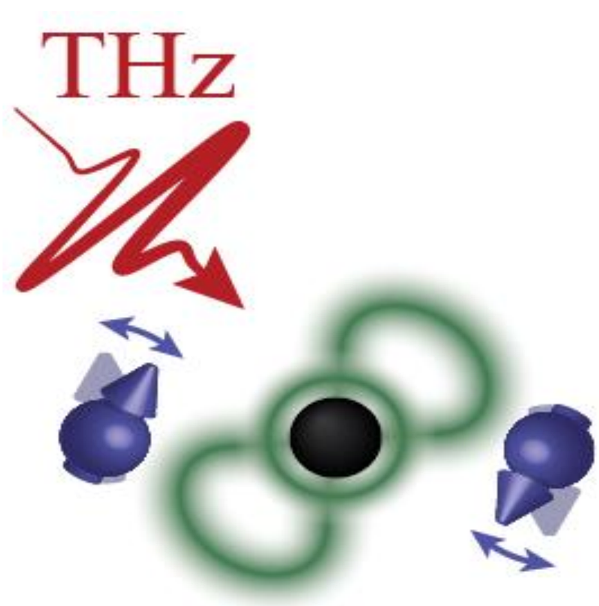
Alternative scenario



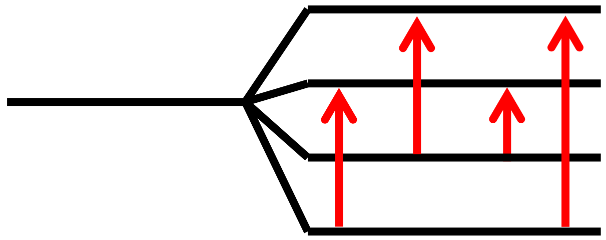
Electric field driven terahertz control

We propose to employ low-energy electric dipole-active excitations coupled to magnetic order

Orbital states set magnetic anisotropy

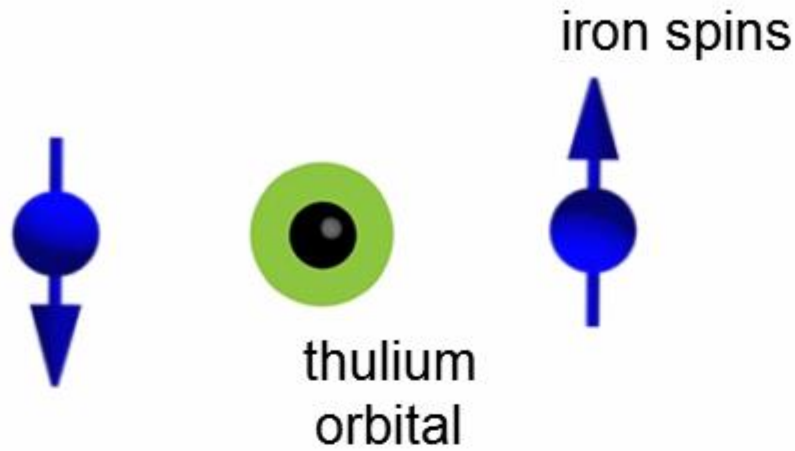


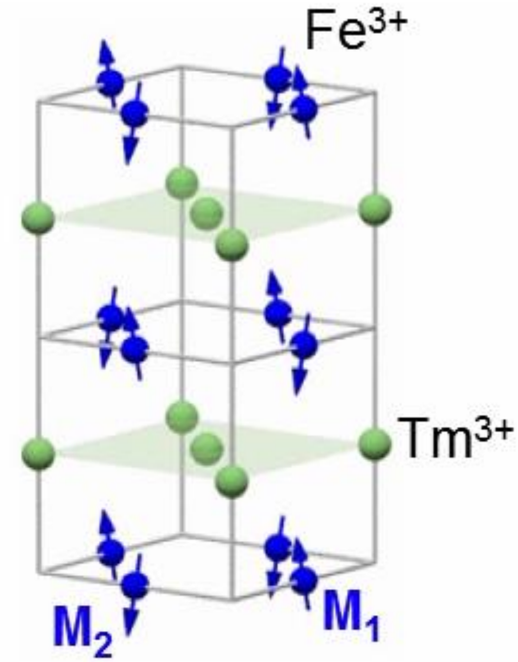
Electronic transitions in crystal-field

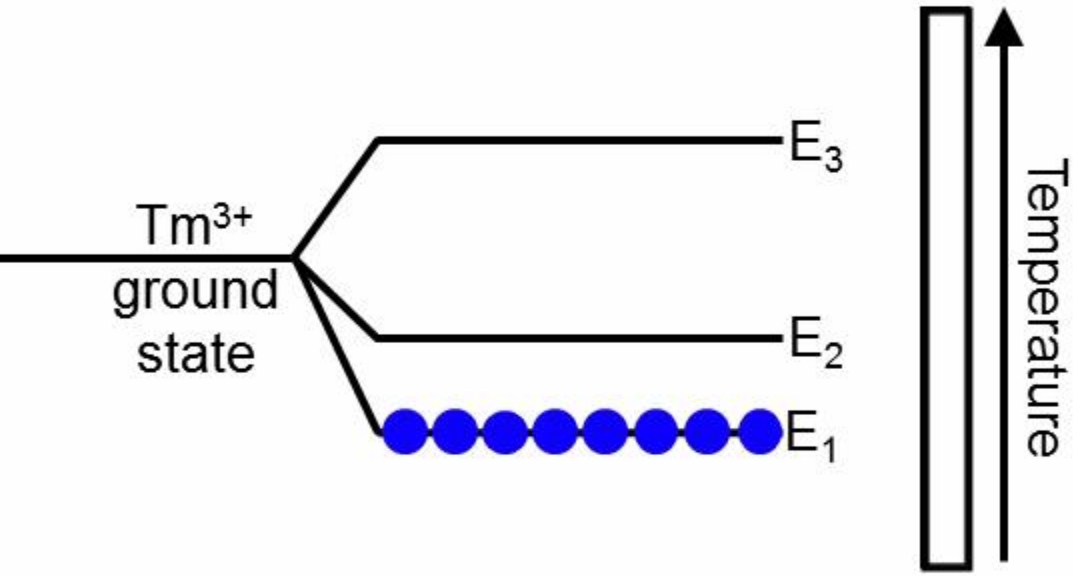


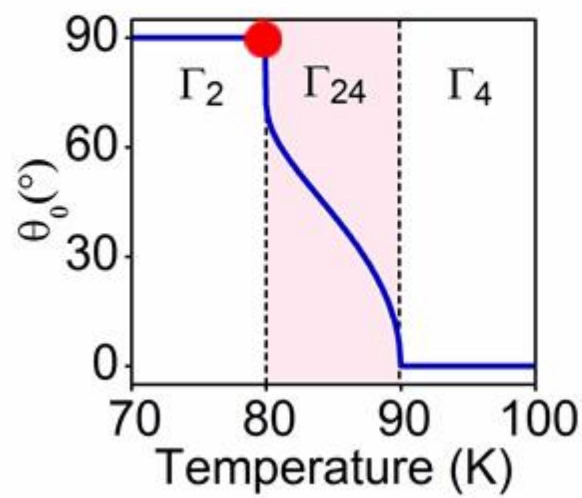
Can THz-population of electronic orbitals drive spin motion?

Spin reorientation in $TmFeO_3$

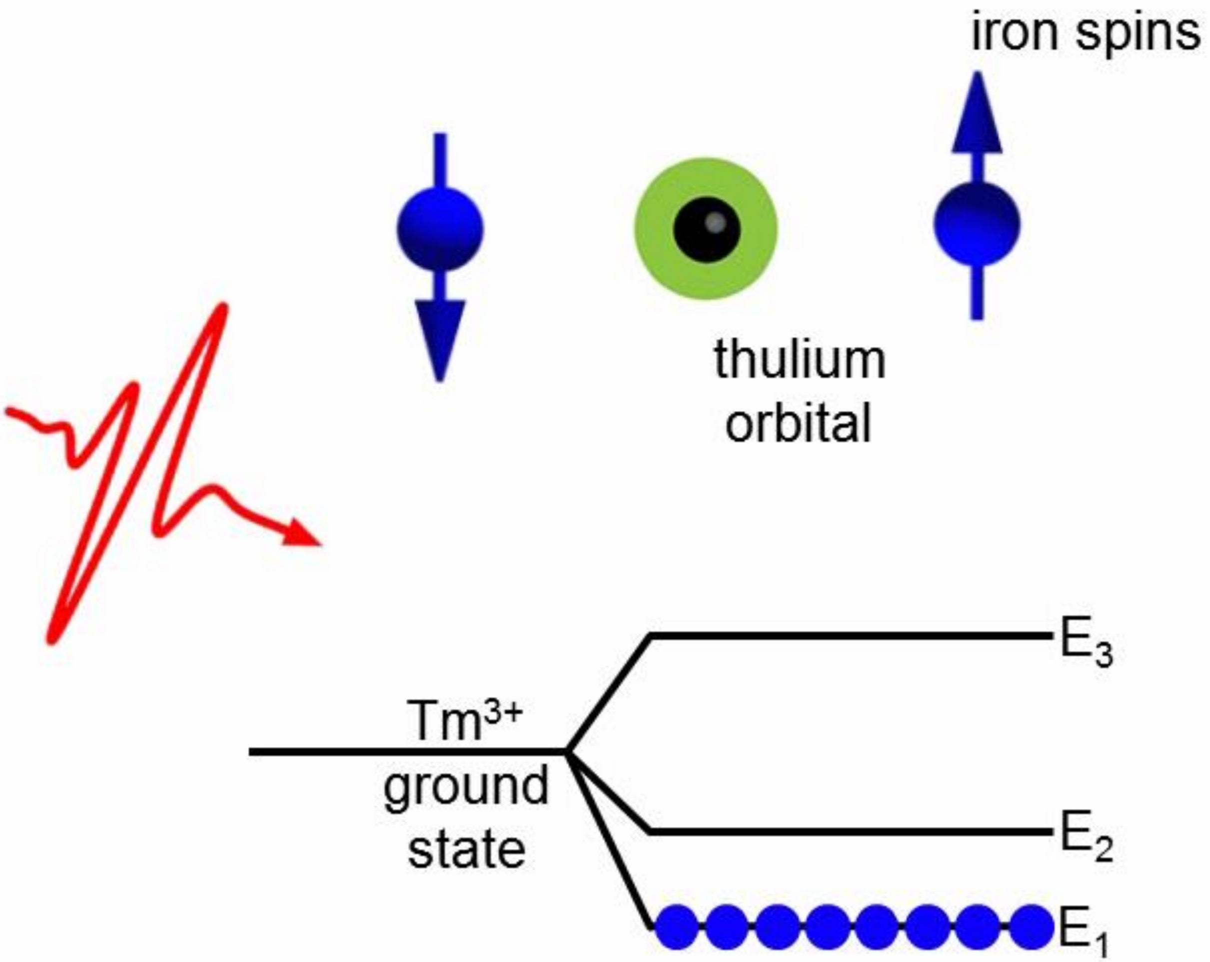




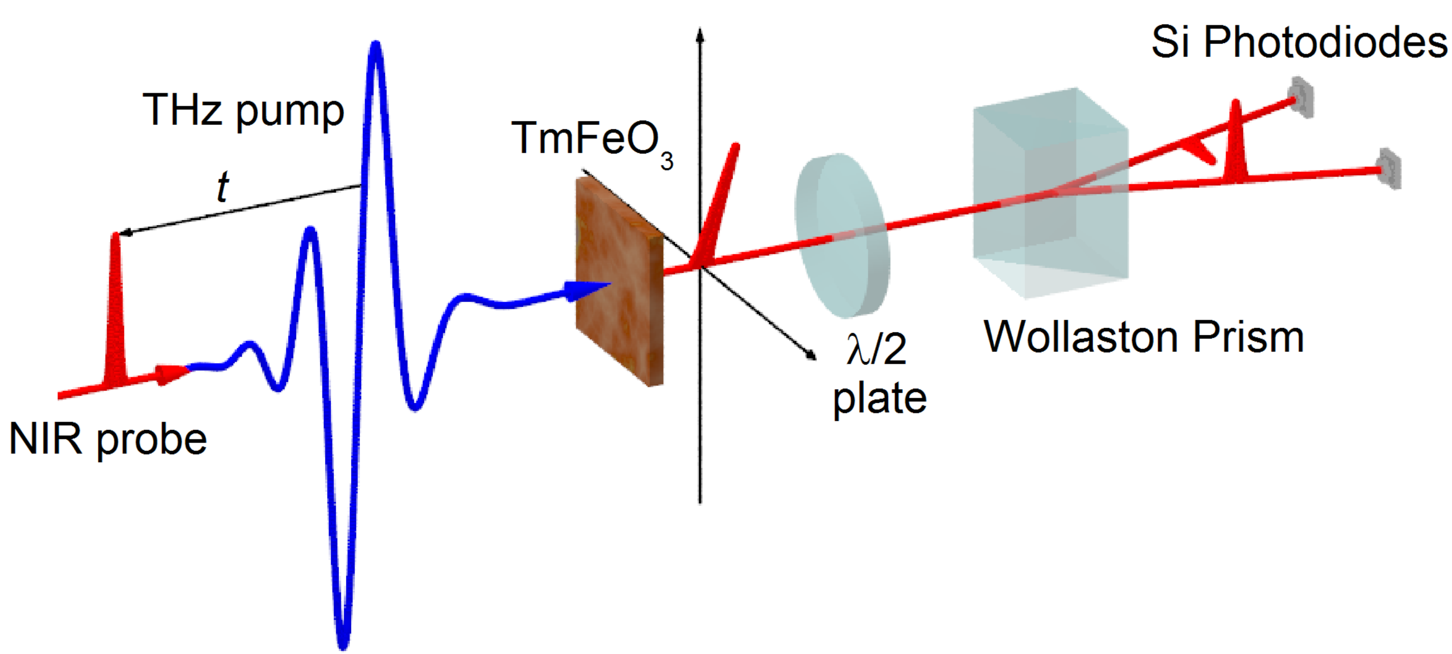


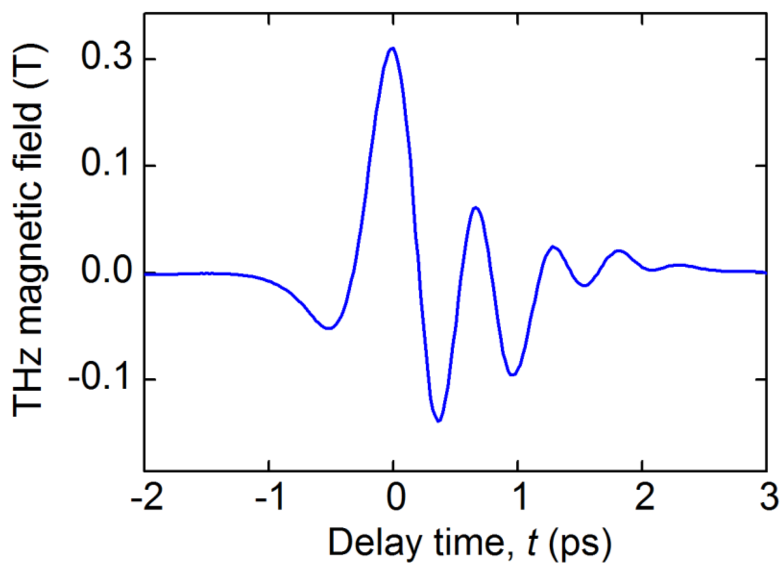


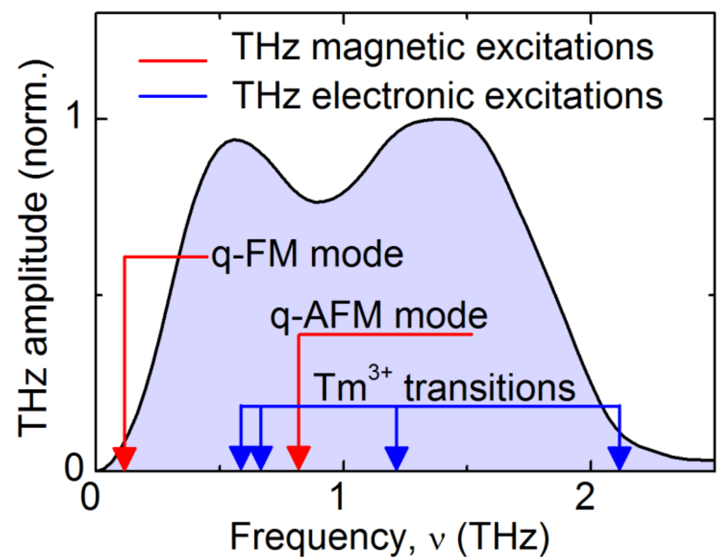
Terahertz-driven spin reorientation



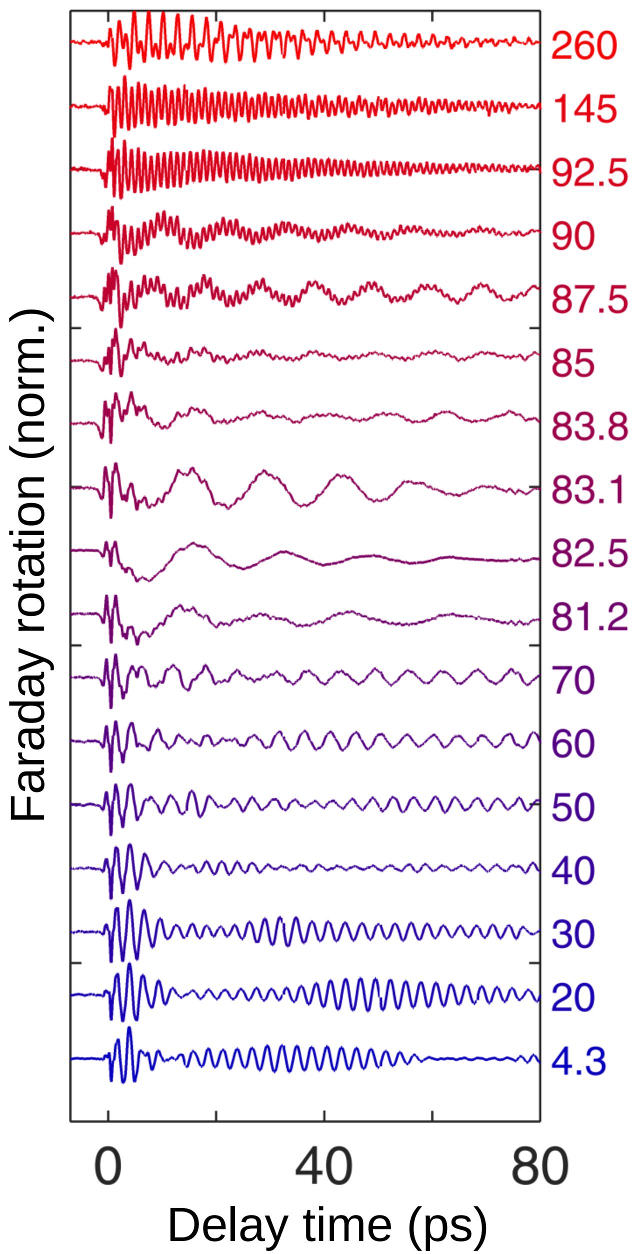
THz-pump optical-probe experiment

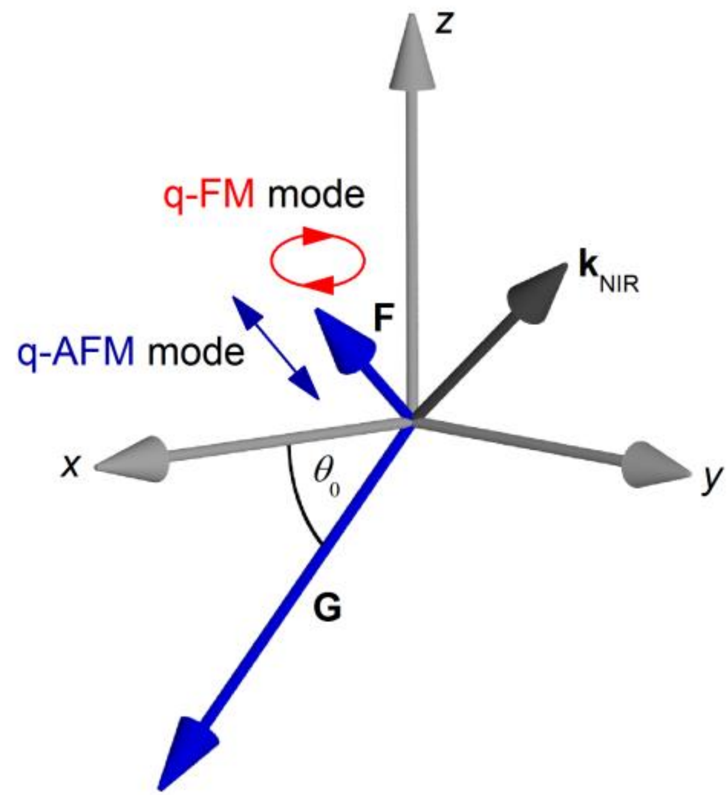






$TmFeO_3$: antiferromagnetic resonance





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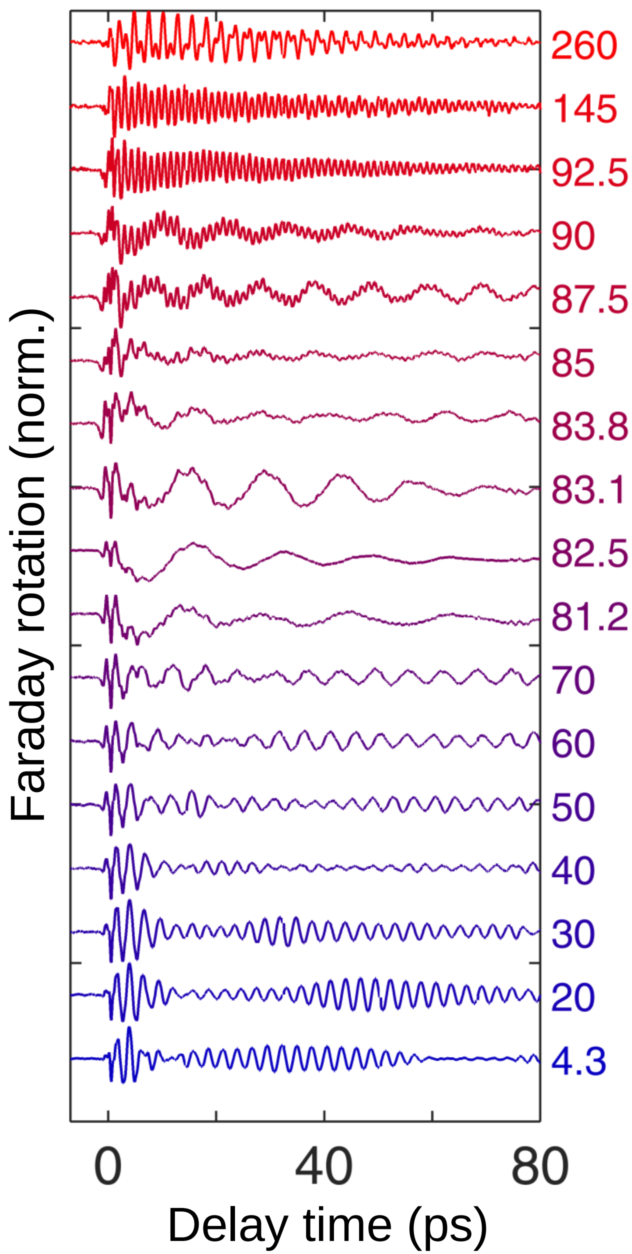
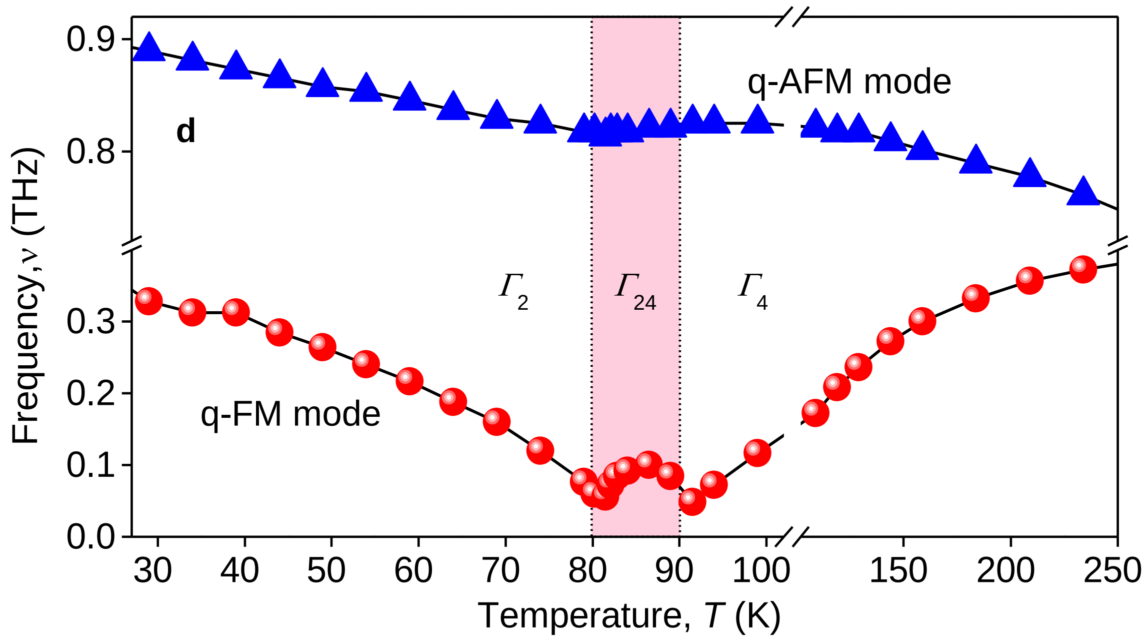
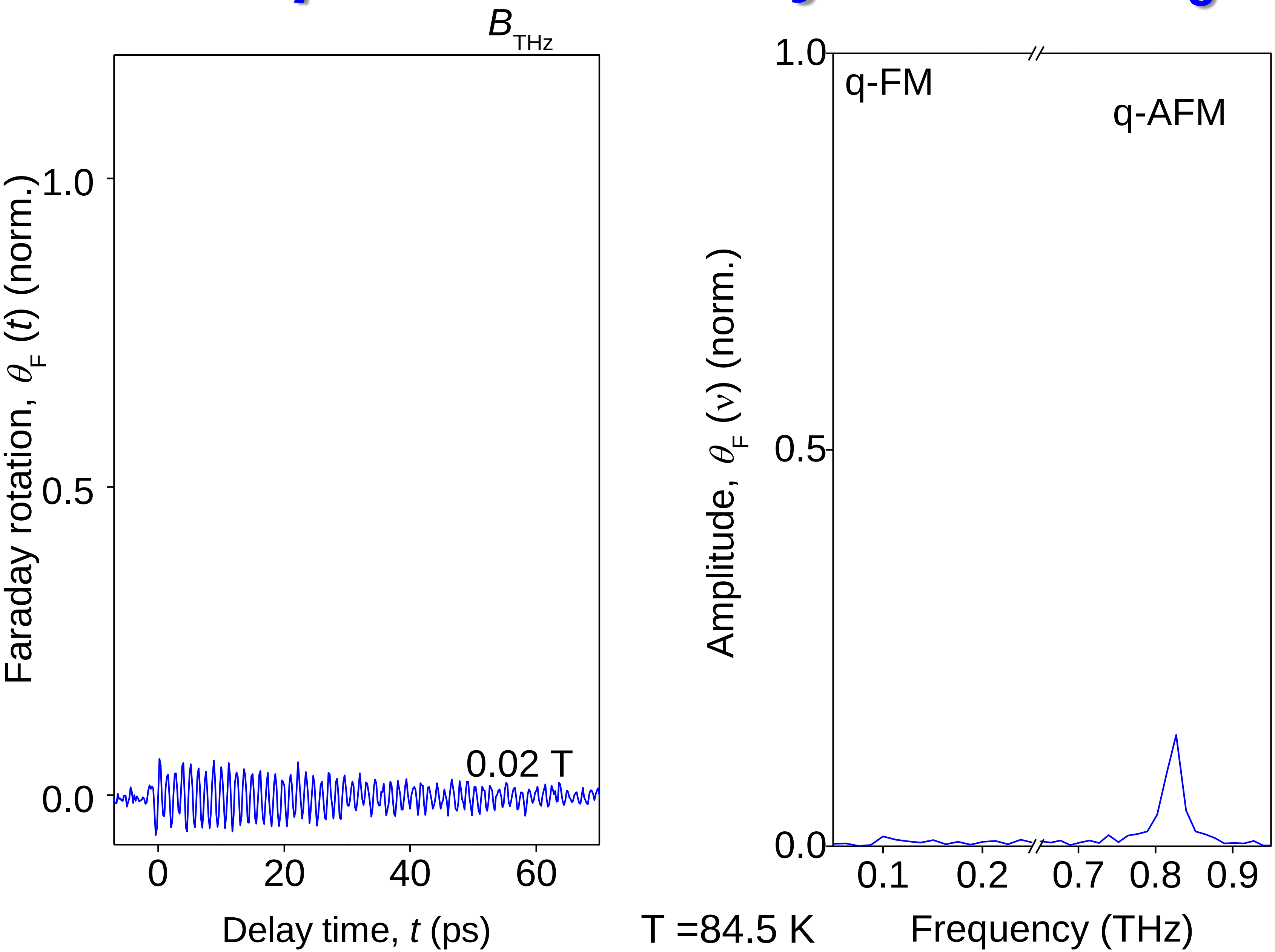


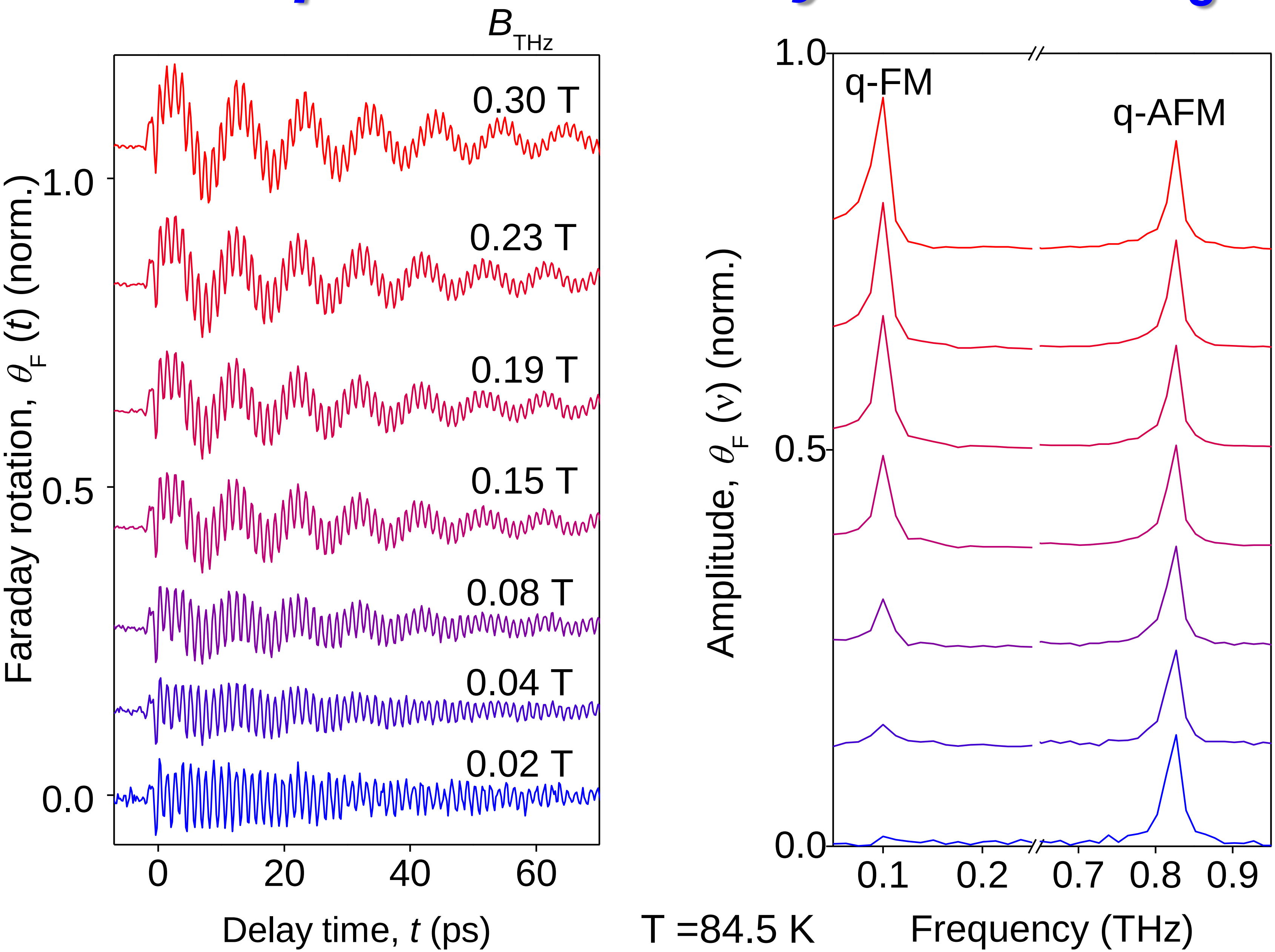


Ferromagnetic mode **softens** at **magnetic phase transitions**

THz-spin nonlinearity in $TmFeO_3$

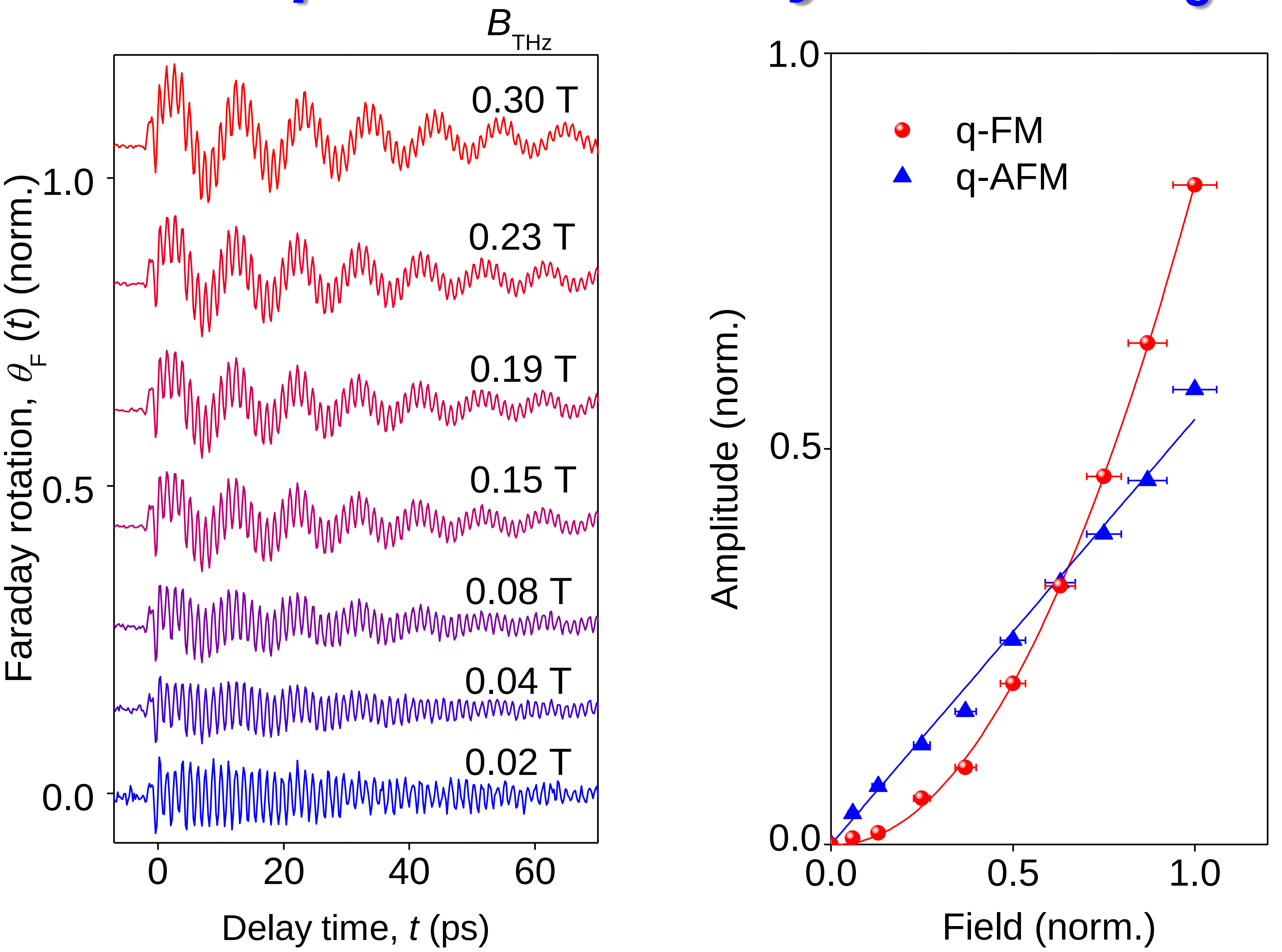


THz-spin nonlinearity in $TmFeO_3$

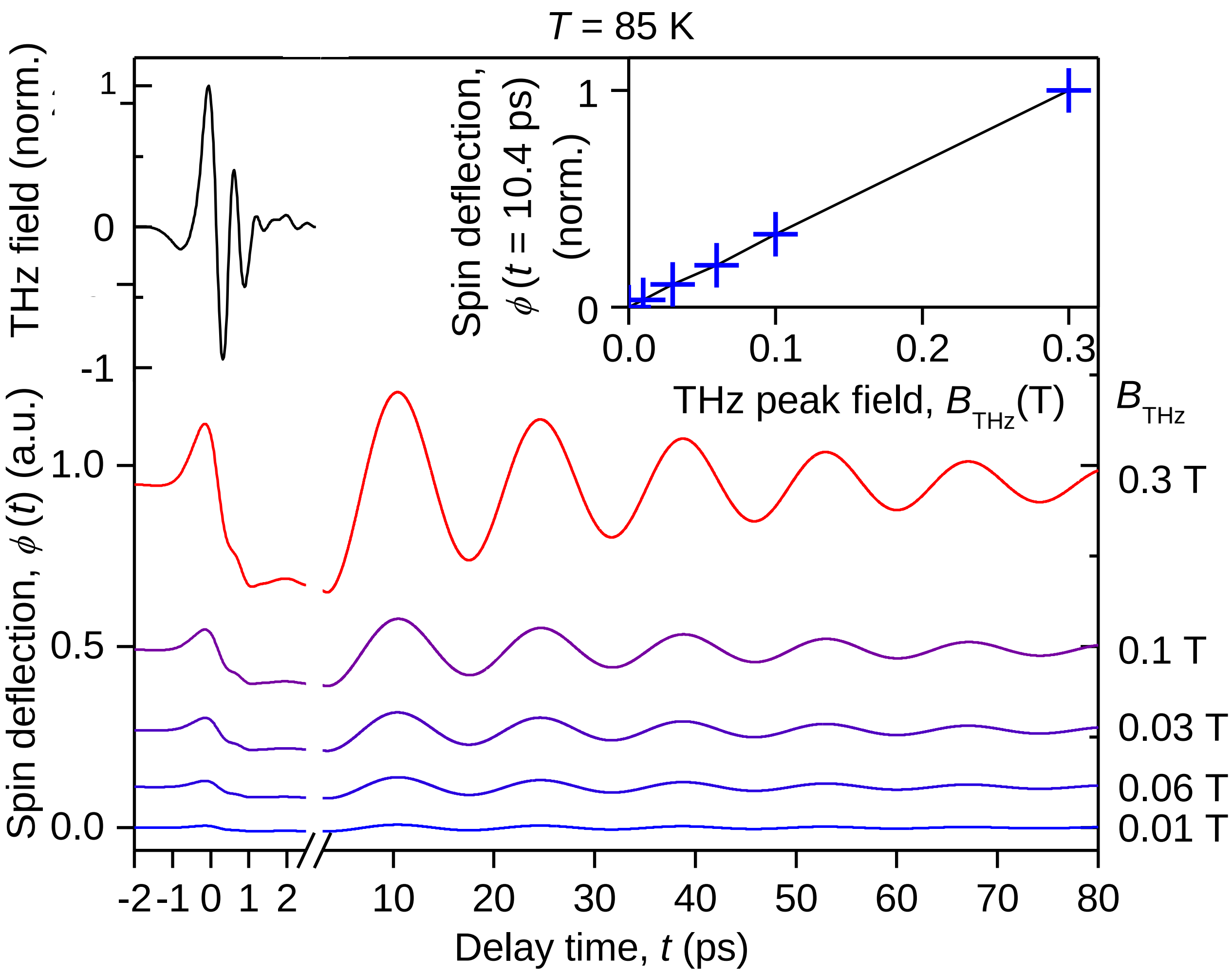


T =84.5 K

THz-spin nonlinearity in $TmFeO_3$



Origin of nonlinearity?



Can Zeeman interaction drive spins into strongly nonlinear regime?

- Quantitative simulation of Lagrangian FM mode dynamics
- **linear behaviour** predicted for isolated iron spin system

Phenomenology of nonlinearity

- Crystal symmetry D_{2h}^{16} allows for free energy terms **quadratic in THz electric field** in the intermediate phase:

$$W = \left(\chi_{xx} E_x^2 + \chi_{yy} E_y^2 + \chi_{zz} E_z^2\right) \sin^2 \theta$$

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- corresponds to **THz induced change in magnetic anisotropy**

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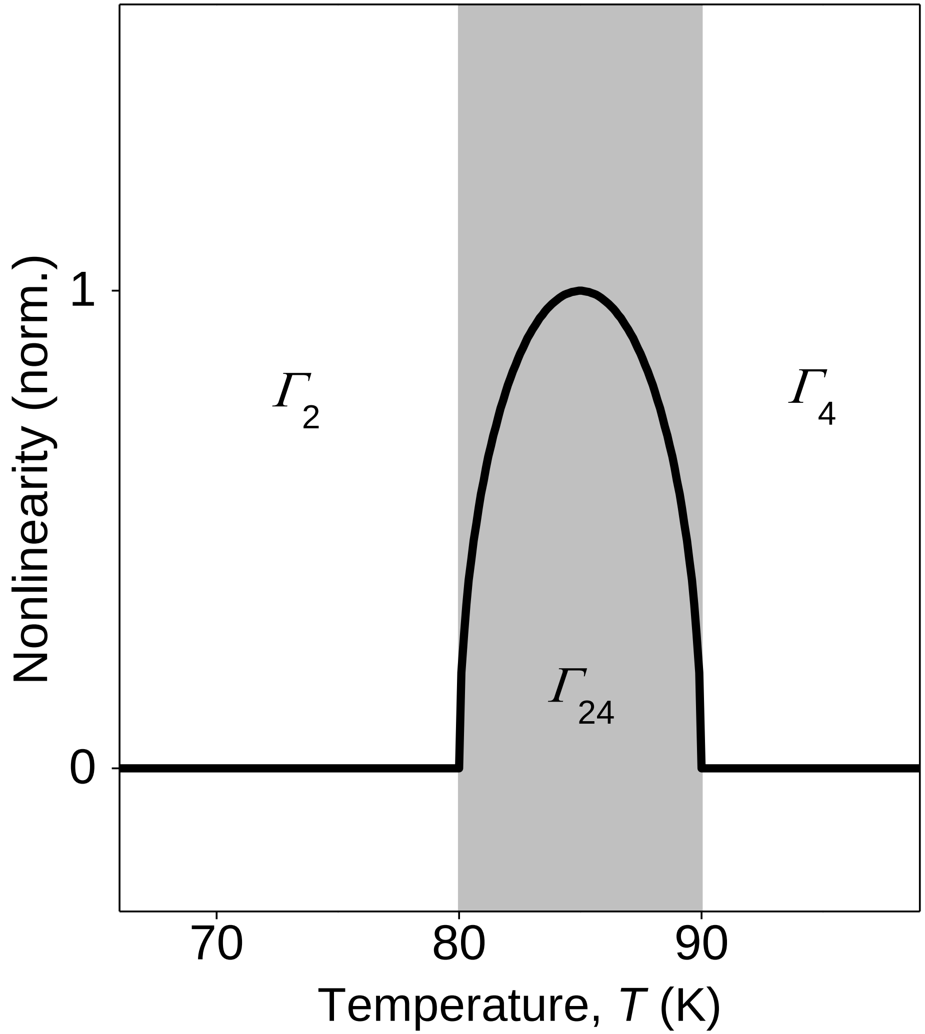

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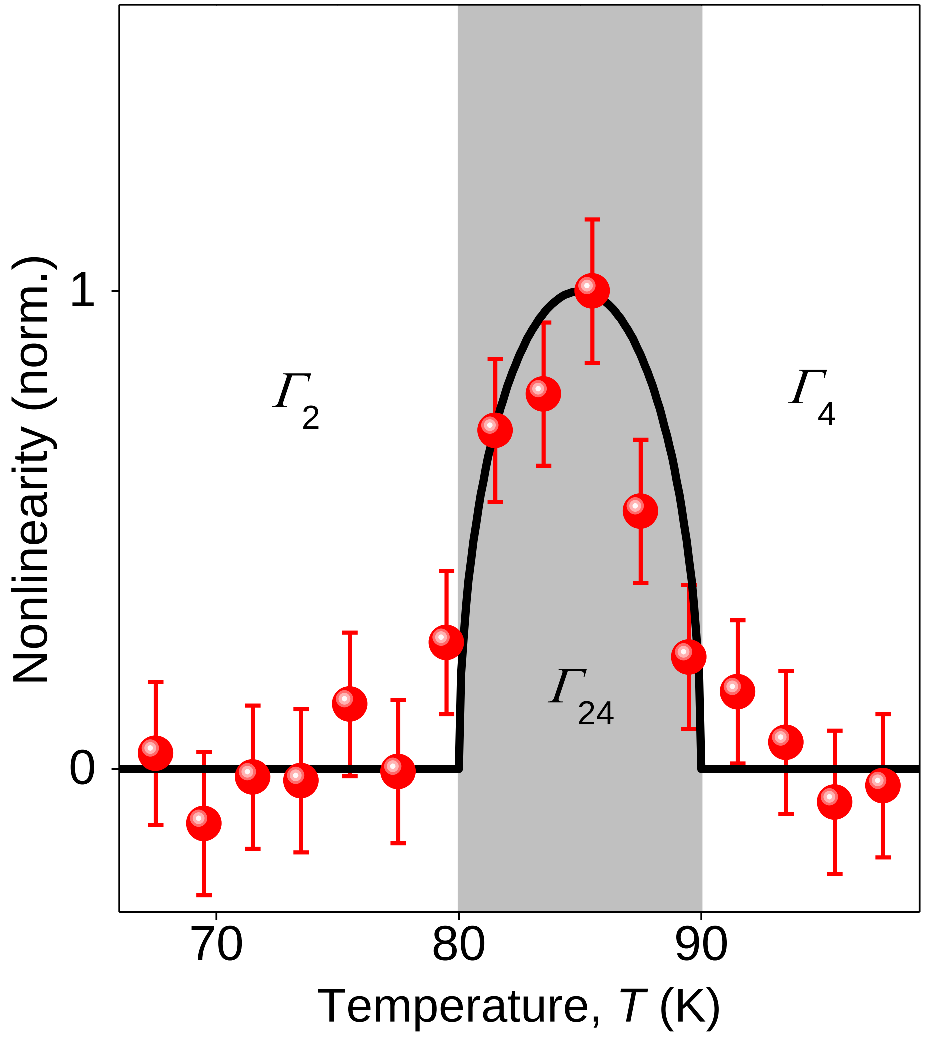
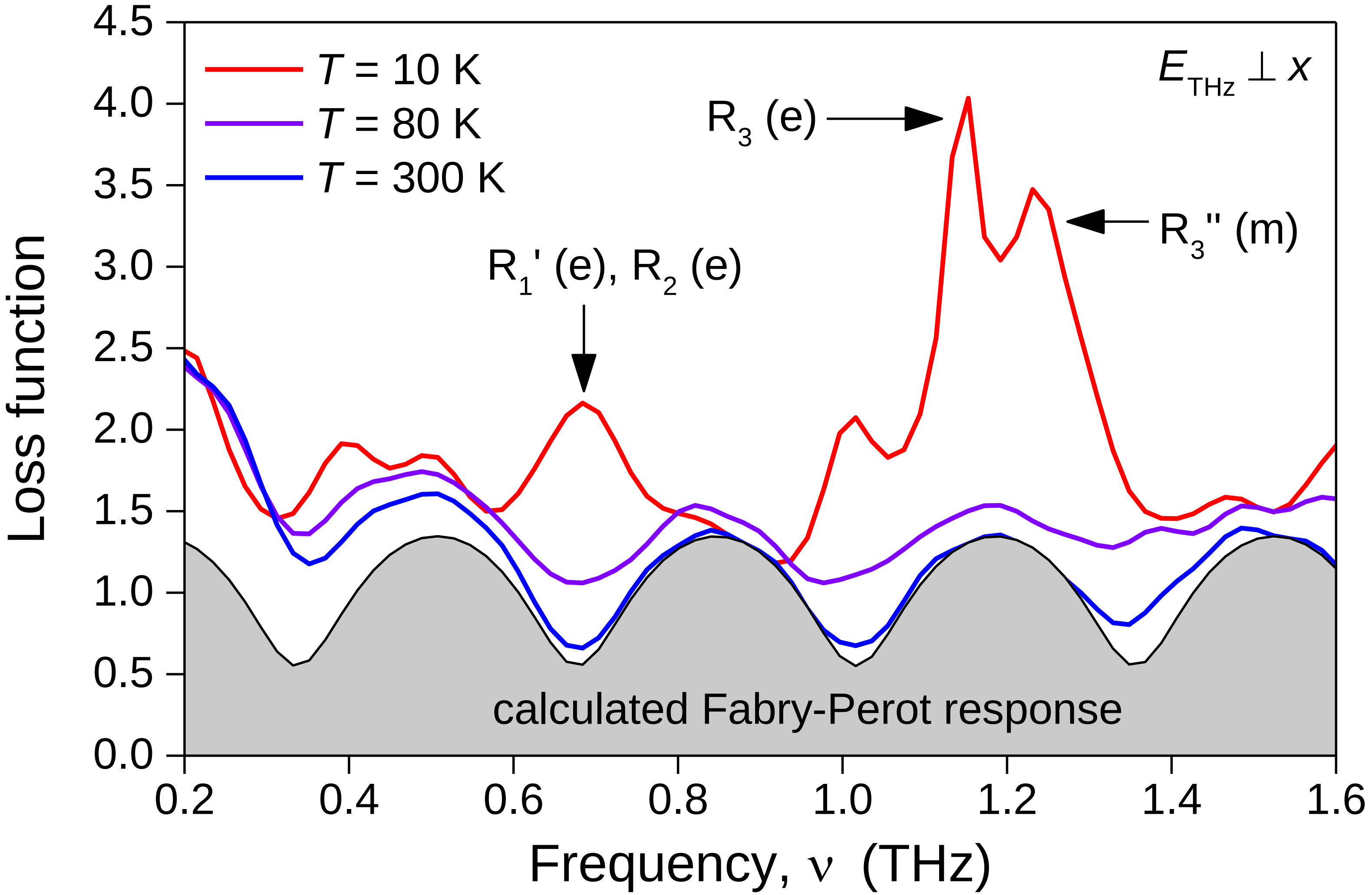
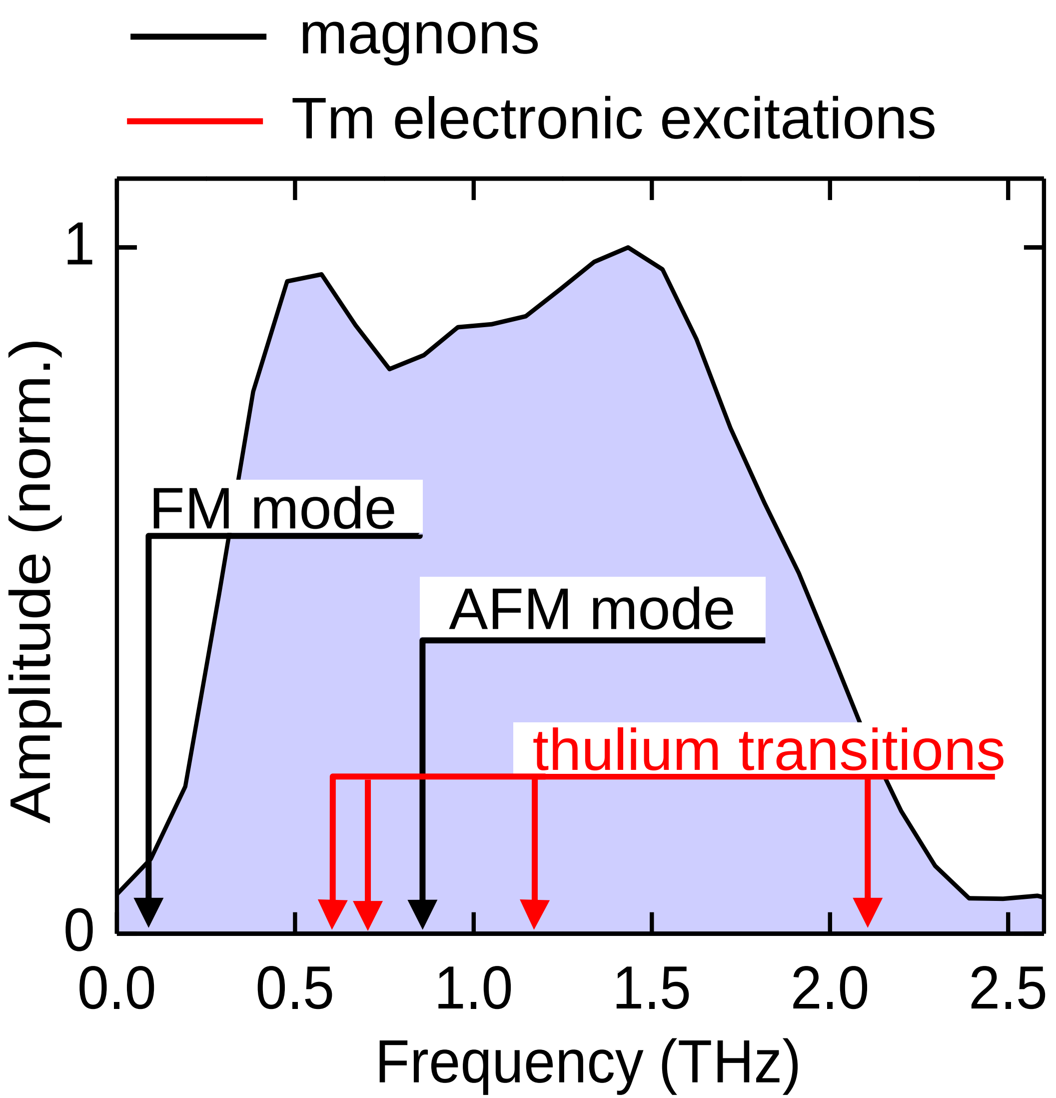


Tm states?

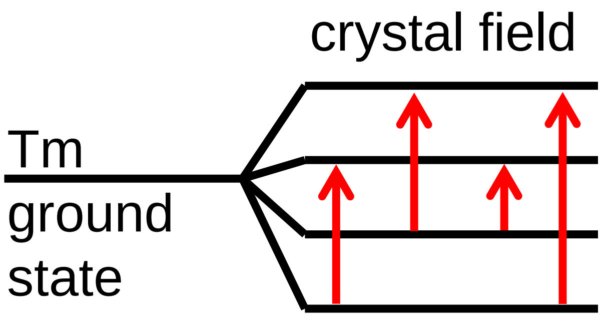
THz excitations in $TmFeO_3$



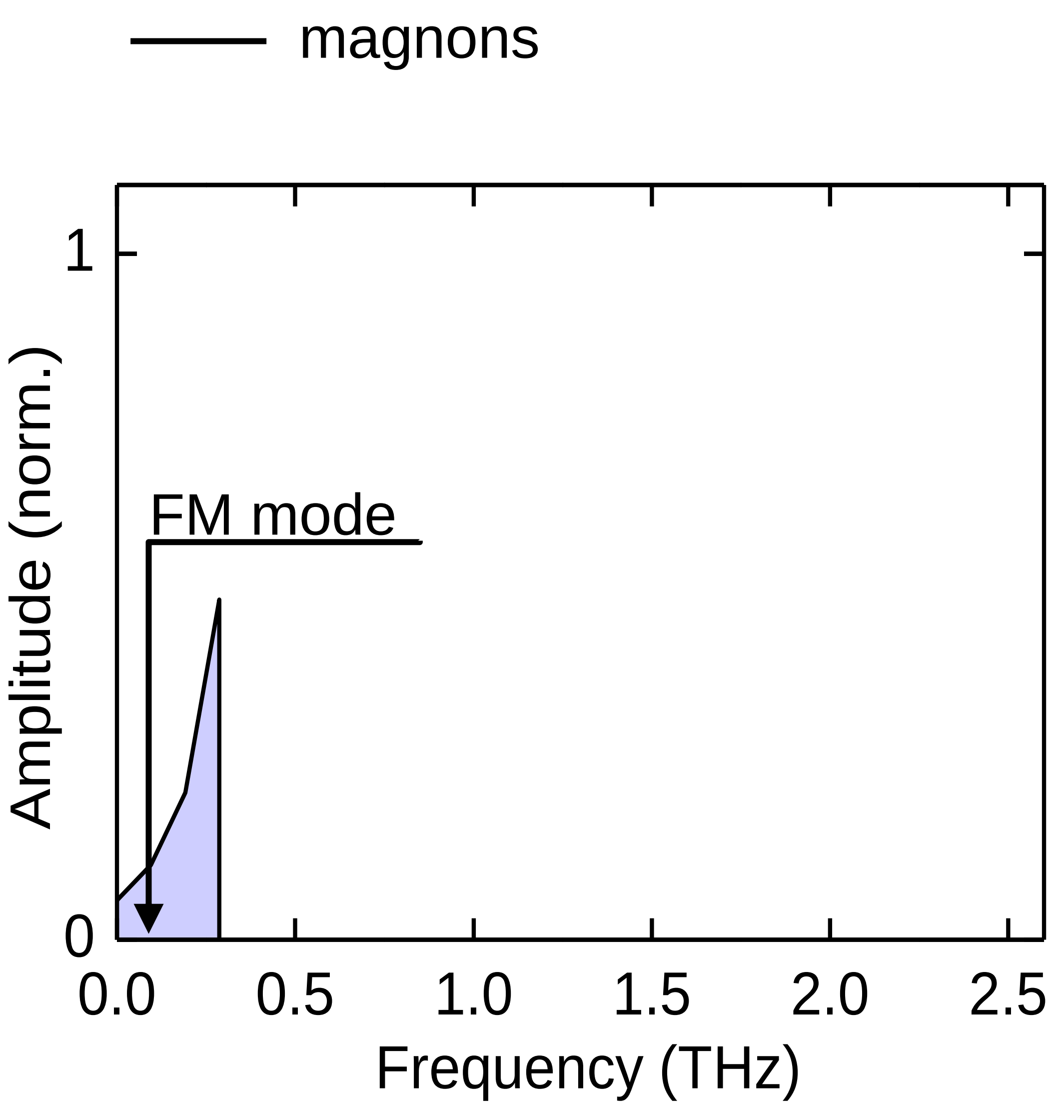
THz excitations in $TmFeO_3$



- Resonant Zeeman-type excitation of magnons by **THz magnetic field**
- **Electric dipole interaction** with 4 electronic excitations of Tm



THz excitations in $TmFeO_3$

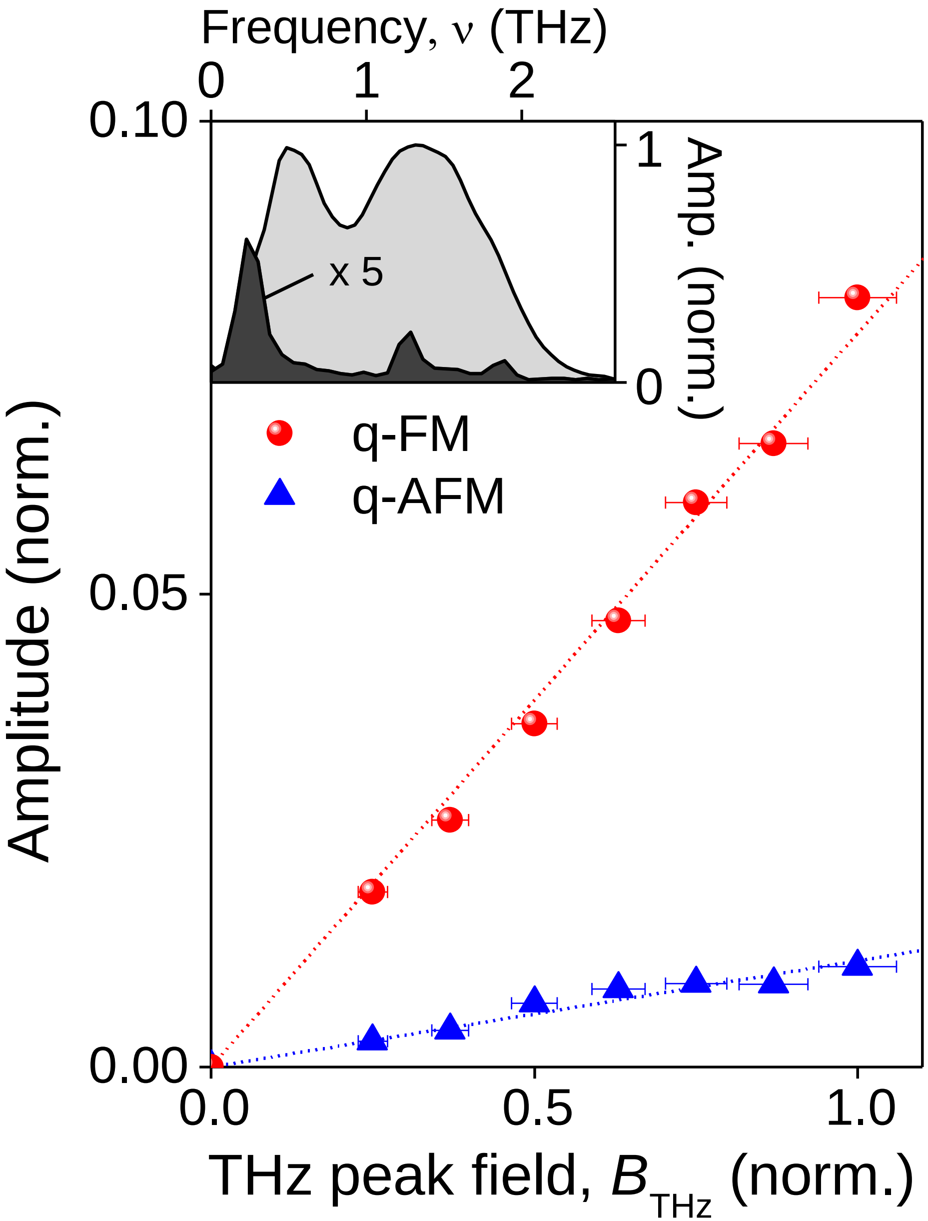


Excite FM mode only by resonant Zeeman interaction

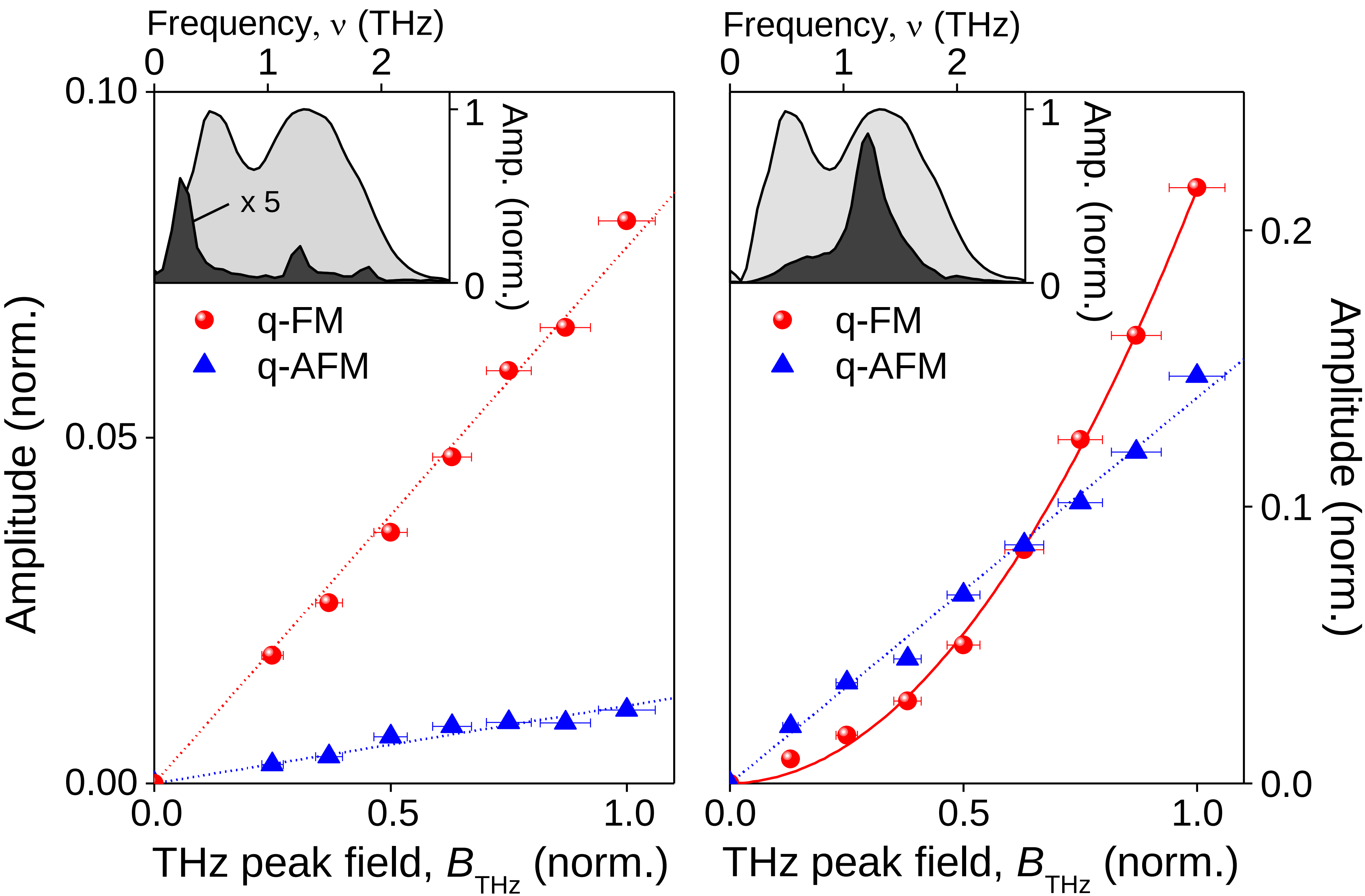
→ **rule out** influence of **electric dipole** transitions by spectral filtering

→ Expectation: **no nonlinearity!**

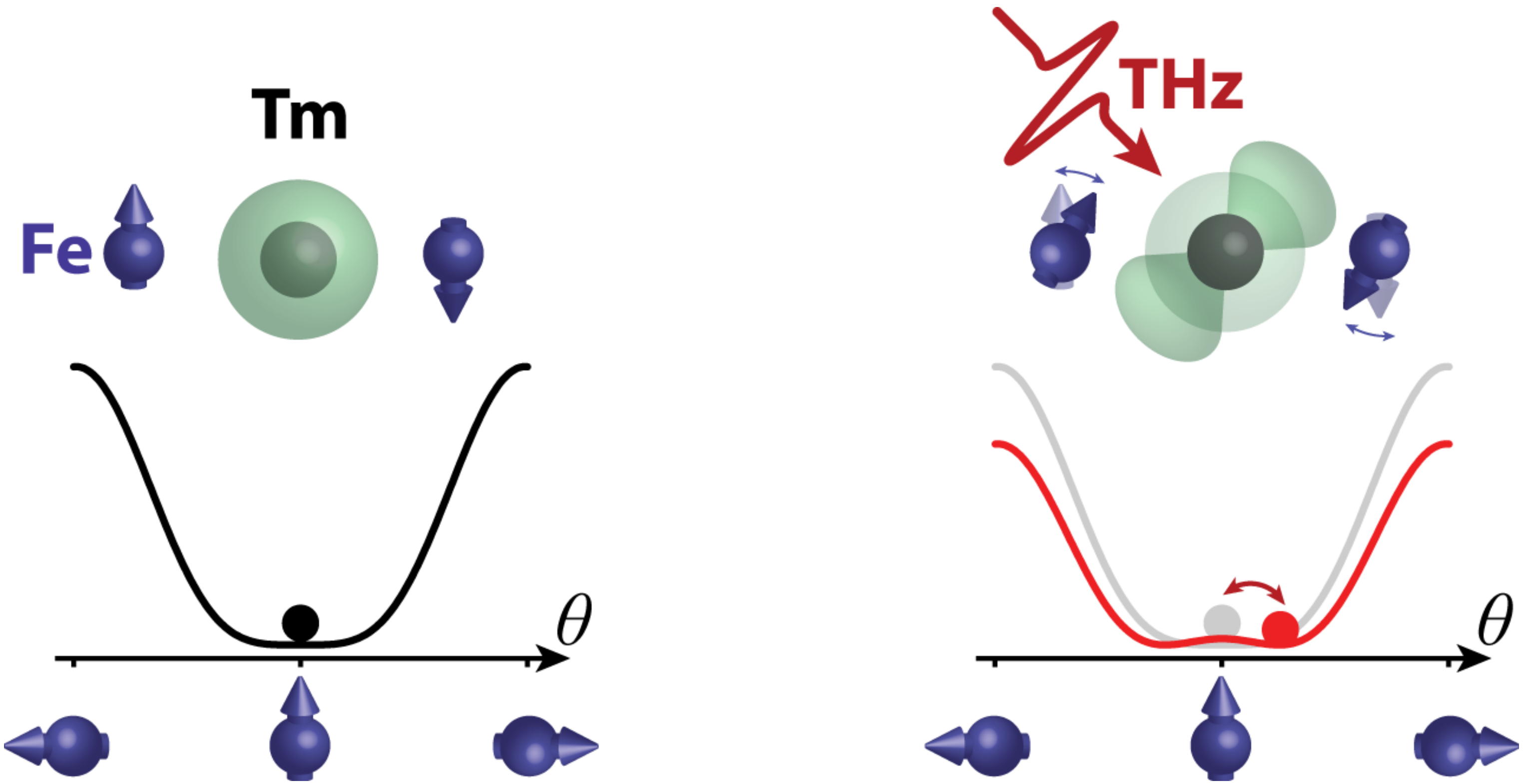
Spectral tailoring



Spectral tailoring



Terahertz-driven anisotropy change

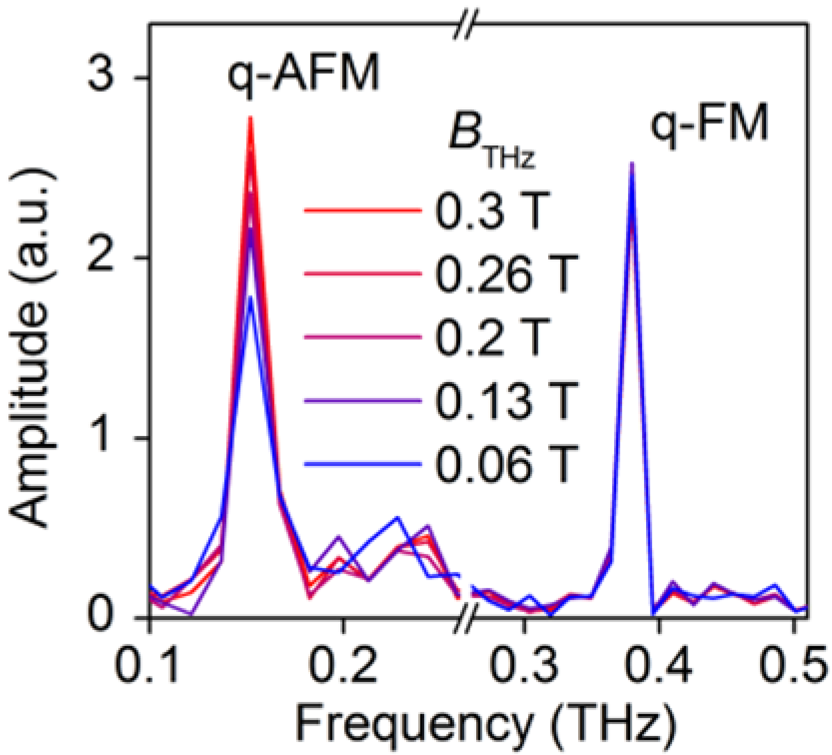


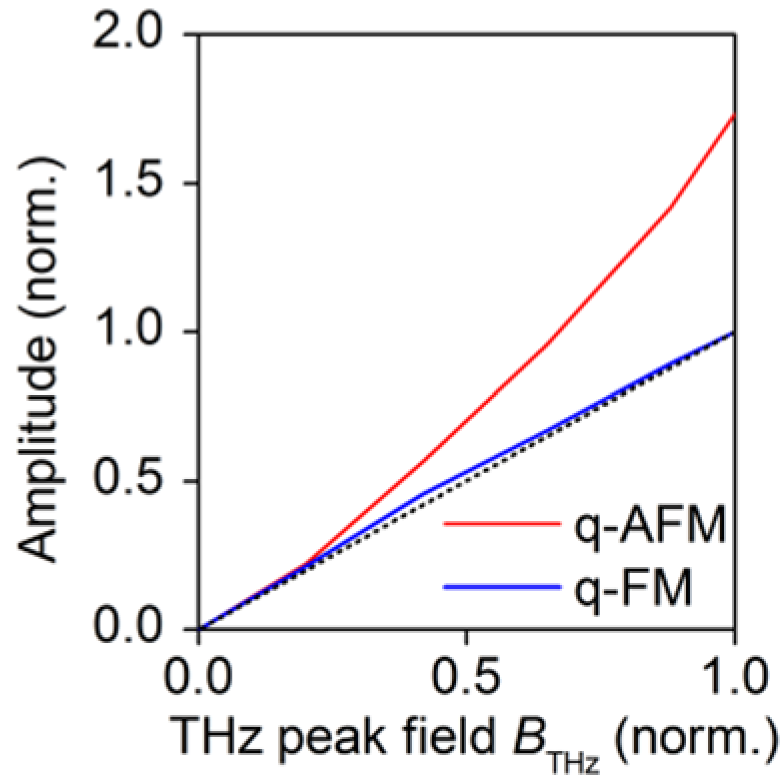
THz-population of electronic orbitals drives spin motion

Anisotropy-driven torque is 8 times larger than Zeeman torque for 0.3 T THz-field

THz-driven anisotropy in $DyFeO_3$

Morin temperature T_M=50 K





Nonlinear excitation of soft q-AFM mode

Summary

✓ **THz magnon-polaritonics**

ACS Photonics

Terahertz Magnon-Polaritons in $TmFeO_3$

Kirill Grishunin,[†] Thomas Huisman,[‡] Guanqiao Li,[‡] Elena Mishina,[†] Theo Rasing,[‡] Alexey V. Kimel,[†,‡] Kailing Zhang,[§] Zuanming Jin,[§] Shixun Cao,[§] Wei Ren,[§] Guo-Hong Ma,[*,§] and Rostislav V. Mikhaylovskiy[*,‡]

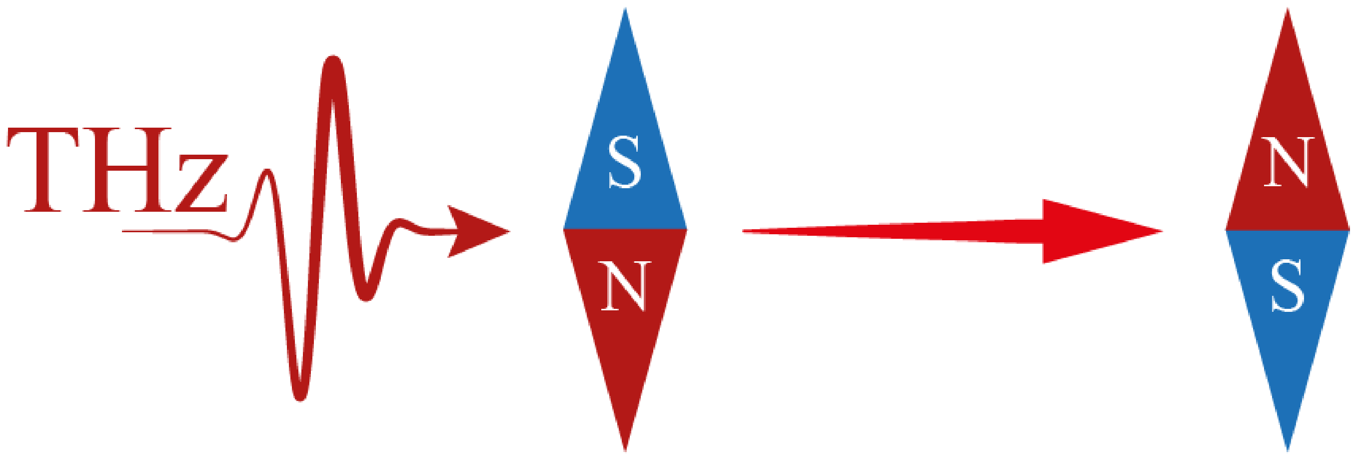
✓ **THz nonlinear spin control**

nature photonics

Nonlinear spin control by terahertz-driven anisotropy fields

S. Baierl[1], M. Hohenleutner[1], T. Kampfrath[2], A. K. Zvezdin[3,4,5], A. V. Kimel[4,6], R. Huber[1*] and R. V. Mikhaylovskiy[6*]

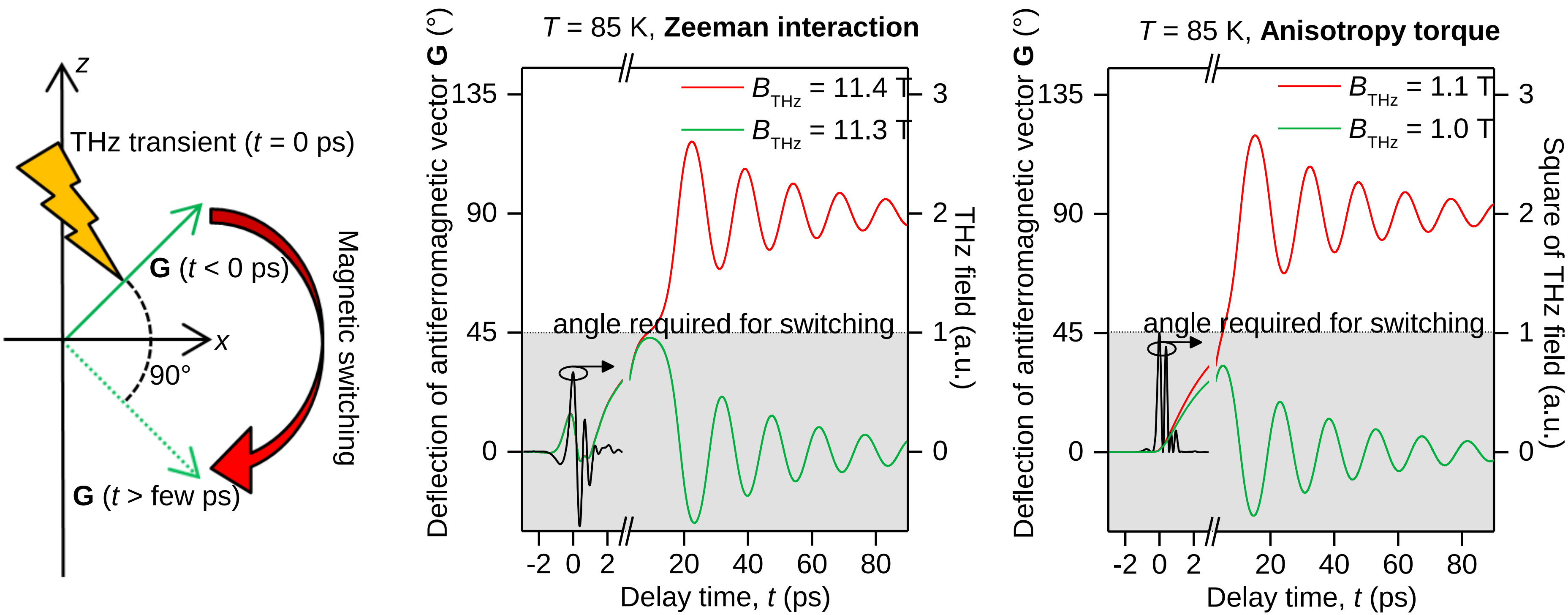
Towards THz-switching



So far: ***nonlinearity with respect to the THz field***

Required: ***nonlinearity in spin dynamics***

Towards THz-switching



How to enhance THz field?

TH
yU
znk

Summary

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Outline

- Introduction: why THz?

- THz linear spin control
  - Magnon-polaritons
  - Internal resonance

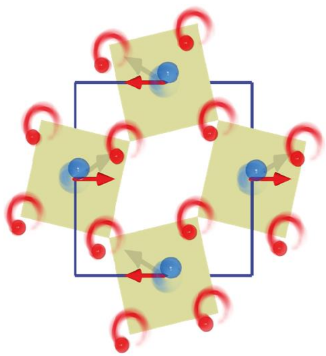
- **THz nonlinear spin control**
  - **THz magneto-phonics**
  - THz-driven anisotropy fields
  - Towards THz spin switching

Electric field driven terahertz control

We propose to employ low-energy electric dipole-active excitations coupled to magnetic order

Nonlinear phononics

IR-active phonons



nature physics

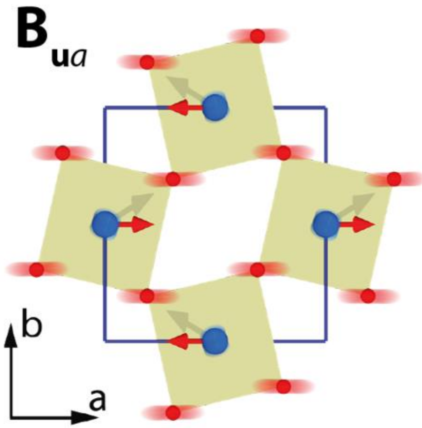
An effective magnetic field from optically driven phonons

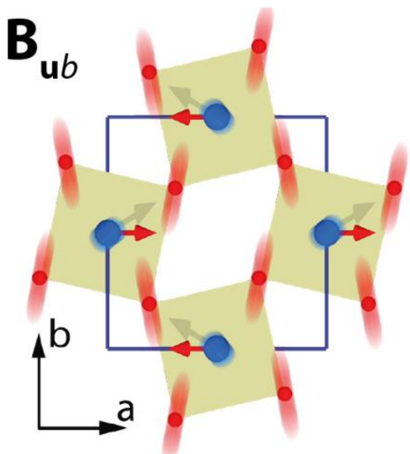
T. F. Nova[1★], A. Cartella[1], A. Cantaluppi[1], M. Först[1], D. Bossini[2†], R. V. Mikhaylovskiy[2], A. V. Kimel[2], R. Merlin[3] and A. Cavalleri[1,4★]

THz nonlinear magneto-phononics

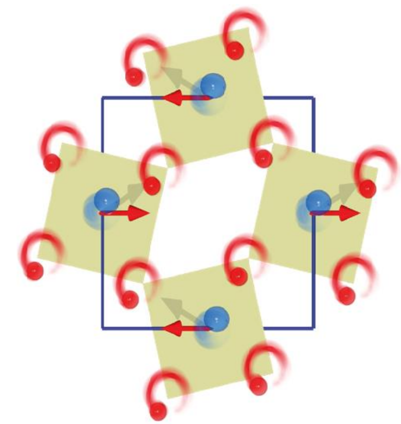
$ErFeO_3$ – orthorombic crystal

Orthogonal phonon modes



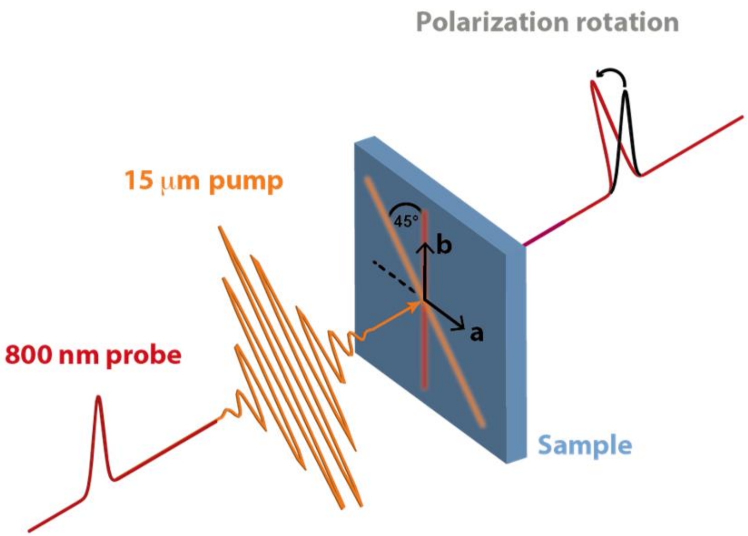


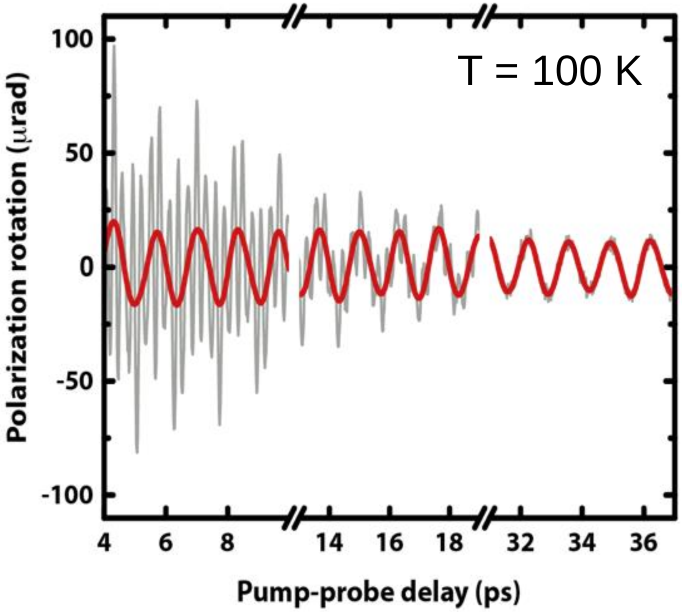
Circular ionic loops

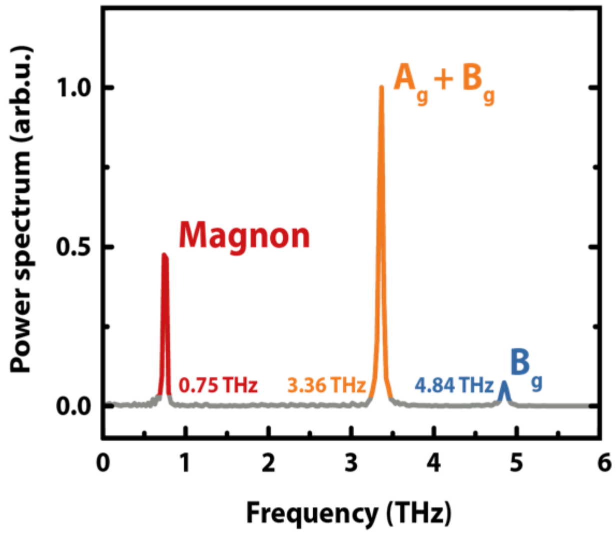


Simultaneous excitation of orthogonal phonons breaks time inversion invariance

THz nonlinear magneto-phononics

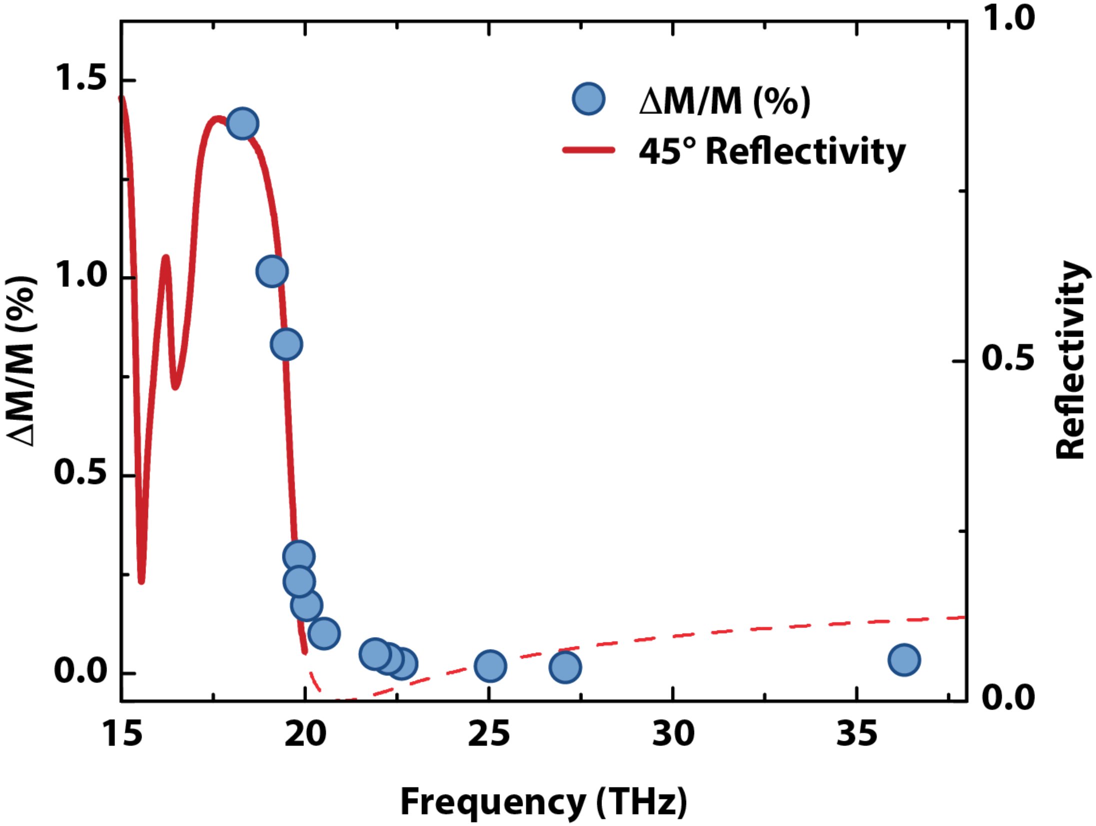






20 THz pump excites quasi-antiferromagnetic resonance

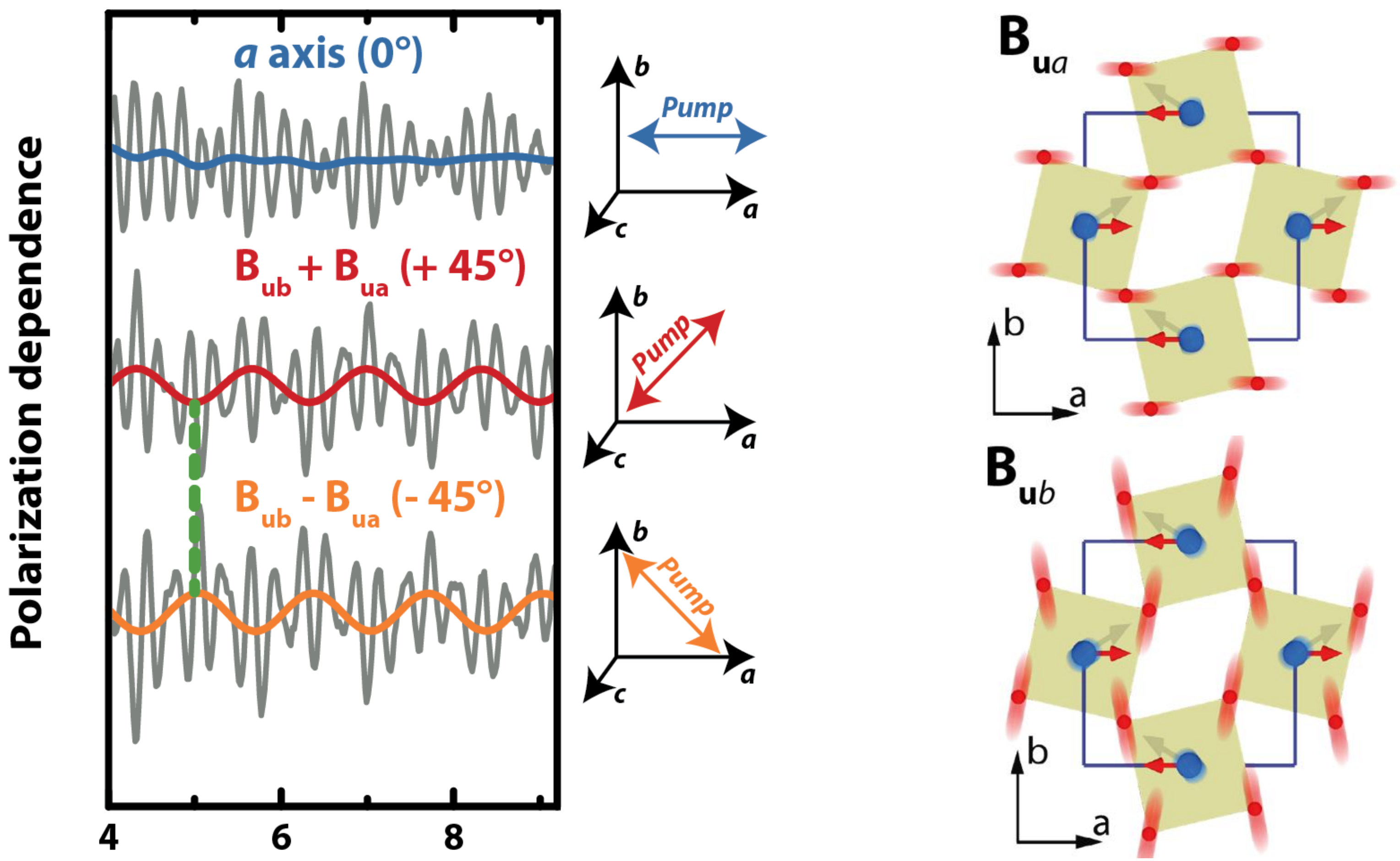
THz nonlinear magneto-phononics



Phonon resonance

Spin deflection scales nonlinearly

THz nonlinear magneto-phononics



Excitation of phonons generates effective magnetic field

Phonon-driven effective field

Phonon-mediated Inverse Faraday effect (ionic impulsive stimulated Raman scattering)?

$$\Phi = i\chi\mathbf{M}\cdot\left[\mathbf{Q}\times\mathbf{Q}^*\right]$$

$$\mathbf{B}_{\mathrm{eff}} = -\frac{\partial\Phi}{\partial\mathbf{M}} = -i\chi\left[\mathbf{Q}\times\mathbf{Q}^*\right]$$

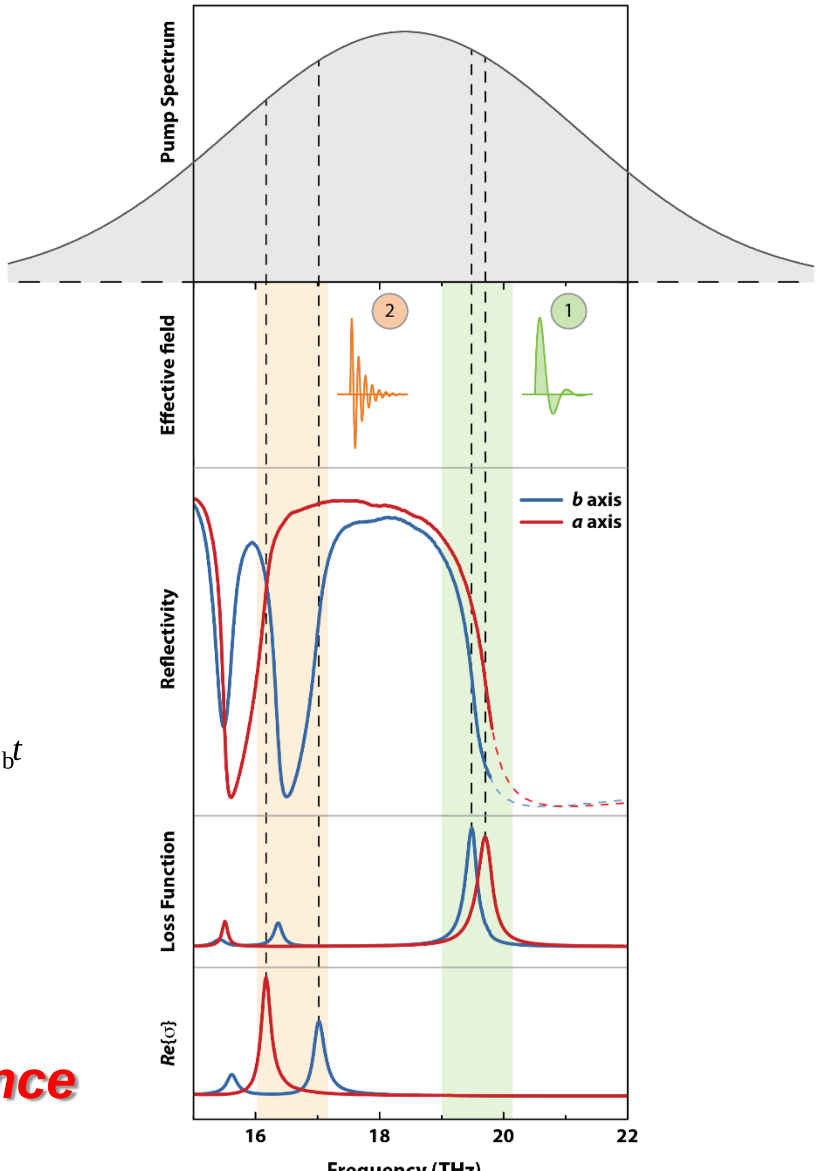
How to create circularly polarized lattice vibration?

$$-i\chi\left[\mathbf{Q}\times\mathbf{Q}^*\right] = -i\chi\left(Q_x Q_y^* - Q_y Q_x^*\right)\mathbf{z}_0$$

$$Q_x = \Theta(t) q_{\mathrm{ua}} e^{i\omega_{\mathrm{ua}}t} e^{-\alpha_{\mathrm{ua}}t}, \quad Q_y = \Theta(t) q_{\mathrm{ub}} e^{i\omega_{\mathrm{ub}}t} e^{-\alpha_{\mathrm{ub}}t}$$

$$\boxed{B_{\mathrm{eff}} = b_0\Theta(t)\sin\left(\omega_{\mathrm{ua}} - \omega_{\mathrm{ub}}\right)t\, e^{-(\alpha_{ua}+\alpha_{ub})t}}$$

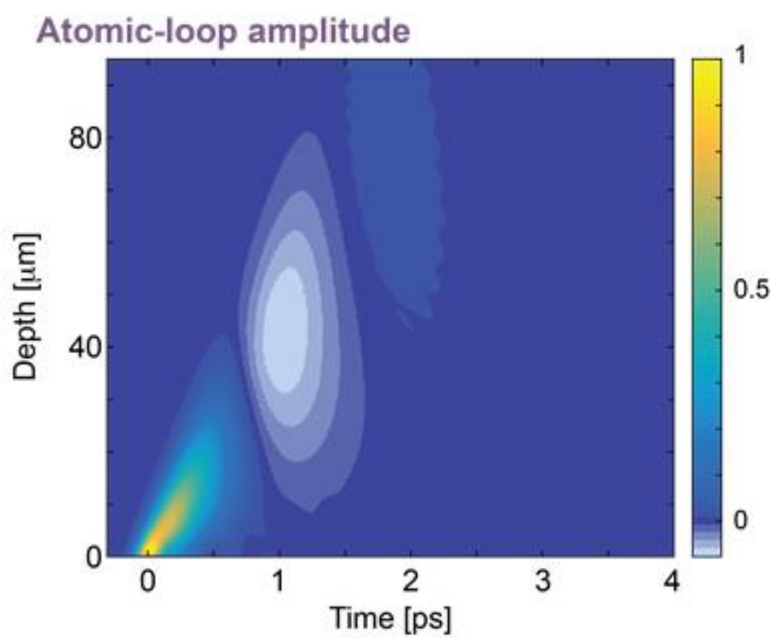
Simultaneous excitation of orthogonal phonons breaks time inversion invariance



Phonon-driven effective field

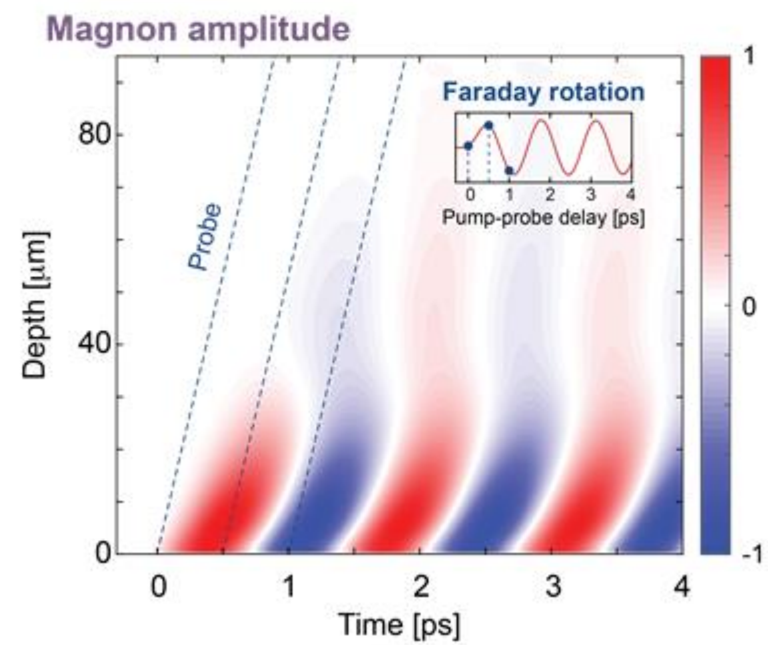
Solve equations of motion for phonon-polaritons

Angular momentum $\mathbf{J} \sim \left[\mathbf{Q} \times \frac{\partial \mathbf{Q}}{\partial t} \right]$



Solve equation of motion for antiferromagnetic vector

$$\mathbf{B}_{\text{eff}} \sim \mathbf{J}$$



Per 1 mJ/cm² $\boxed{B_{\text{eff}} \approx 100 \ \text{Gauss}}$