Terahertz control of magnetic materials

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Knut och Alice Wallenbergs Stiftelse

Vetenskapsrådet

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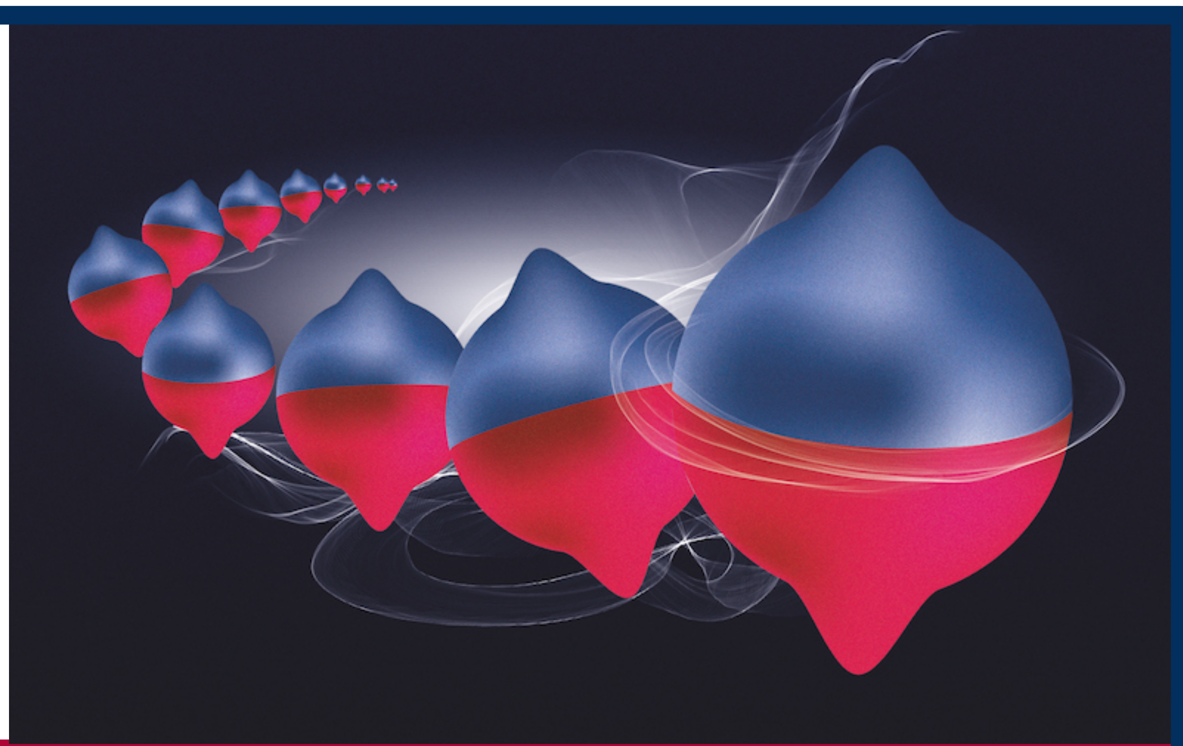

www.magnetic-speed-limit.eu

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Matter manipulation with extreme terahertz light: Progress in the enabling THz technology

Peter Salén [a], Martina Basini [b], Stefano Bonetti [b,c], János Hebling [d,e], Mikhail Krasilnikov [f], Alexey Y. Nikitin [g,h], Georgii Shamuilov [a], Zoltán Tibai [d], Vitali Zhaunerchyk [i], Vitaliy Goryashko [a,*]

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Interaction of THz light with magnetic materials

- Coupling of $\mathbf{H}_{THz}$ with magnetization: $\mathbf{H}_{THz} \times \mathbf{M}$

- Coupling of $\mathbf{E}_{THz}$ with electrons, phonons, electromagnons, ...: $\mathbf{p} \cdot \mathbf{E}_{THz}$

- Heat deposited in the material: $\sim H^2_{THz}$

First THz torque experiments

nature photonics

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nature › nature photonics › letters › article

Published: 21 November 2010

Coherent terahertz control of antiferromagnetic spin waves

Tobias Kampfrath, Alexander Sell, Gregor Klatt, Alexej Pashkin, Sebastian Mährlein, Thomas Dekorsy, Martin Wolf, Manfred Fiebig, Alfred Leitenstorfer & Rupert Huber

Nature Photonics **5**, 31–34 (2011) | Cite this article

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Published: 11 August 2013

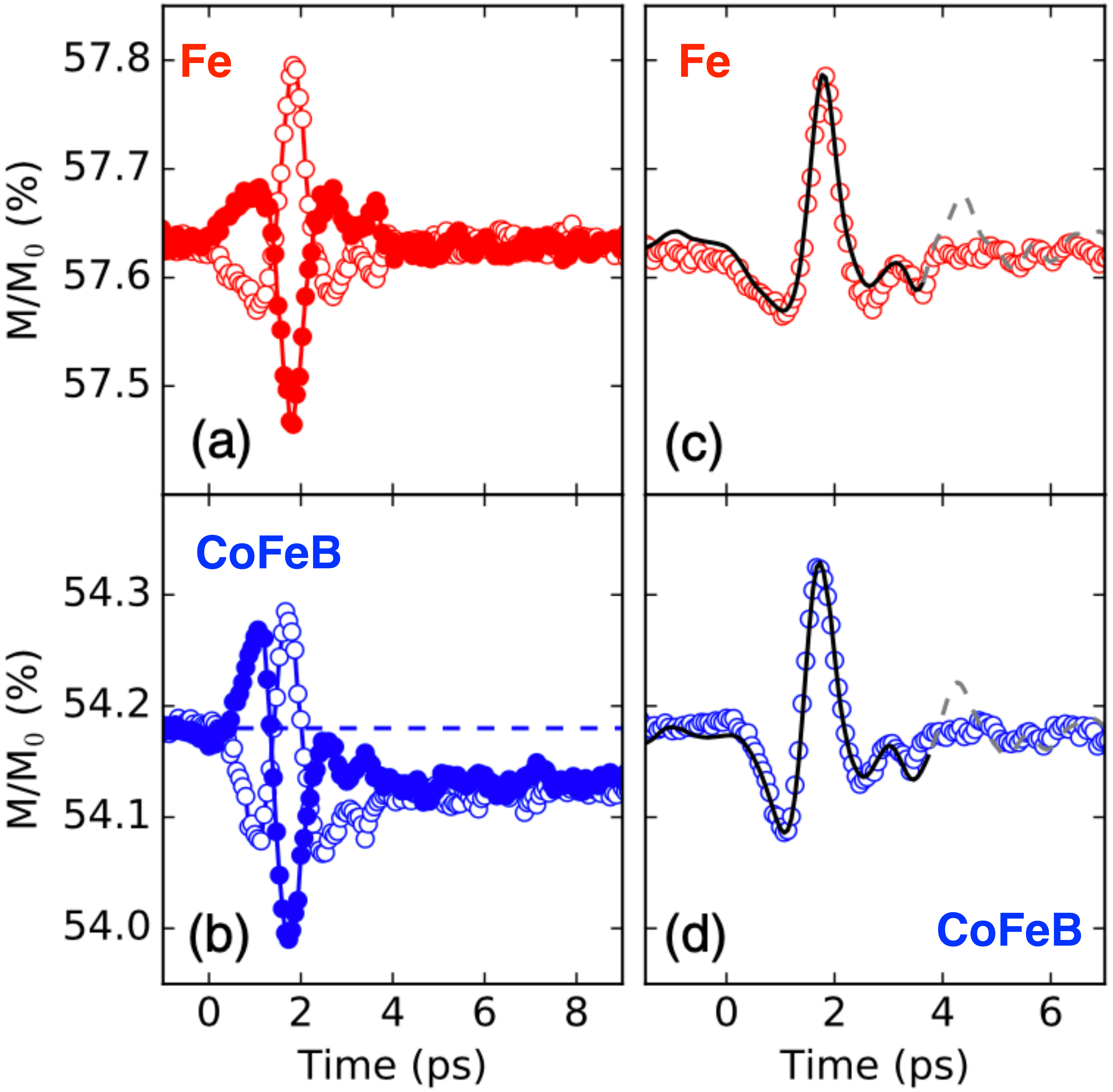
Off-resonant magnetization dynamics phase-locked to an intense phase-stable terahertz transient

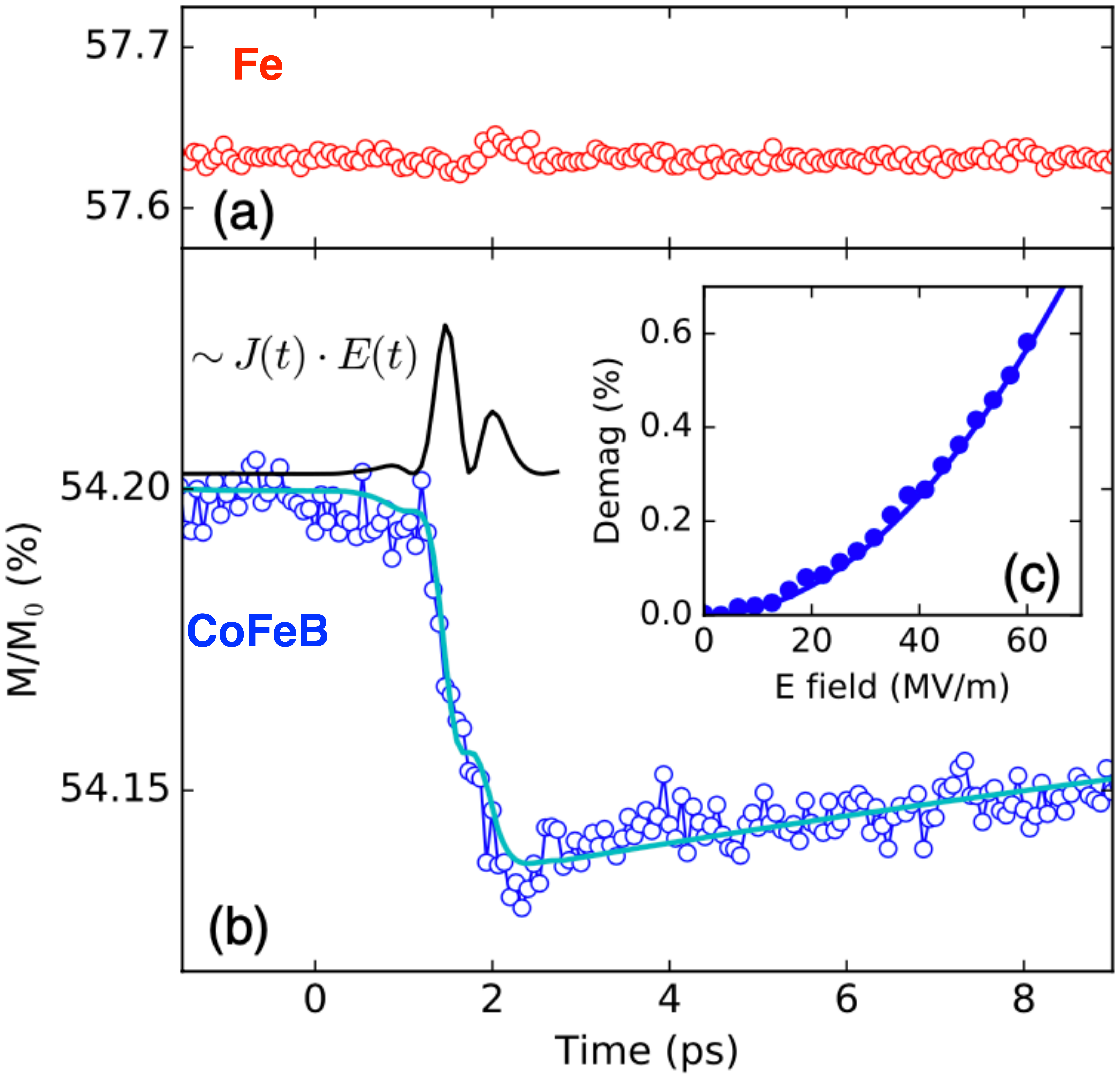
C. Vicario, C. Ruchert, F. Ardana-Lamas, P. M. Derlet, B. Tudu, J. Luning & C. P. Hauri

Nature Photonics **7**, 720–723 (2013) | Cite this article

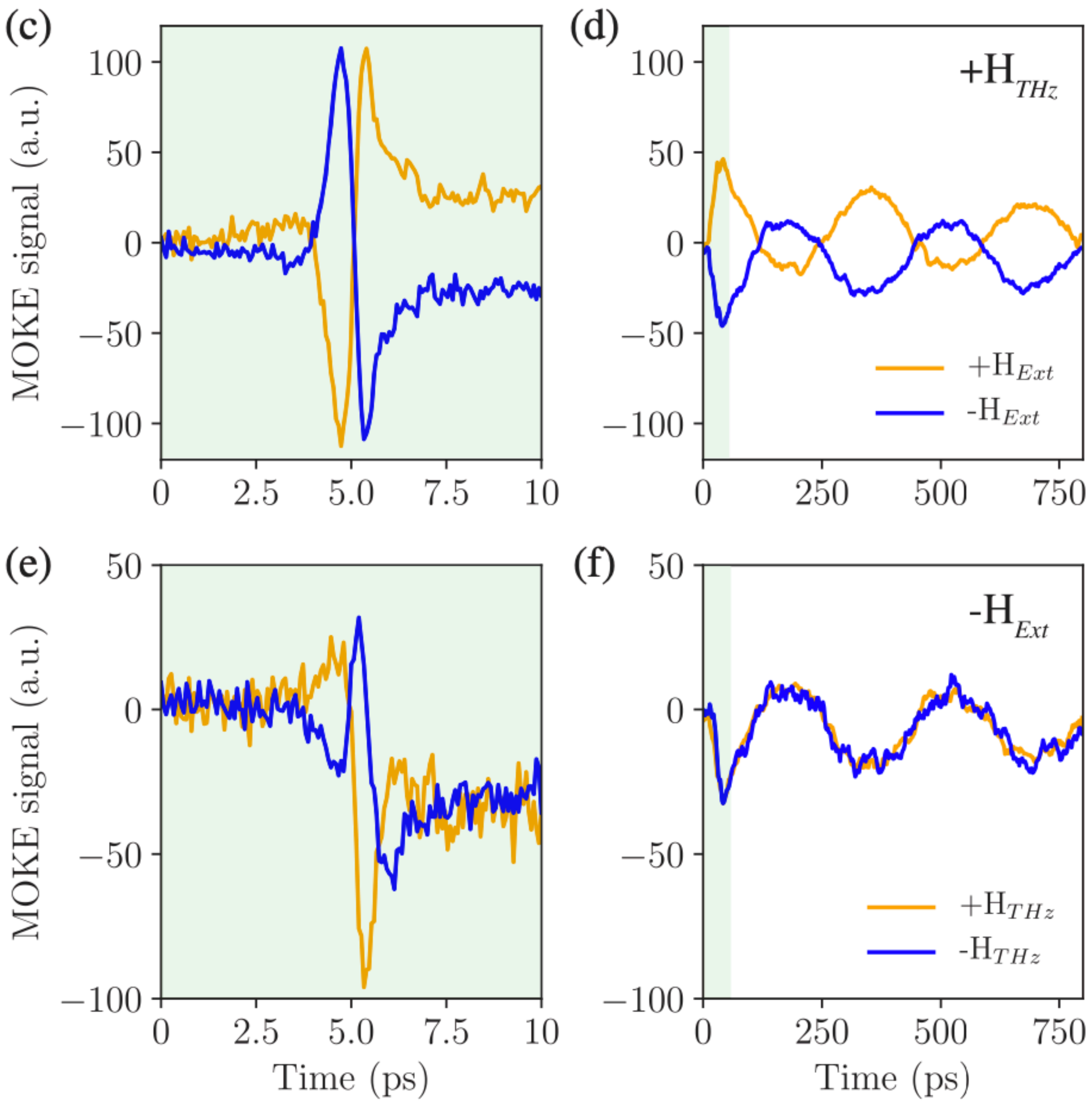
Part one: demagnetization vs coherent response

Torque and demagnetization dynamics



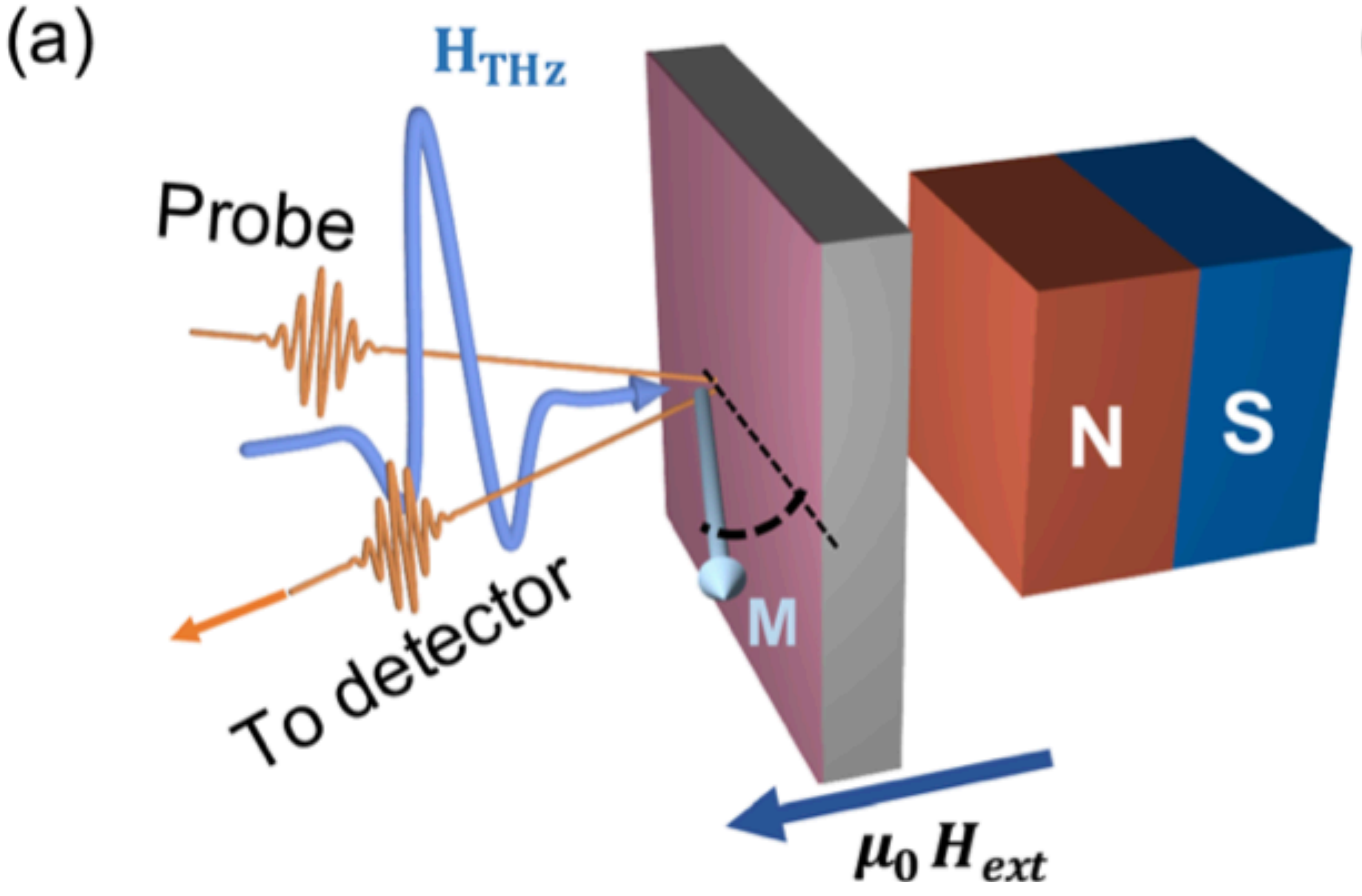


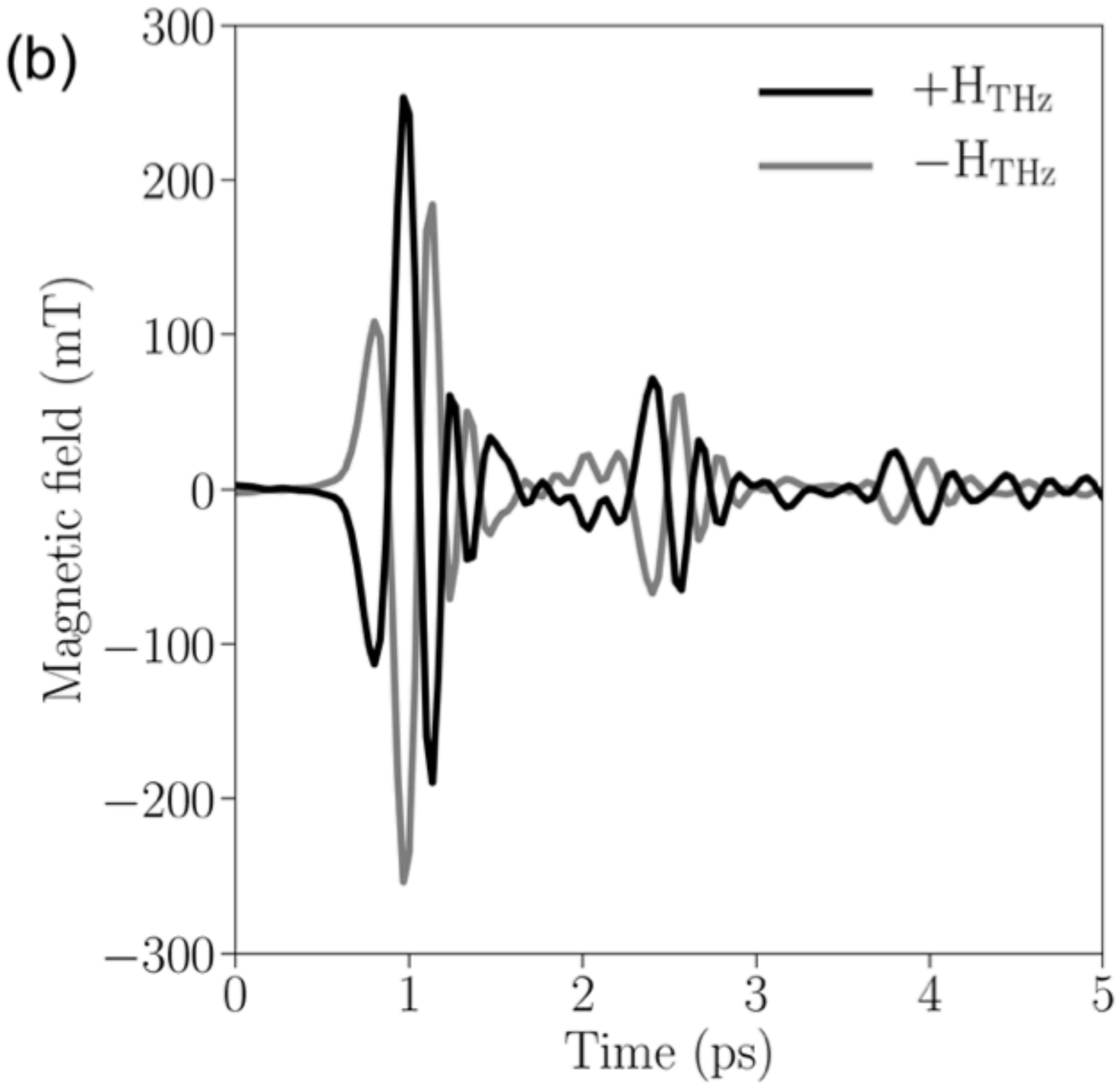
S. Bonetti, M.C. Hoffmann, M.-J. Sher, Z. Chen, S.-H. Yang, M. Samant, S.S.P. Parkin, H.A. Dürr, THz-driven ultrafast spin-lattice scattering in amorphous metallic ferromagnets, *Physical Review Letters* **117**, 087205 (2016)



- Flipping DC magnetic field changes the overall sign of the MOKE (magnetic origin)
- Flippling THz magnetic field reverses coherent response (linear in H), but not FMR precession (triggered by demagnetization proportional to H^2)

M. Hudl Waltin et al., Nonlinear magnetization dynamics driven by strong terahertz fields, *Physical Review Letters* **123**, 197204 (2019)

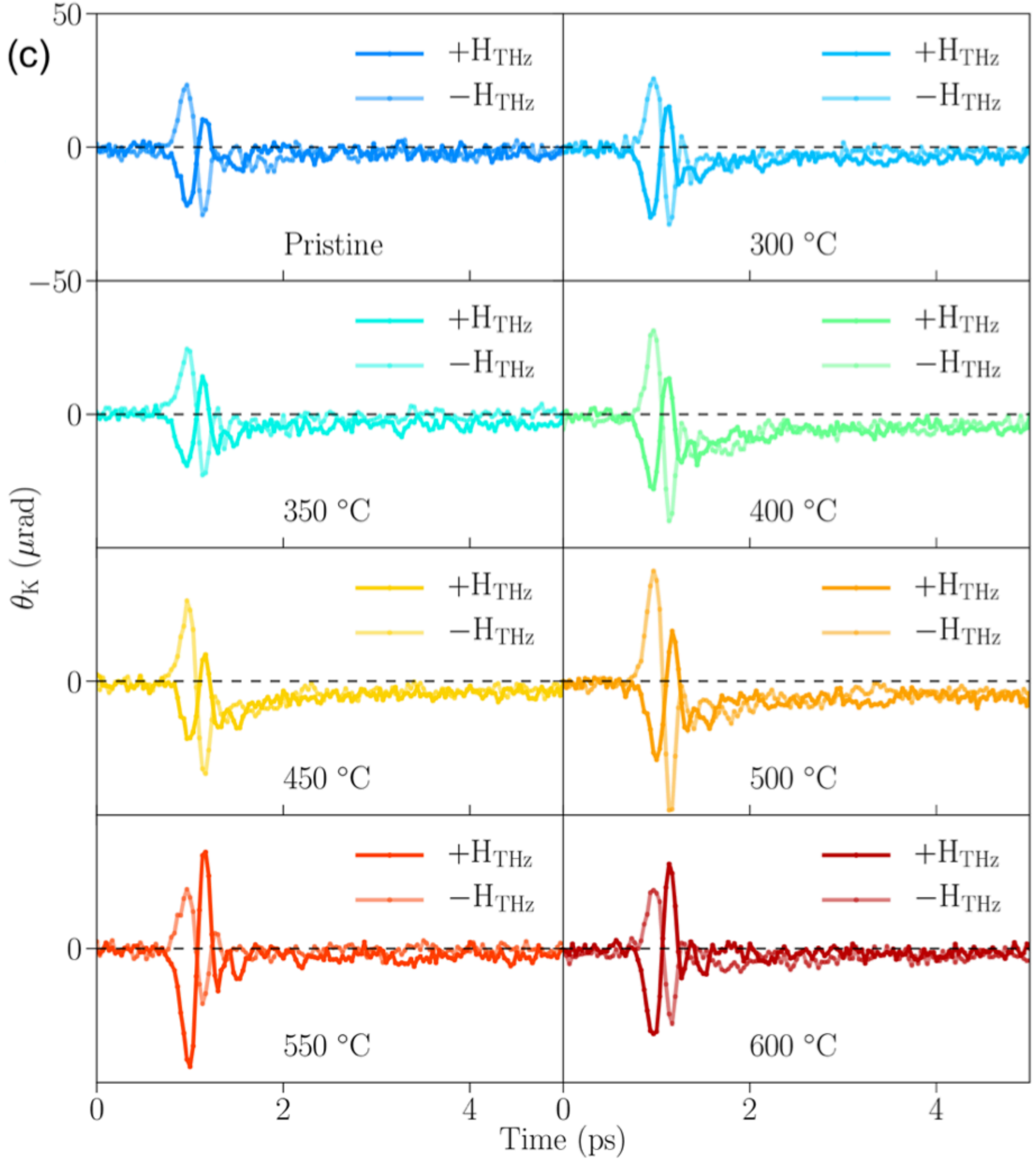


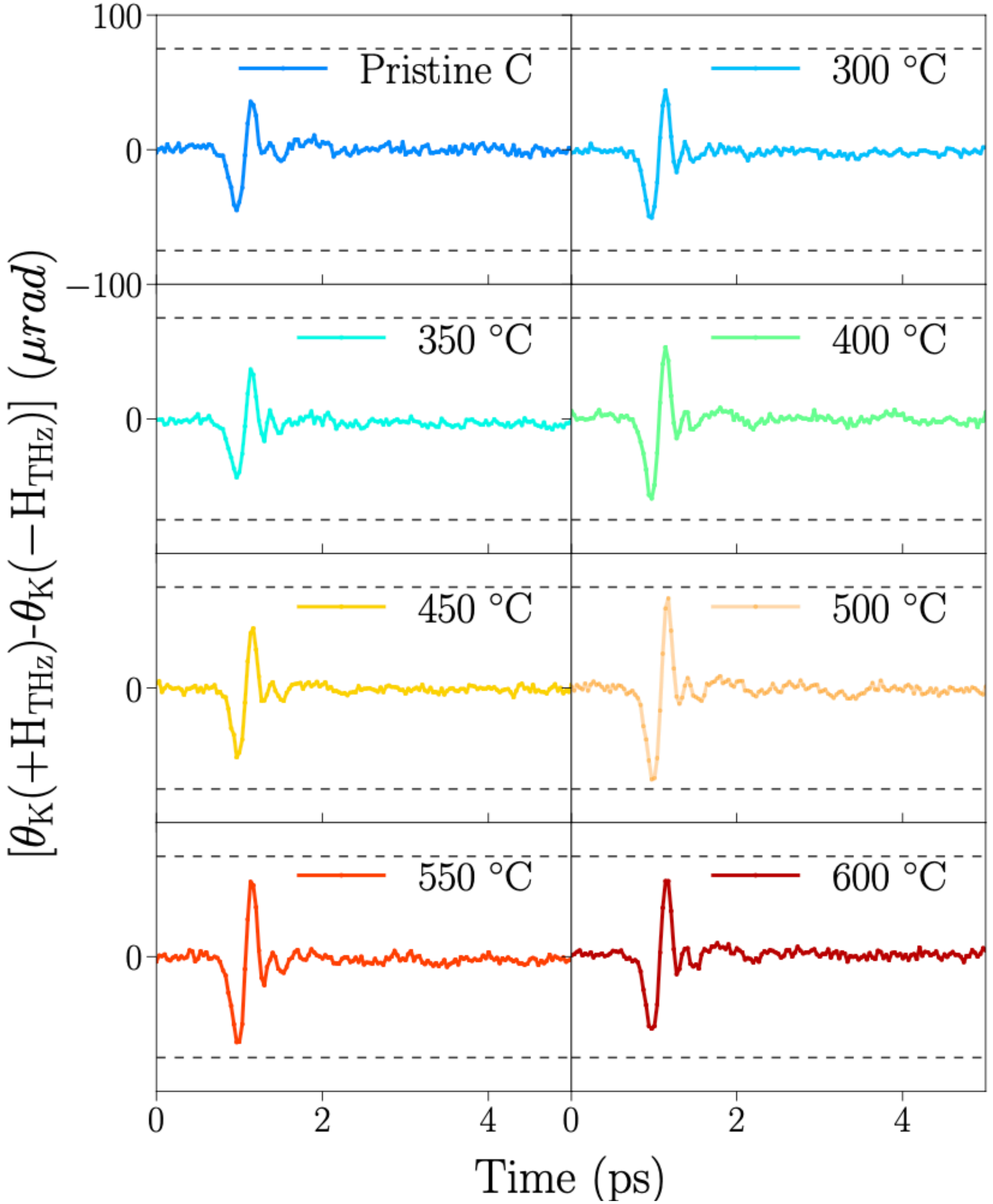


Study THz driven magnetization dynamics in set of CoFeB films annealed at different temperatures

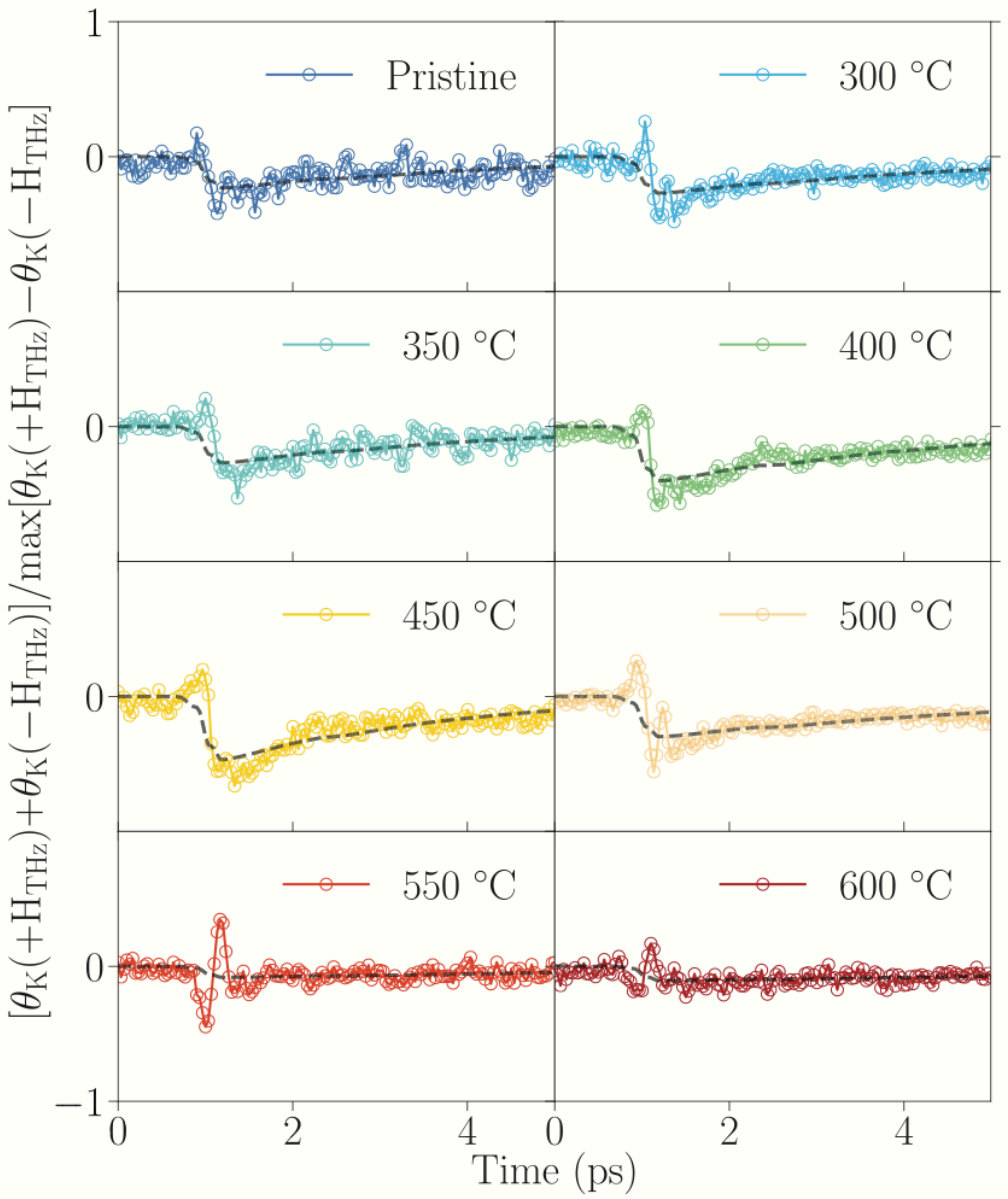
K. Neeraj et al., Terahertz charge and spin transport in metallic ferromagnets: the role of crystalline and magnetic order, *Applied Physics Letters* **120**, 102406 (2022)

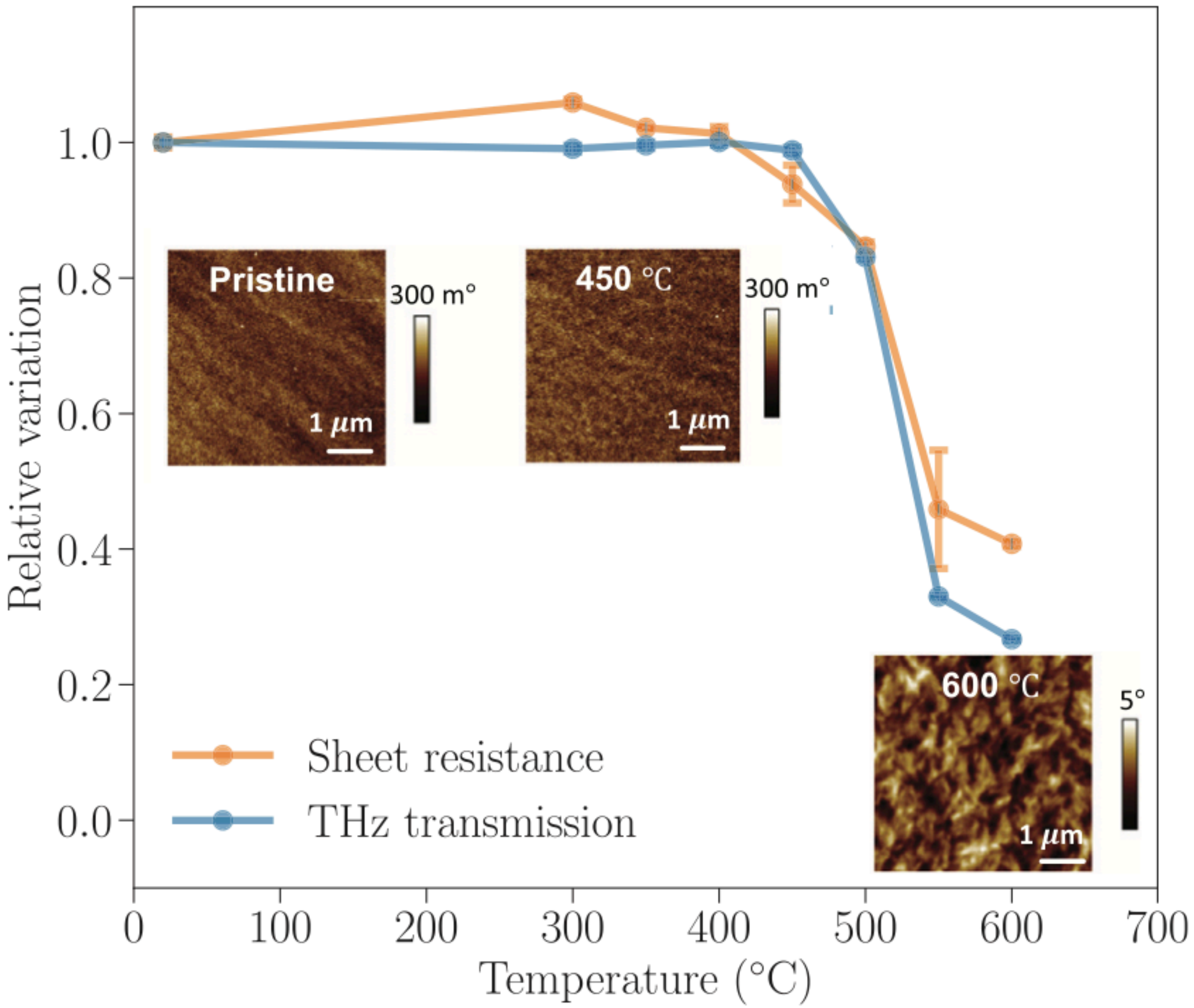
Total and difference response



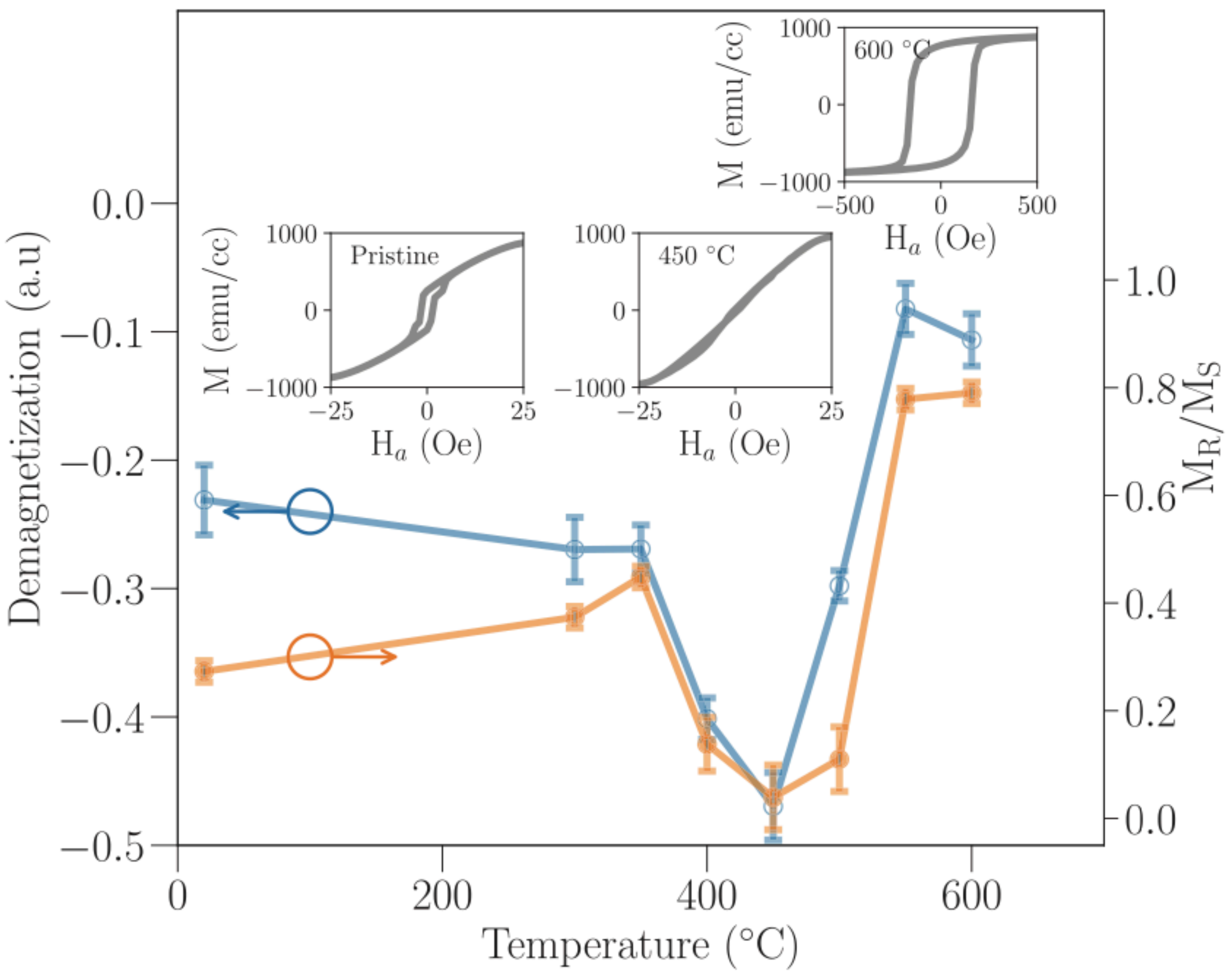


Sum response and transmission / resistivity





K. Neeraj et al., Terahertz charge and spin transport in metallic ferromagnets: the role of crystalline and magnetic order, *Applied Physics Letters* **120**, 102406 (2022)



- Demagnetization largest where remanence is minimum
- Overall demagnetization amplitude follow the strength of the in-plane anisotropy of the sample
- Behavior not observed when using NIR pump

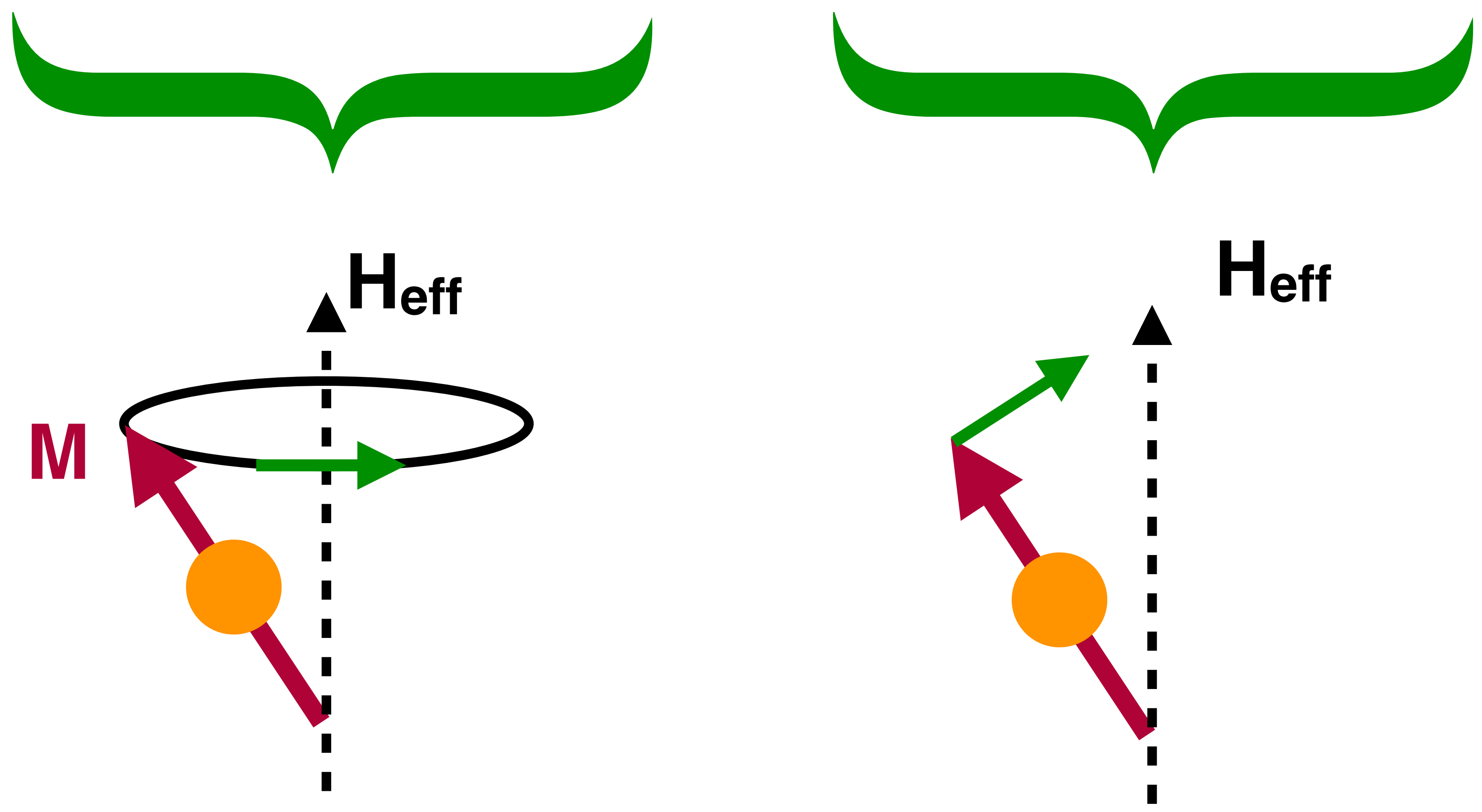
K. Neeraj et al., Terahertz charge and spin transport in metallic ferromagnets: the role of crystalline and magnetic order, *Applied Physics Letters* **120**, 102406 (2022)

Part two:
nutating magnets

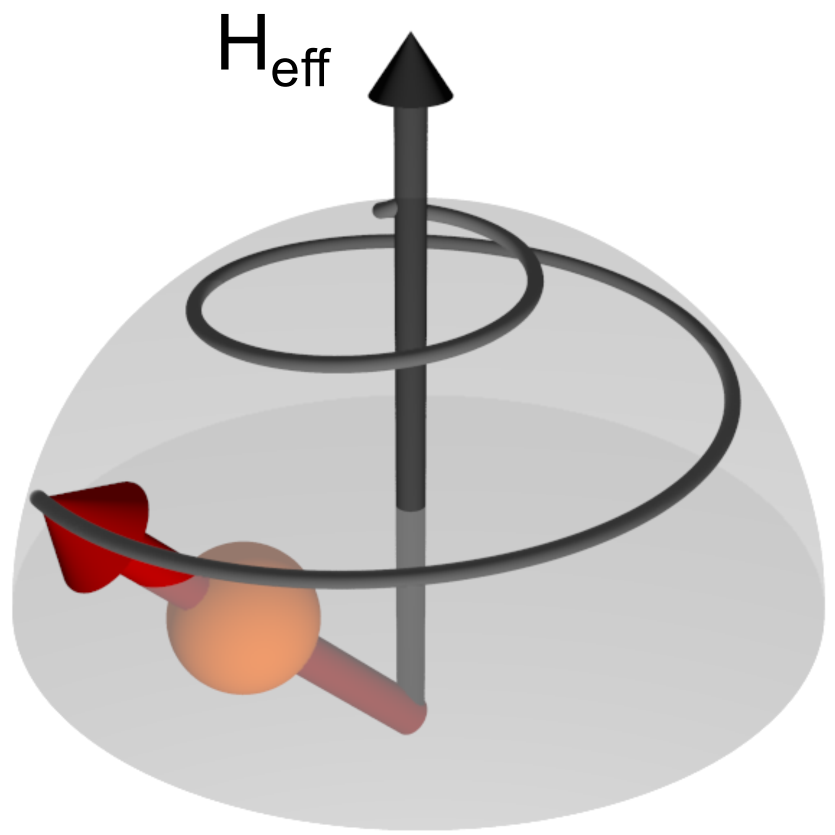
The LLG equation

- Landau-Lifshitz-Gilbert equation: dynamics of (spin) angular momentum

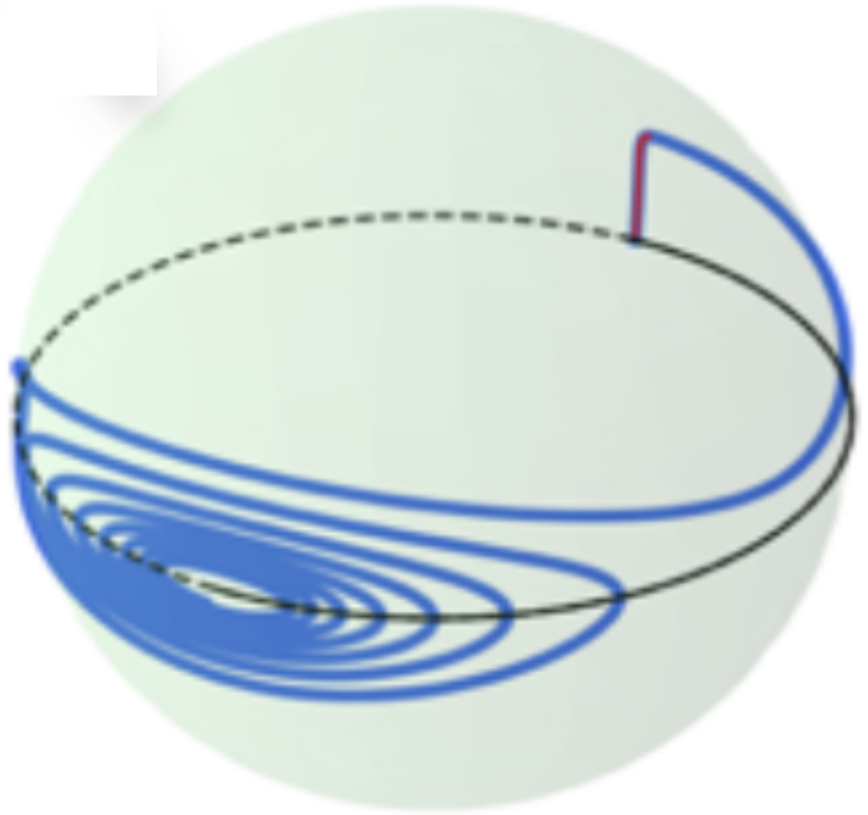
$$\frac{dM}{dt} = -\gamma\, M \times H_{eff} + \alpha M \times \frac{dM}{dt}$$



Precession



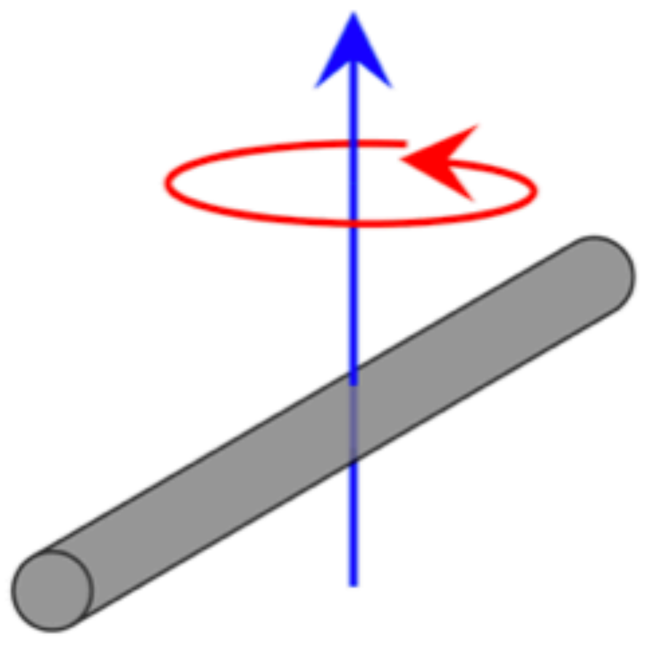
Switching



- LLG equation: unphysical inertia tensor. Gilbert, the "G" in the equation, noticed it:

Gilbert (2004): "I was unable to conceive of a physical object with an inertial tensor of this kind"

$$I = \begin{pmatrix} \cancel{I_1}^{\,0} & 0 & 0 \\ 0 & \cancel{I_2}^{\,0} & 0 \\ 0 & 0 & I_3 \end{pmatrix} \qquad I = \begin{pmatrix} ml^2/12 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & ml^2/12 \end{pmatrix}$$



- How can it be wrong if it is being used every day in data centers?

- Degrees of freedom separated by energy scales orders of magnitude apart

J.-E. Wegrowe and M.-C. Ciornei, "Magnetization dynamics, gyromagnetic relation, and inertial effects", *American Journal of Physics* **80**, 607 (2012)

PREFACE

THE present volume in the *Course of Theoretical Physics* deals with the theory of electromagnetic fields in matter and with the theory of the macroscopic electric and magnetic properties of matter. These theories include a very wide range of topics, as may be seen from the Contents.

In writing this book we have experienced considerable difficulties, partly because of the need to make a selection from the extensive existing material, and partly because the customary exposition of many topics to be included does not possess the necessary physical clarity, and sometimes is actually wrong. We realise that our own treatment still has many defects, which we hope to correct in future editions.

We are grateful to Professor V. L. GINZBURG, who read the book in manuscript and made some useful comments. I. E. DZYALOSHINSKIĬ and L. P. PITAEVSKIĬ gave great help in reading the proofs of the Russian edition. Thanks are due also to Dr SYKES and Dr BELL, who not only carried out excellently the arduous task of translating the book, but also made some useful comments concerning its contents.

L. D. LANDAU
E. M. LIFSHITZ

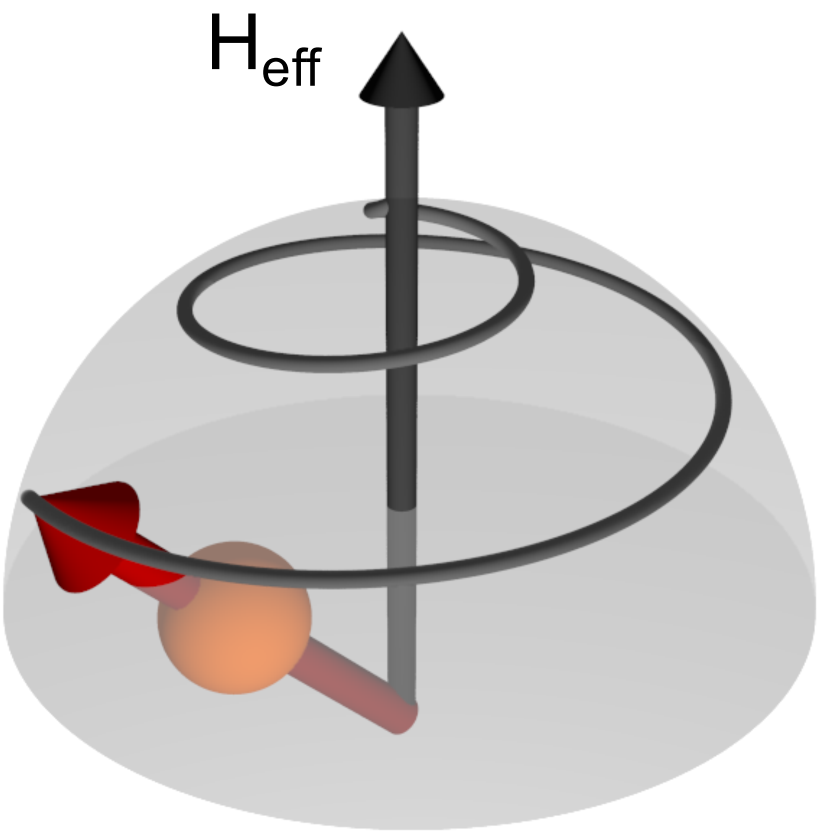
Moscow
June, 1959

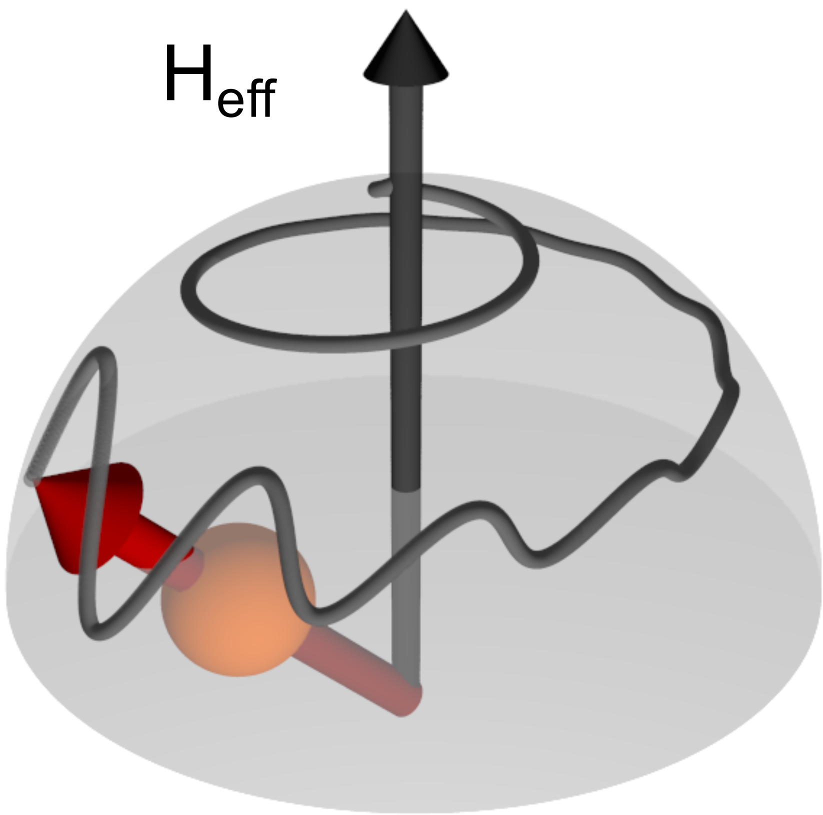
The inertial LLG (iLLG)

Angular momentum relaxation time

$$\frac{d\mathbf{M}}{dt} = -\gamma\,\mathbf{M} \times \mathbf{H}_{\mathrm{eff}} + \alpha\,\mathbf{M} \times \left(\frac{d\mathbf{M}}{dt} + \tau\frac{d^2\mathbf{M}}{dt^2}\right)$$

J.-E. Wegrowe and M.-C. Ciornei, "Magnetization dynamics, gyromagnetic relation, and inertial effects", *American Journal of Physics* **80**, 607 (2012)





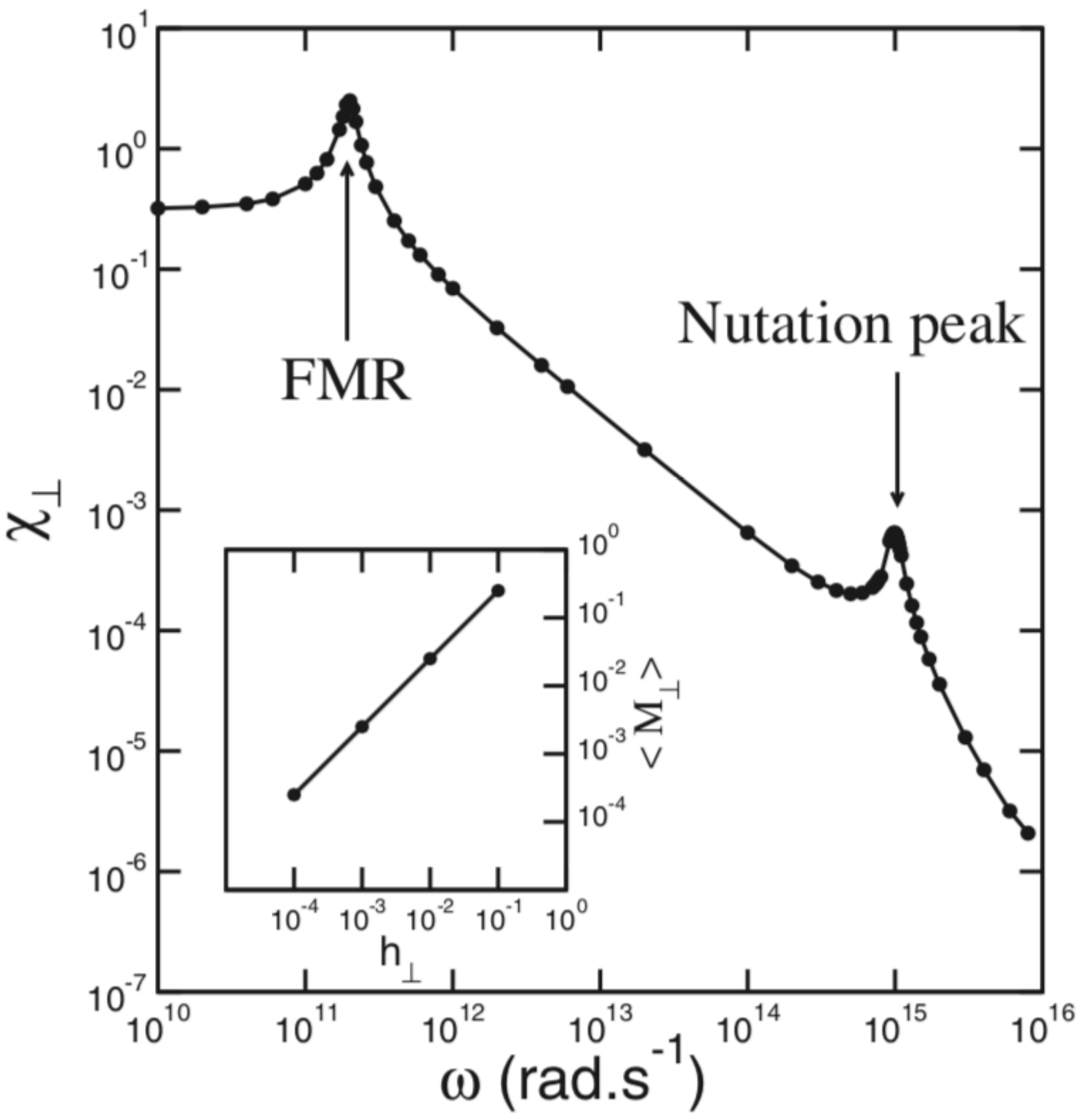
K. Neeraj et al., "Inertial spin dynamics in ferromagnets", *Nature Physics* **17**, 245 (2020)

$$\mathcal{H} = c\underline{\boldsymbol{\alpha}} \cdot (\boldsymbol{p} - e\boldsymbol{A}) + (\underline{\beta} - \underline{\mathbb{1}})mc^2 + V\underline{\mathbb{1}}$$

$$\mathcal{H}^S = -\frac{e}{m}\boldsymbol{S}\cdot\boldsymbol{B} + \frac{e}{2m^3c^2}\boldsymbol{S}\cdot\boldsymbol{B}\left[p^2 - 2e\boldsymbol{A}\cdot\boldsymbol{p} + \frac{3e^2}{2}A^2\right]$$
$$-\frac{e}{2m^2c^2}\boldsymbol{S}\cdot[\boldsymbol{E}_{\mathrm{tot}} \times (\boldsymbol{p} - e\boldsymbol{A})] + \frac{ie\hbar}{4m^2c^2}\boldsymbol{S}\cdot\partial_t\boldsymbol{B}$$
$$+\frac{e\hbar^2}{8m^3c^4}\boldsymbol{S}\cdot\partial_{tt}\boldsymbol{B}$$

Ritwik Mondal, Marco Berritta, Ashis K. Nandy, and Peter M. Oppeneer, *Phys. Rev. B* **96**, 024425 (2017)

Susceptibility spectrum with inertial LLG equation

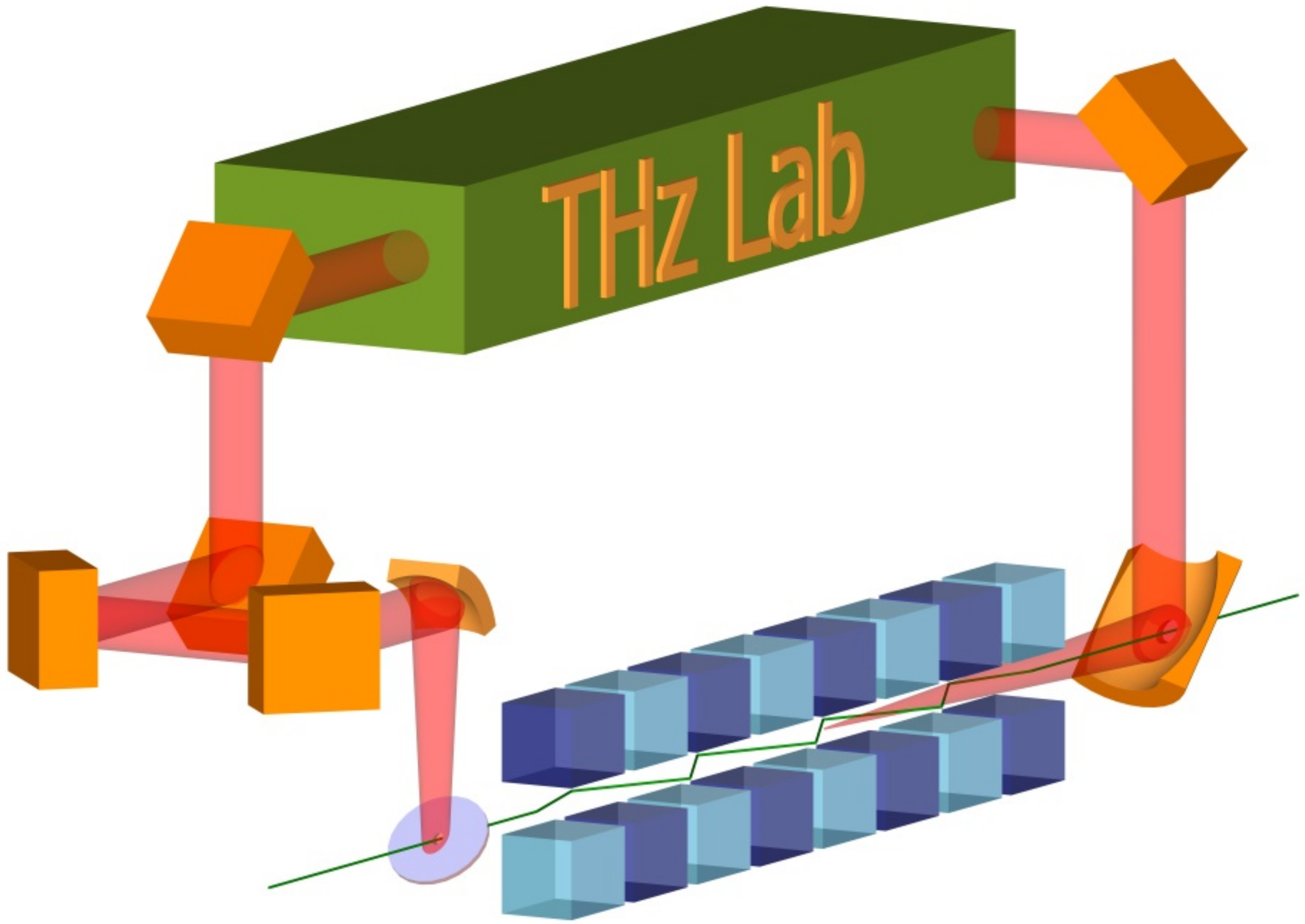


- 2017: met Jean-Eric at the Ultrafast Magnetism Conference in Kaiserslautern

- In 2014 saw unexplained oscillations at 0.3 THz in a ferromagnet driven by THz fields

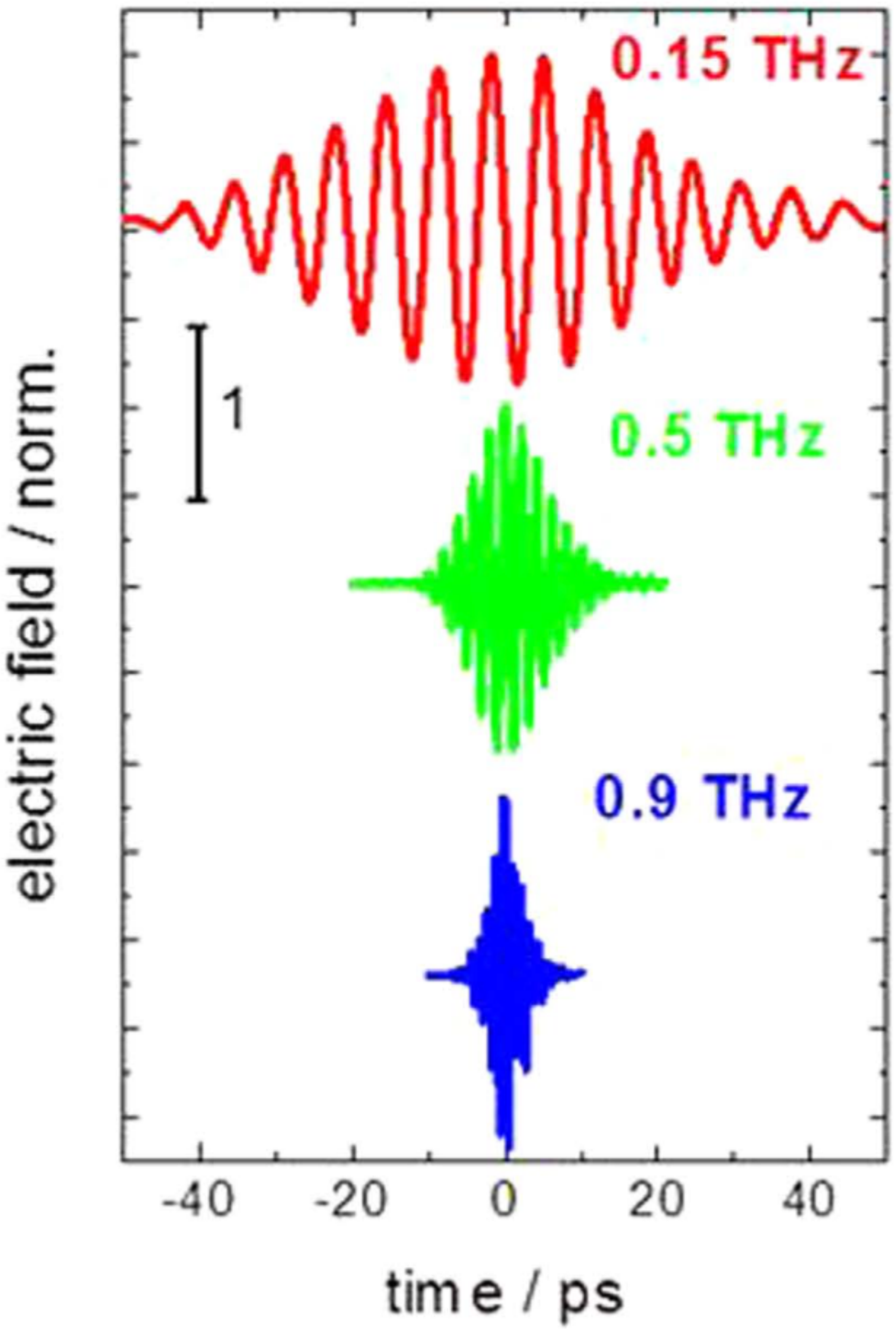
- Idea: ***forced oscillator experiment***. Facility TELBE was starting operation.

Terahertz spectroscopy at TELBE

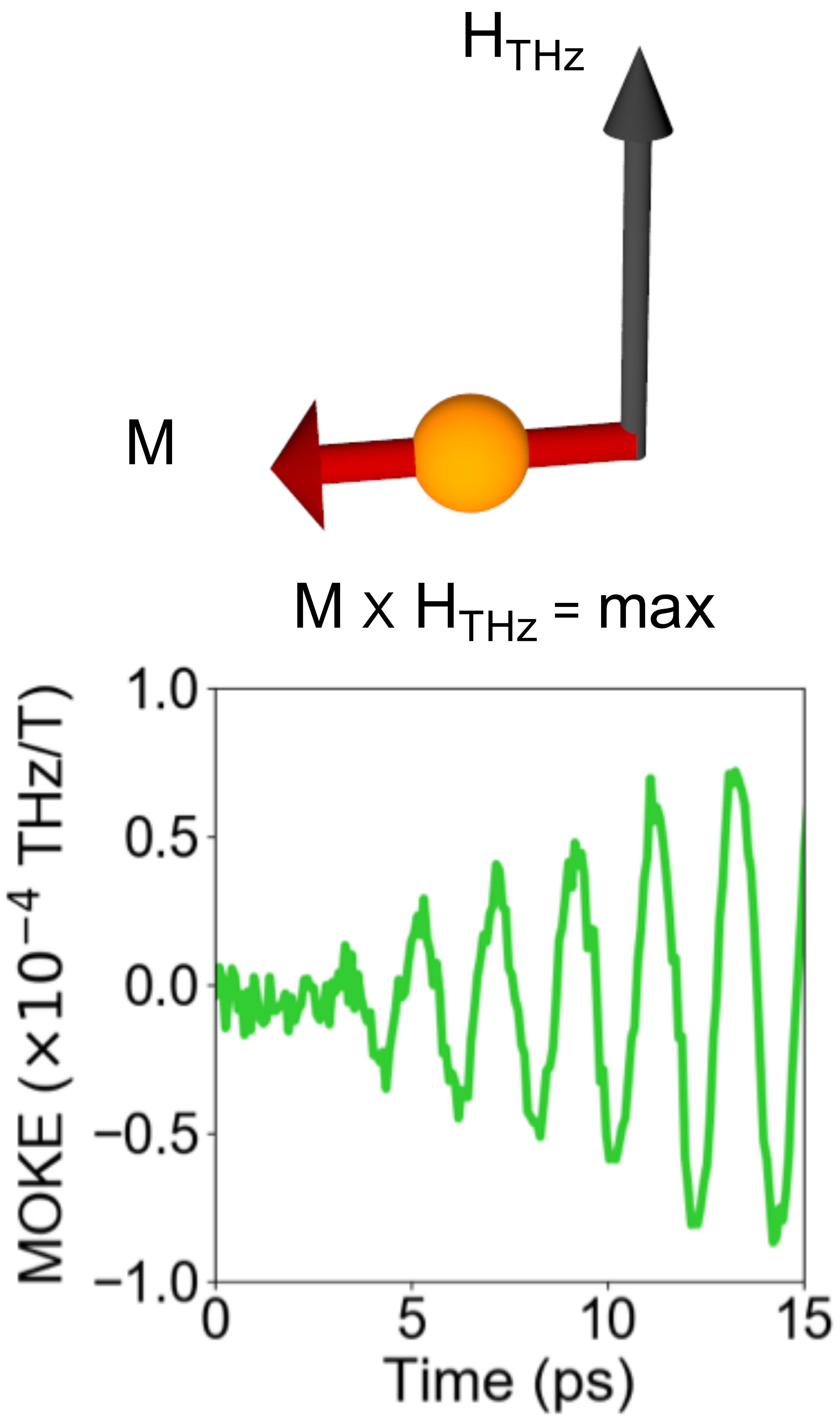


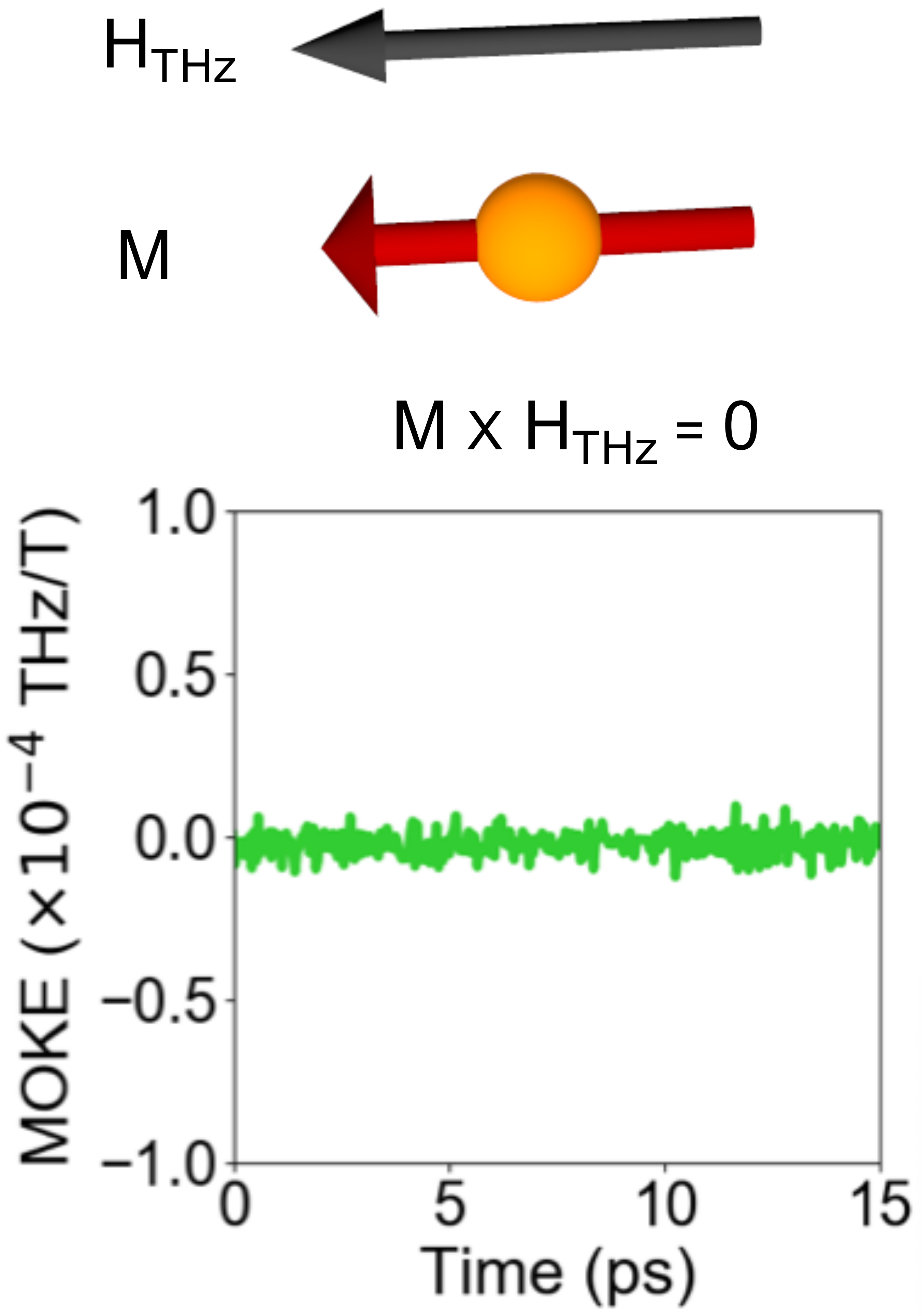
0.1 - 1.2 THz, 1 µJ

B. Green et al., "High-Field High-Repetition-Rate Sources for the Coherent THz Control of Matter", *Scientific Reports* **6**, 22256 (2016)



Check magnetic torque dynamics

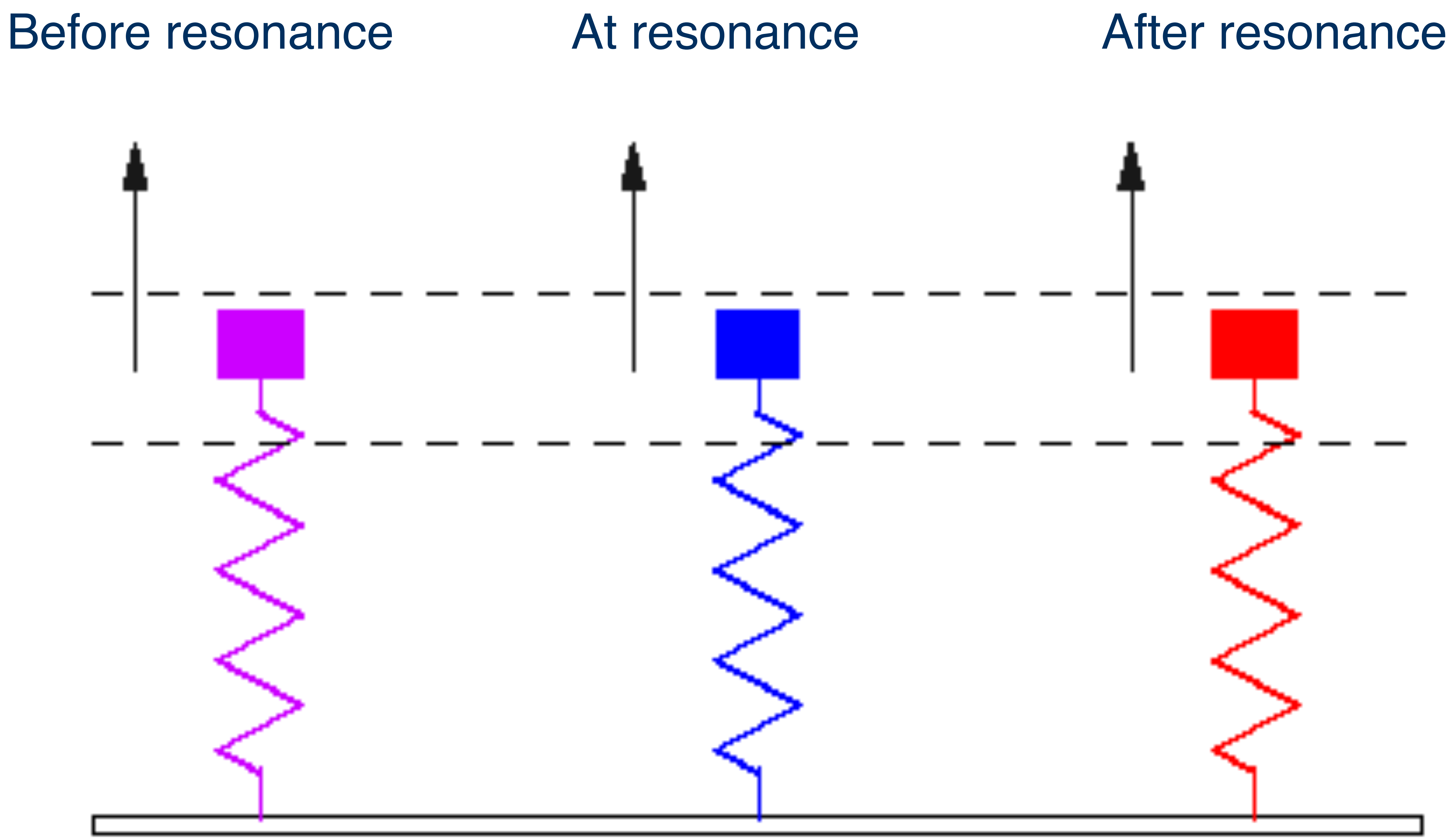




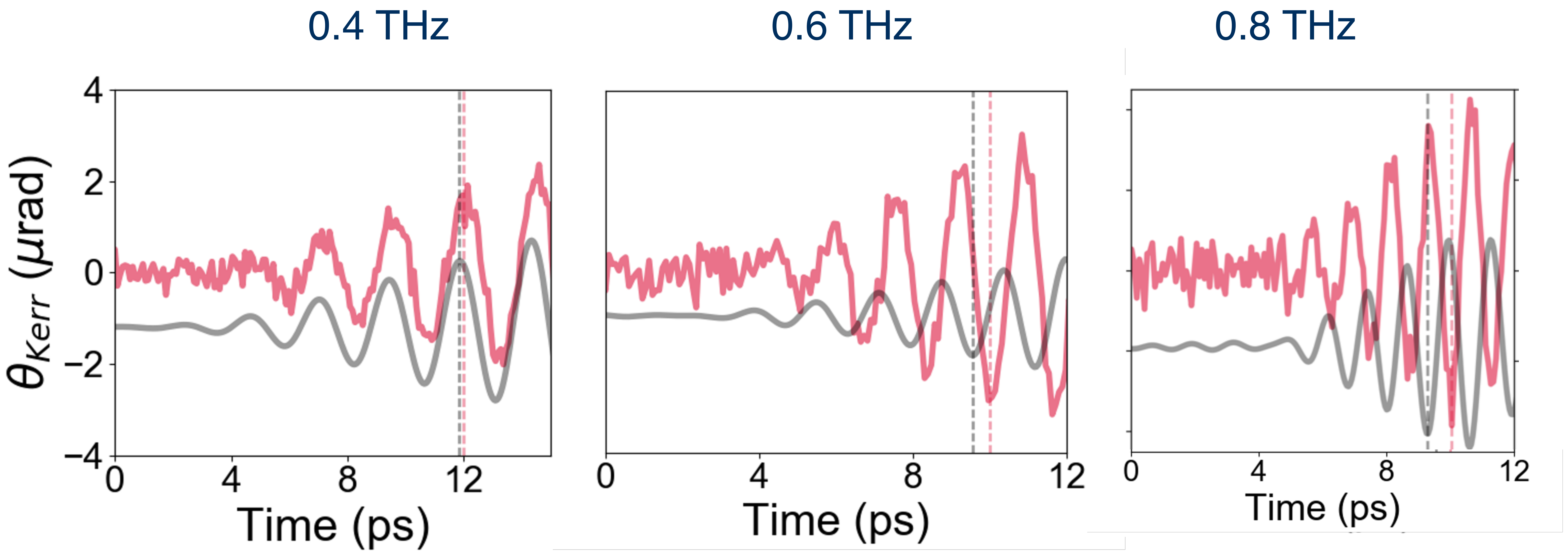
K. Neeraj, N. Awari, S. Kovalev, D. Polley, N.Z. Hagström, S.S.P. Kanth Arekapudi, A. Semisalova, K. Lenz, B. Green, J.-C. Deinert, I. Ilyakov, M. Chen, M. Bawatna, V. Scalera, M. d'Aquino, C. Serpico, O. Hellwig, J.-E. Wegrowe, M. Gensch, **S. Bonetti**, Inertial spin dynamics in ferromagnets, *Nature Physics* **17**, 245 (2020)

Forced Lorentz oscillator

- Largest ***amplitude*** of response when driving force has same frequency of intrinsic resonance
- ***Phase*** shift between driving force and oscillator varies monotonously with frequency (90 degrees at resonance)

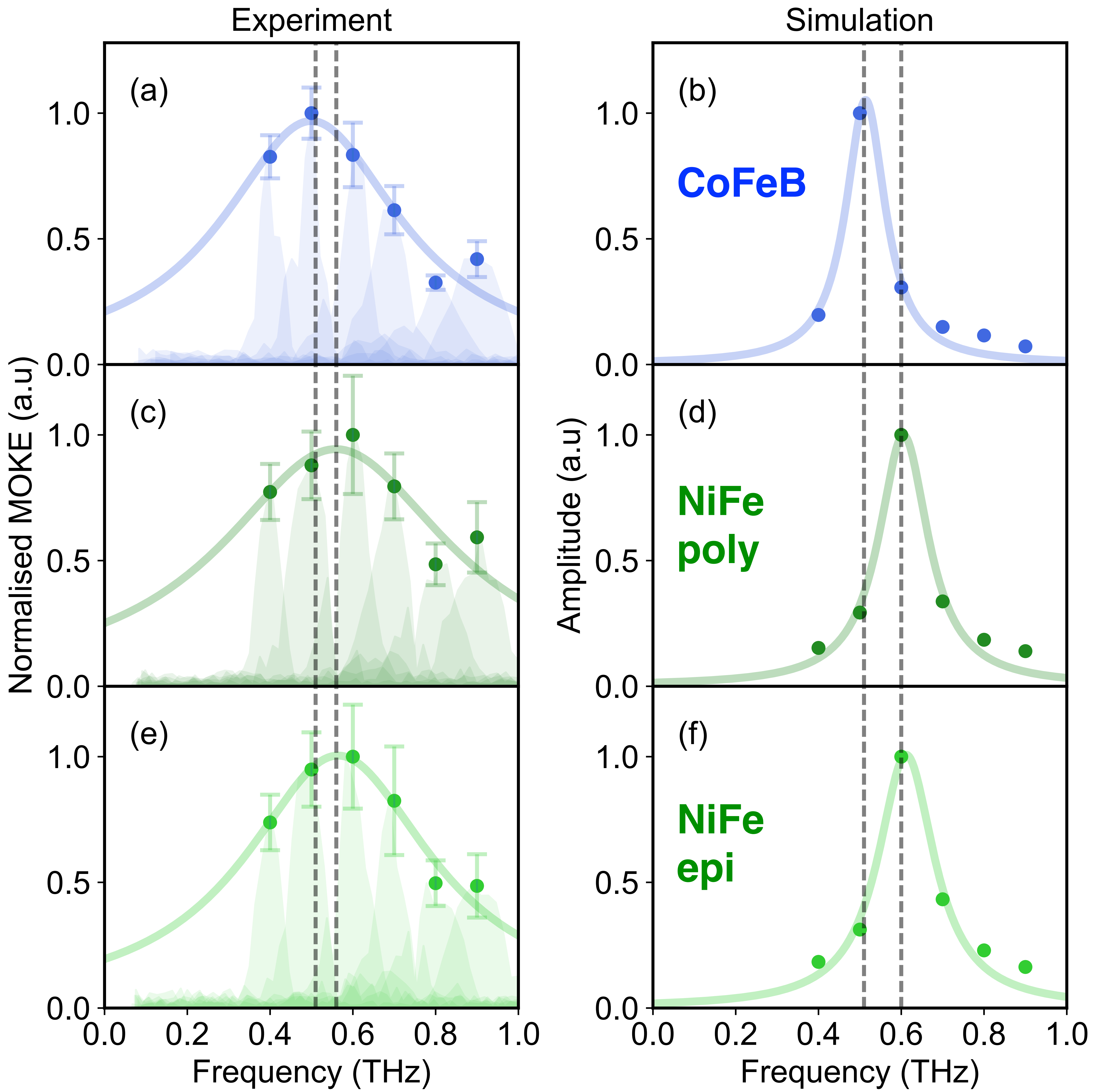


https://www.acs.psu.edu/drussell/Demos/SHO/mass-force.html



K. Neeraj et al., "Inertial spin dynamics in ferromagnets", *Nature Physics* **17**, 245 (2020)

FFT summary for three samples

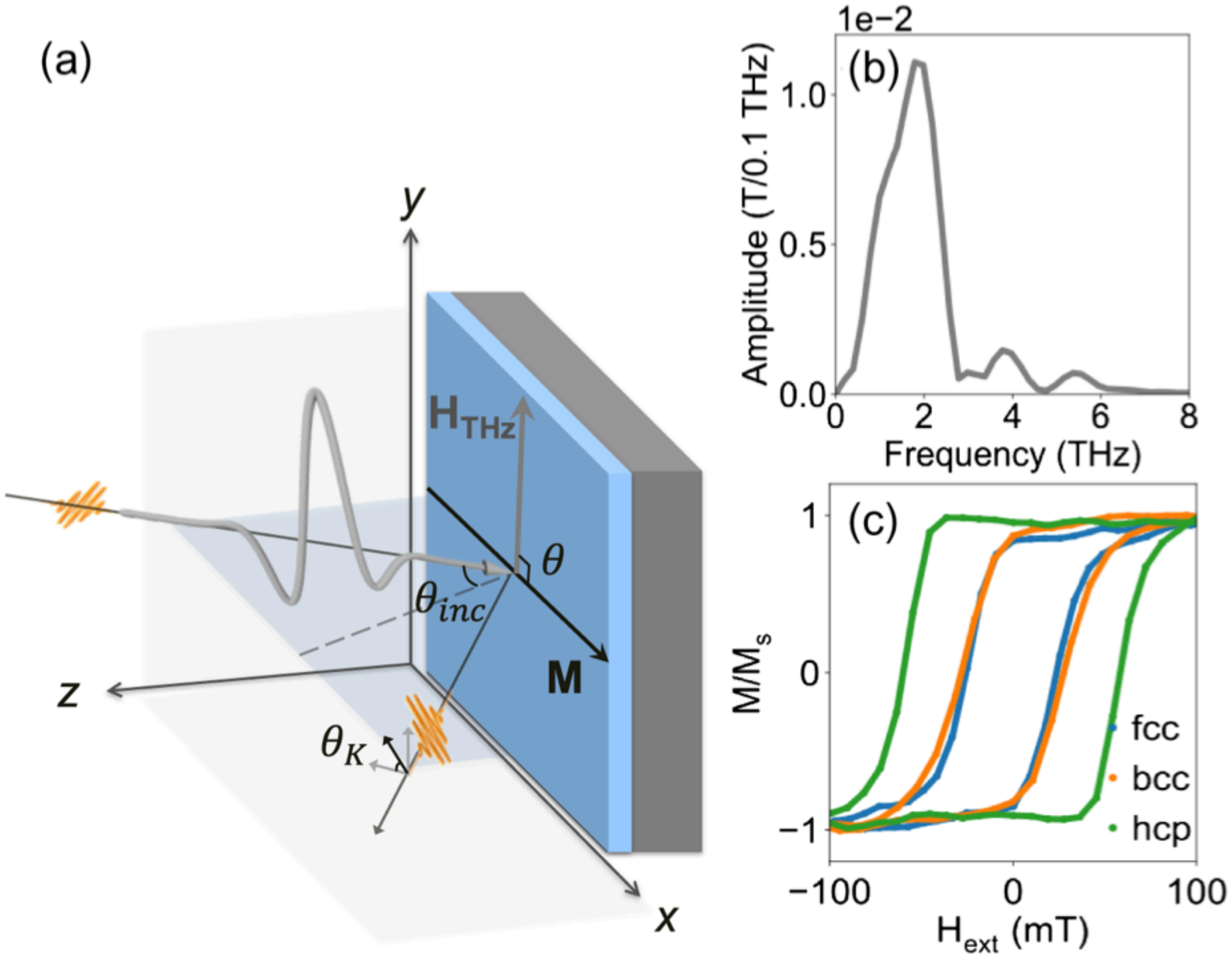


- Forced resonance at 100-1000x higher frequency than FMR
- Peak shifts (slightly) for different materials
- Peak frequency and phase shift reproduced by inertial LLG, not linewidth

K. Neeraj et al., “Inertial spin dynamics in ferromagnets”, *Nature Physics* **17**, 245 (2020)

Next step

- Microscopically, inertial effects found when including higher-order spin-orbit terms in Dirac Hamiltonian
- Idea 1: let’s measure materials where we can control the magneto-crystalline anisotropy
- Idea 2: try with single-cycle, broadband intense THz fields (~1 MV/cm)

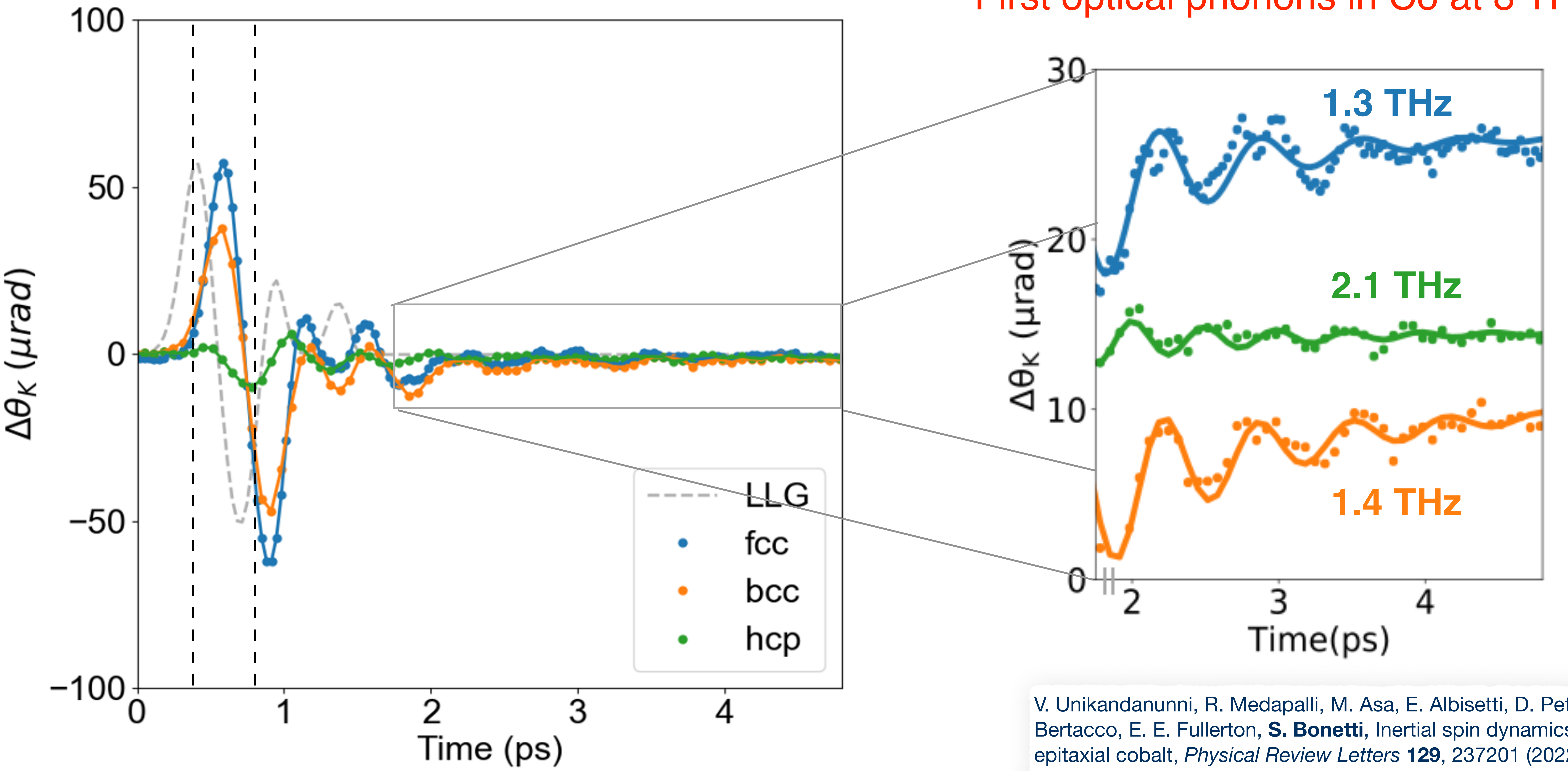


- Use broadband THz pump radiation centered at 1.5 THz, peak field at 1.3 THz
- Measure with time-resolved (~50 fs) MOKE
- Compare epitaxial fcc, bcc and hcp cobalt films

V. Unikandanunni, R. Medapalli, M. Asa, E. Albisetti, D. Petti, R. Bertacco, E. E. Fullerton, **S. Bonetti**, Inertial spin dynamics in epitaxial cobalt, *Physical Review Letters* **129**, 237201 (2022) arXiv:2109.03076

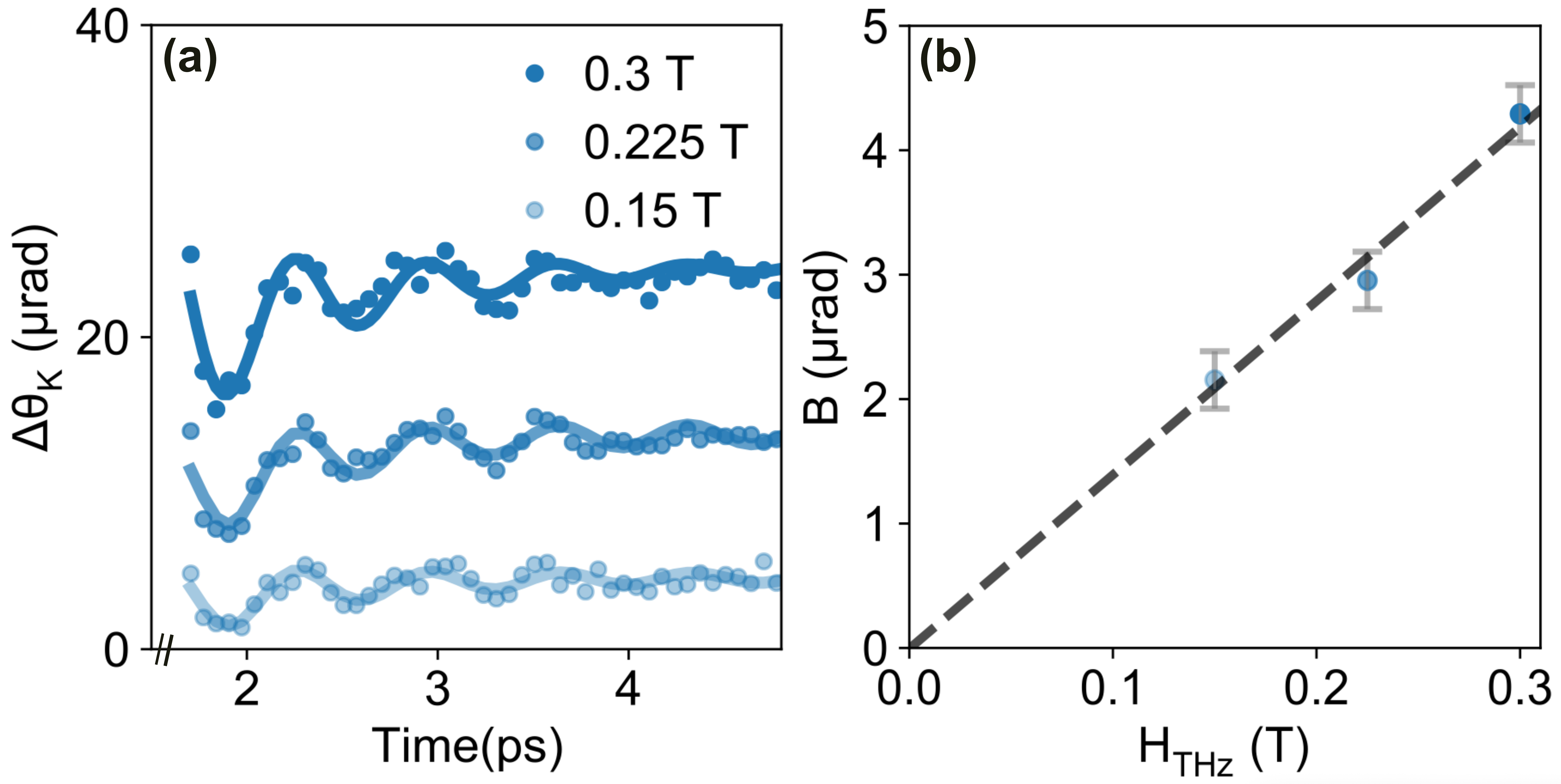
Broadband THz magnetic response

First optical phonons in Co at 8 THz!



V. Unikandanunni, R. Medapalli, M. Asa, E. Albisetti, D. Petti, R. Bertacco, E. E. Fullerton, **S. Bonetti**, Inertial spin dynamics in epitaxial cobalt, *Physical Review Letters* **129**, 237201 (2022) arXiv:2109.03076

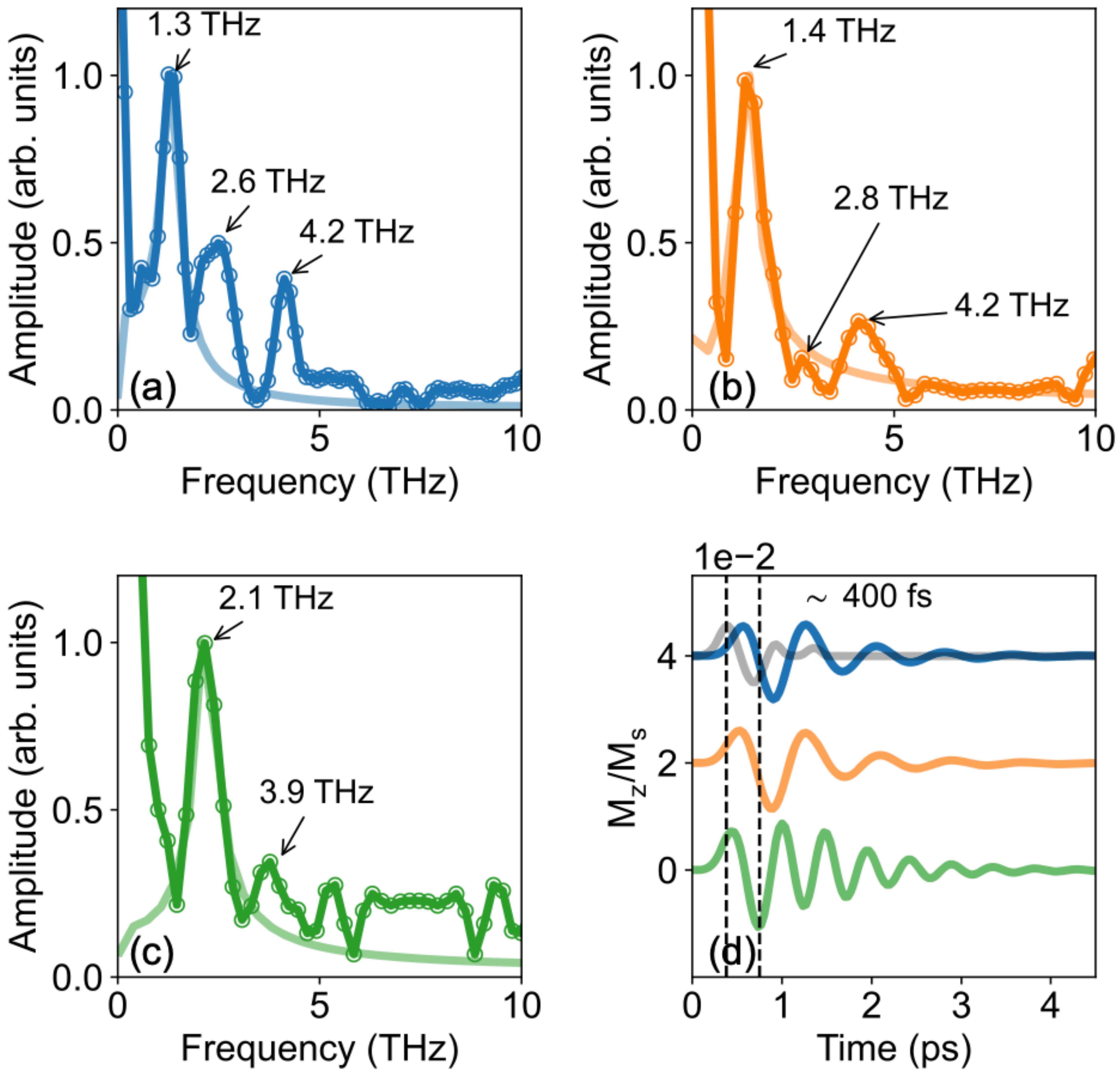
Magnetic origin, linear dependence



- Linear scaling with THz magnetic field
- Measured linewidth 10x FMR one
- Data reproduced by inertial LLG

V. Unikandanunni, R. Medapalli, M. Asa, E. Albisetti, D. Petti, R. Bertacco, E. E. Fullerton, S. Bonetti, Inertial spin dynamics in epitaxial cobalt, *Physical Review Letters* **129**, 237201 (2022) arXiv:2109.03076

Frequency-domain response



- Inertial LLG reproduces nutation frequency, linewidth and temporal shift
- Only input parameters: measured damping (10x Gilbert FMR damping) and tau
- Higher order harmonics not reproduced

V. Unikandanunni, R. Medapalli, M. Asa, E. Albisetti, D. Petti, R. Bertacco, E. E. Fullerton, **S. Bonetti**, Inertial spin dynamics in epitaxial cobalt, *Physical Review Letters* **129**, 237201 (2022) arXiv:2109.03076

$$\frac{d\mathbf{M}}{dt} = \mathbf{M} \times \left(-\gamma_0 \mathbf{H}_{\text{eff}} + \Gamma \frac{d\mathbf{M}}{dt} + I \frac{d^2\mathbf{M}}{dt^2} \right)$$

Dependent on magneto-crystalline anisotropy

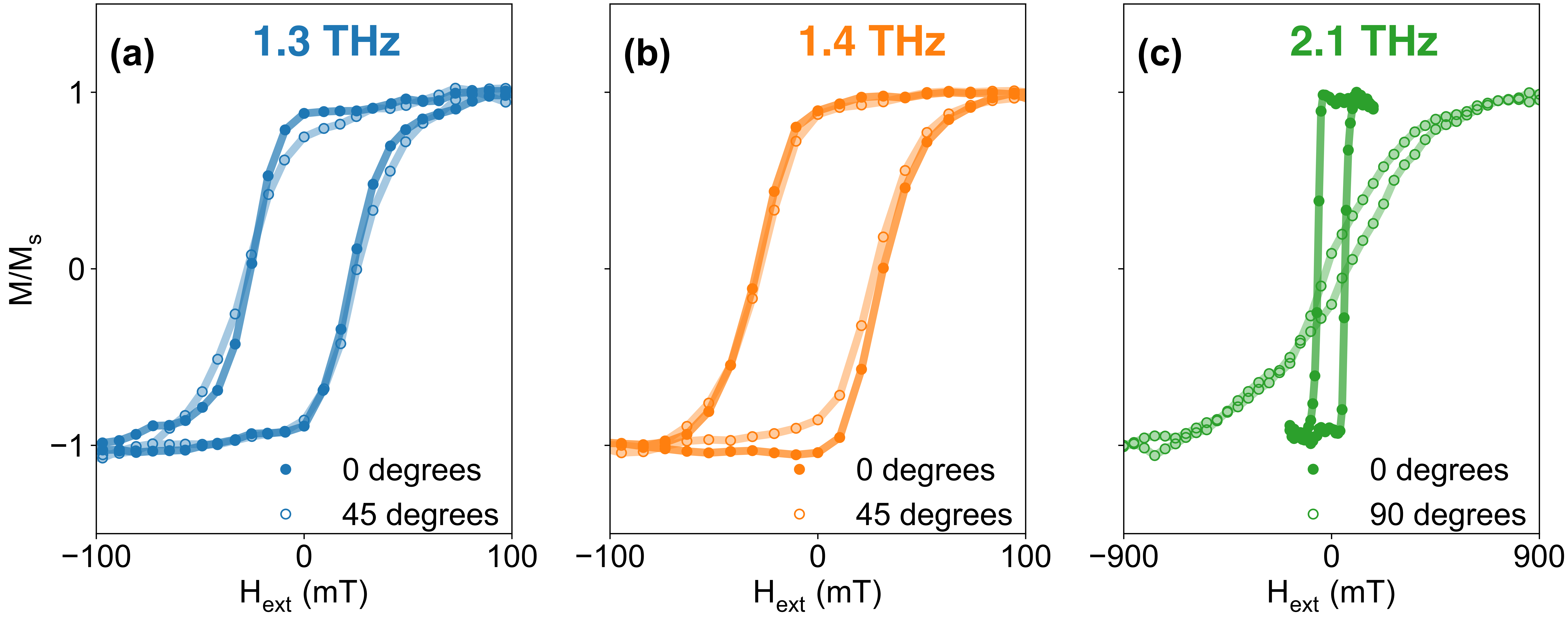
$$\Gamma_{ij} = A_{ij} + \mu_0 \delta \partial_t \left(\overline{\chi_m^{-1}} \right)_{ij}$$

$$I_{ij} = \mu_0 \delta \left(1 + \overline{\chi_m^{-1}} \right)_{ij}$$

$$\frac{I}{\Gamma} = 746 \pm 46 \text{ fs}$$

Only one free parameter needed for inertial LLG to reproduce experiments!

Magneto-crystalline anisotropy and nutation frequency



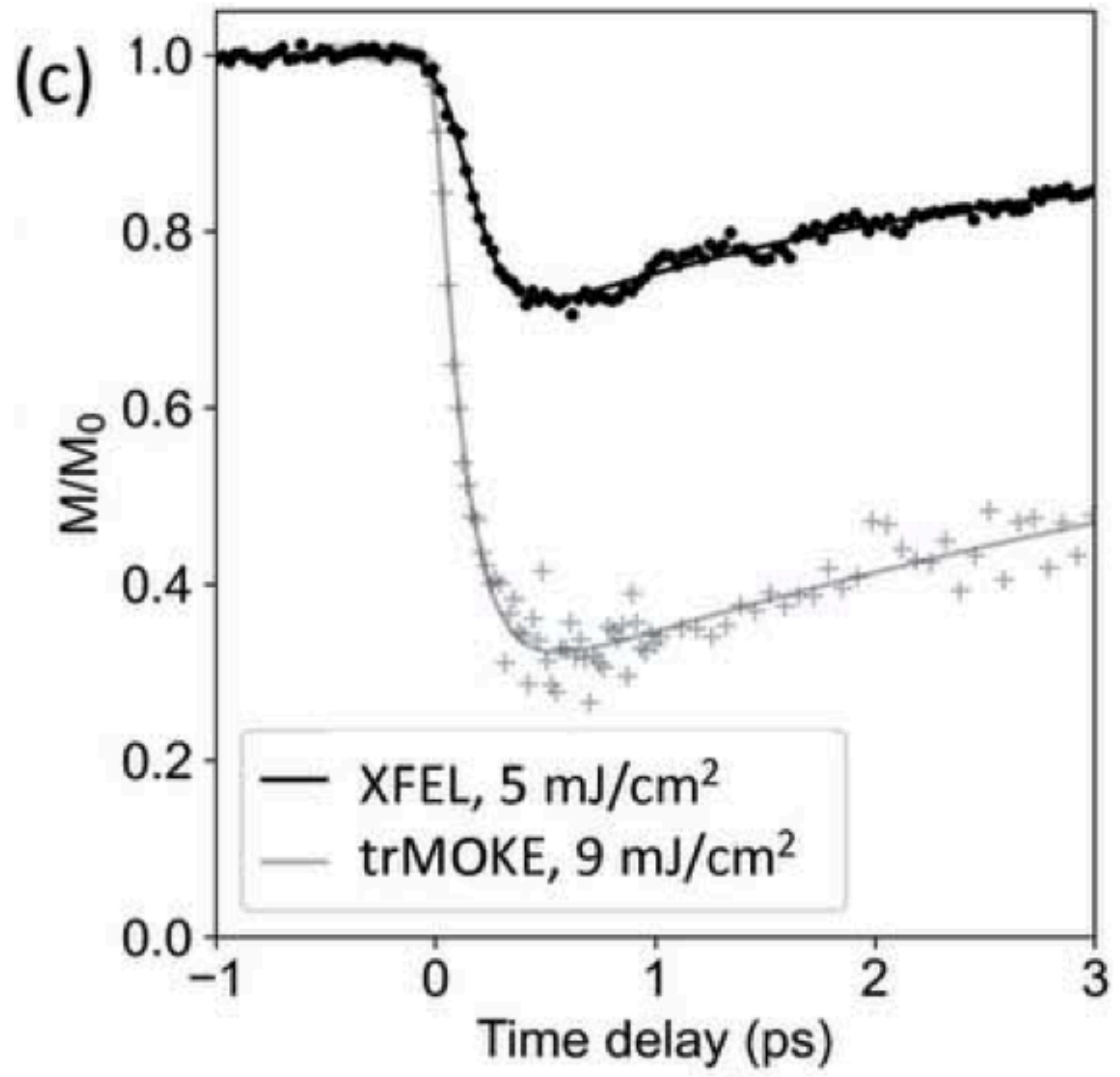
UC San Diego, USA

Politecnico di Milano, Italy

UC San Diego, USA

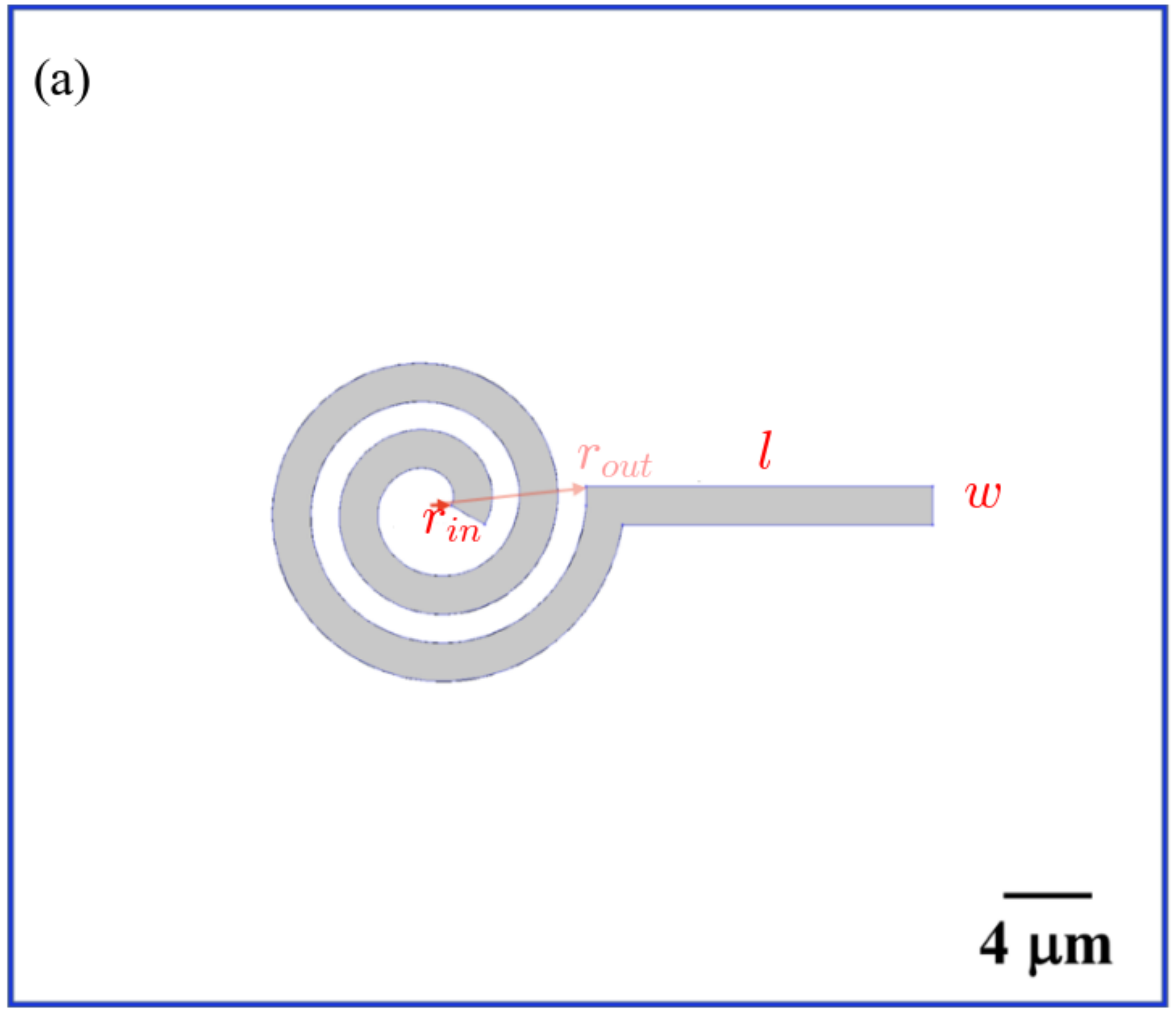
Why so few experiments?

- SNR at TELBE with intense narrowband fields and 250 kHz rep rate is extremely high, able to see in NiFe and CoFeB
- Table-top: quality of films essential. So far nutation seen only in epitaxial cobalt films (from two groups, UCSD and PoliMI)
- However, there seems to be overlooked oscillations in literature (CoFe/Ni)
- Dürr's group has similar oscillations in FePt



Part three:
magnetic metamaterials

Asymmetric spiral metamaterial



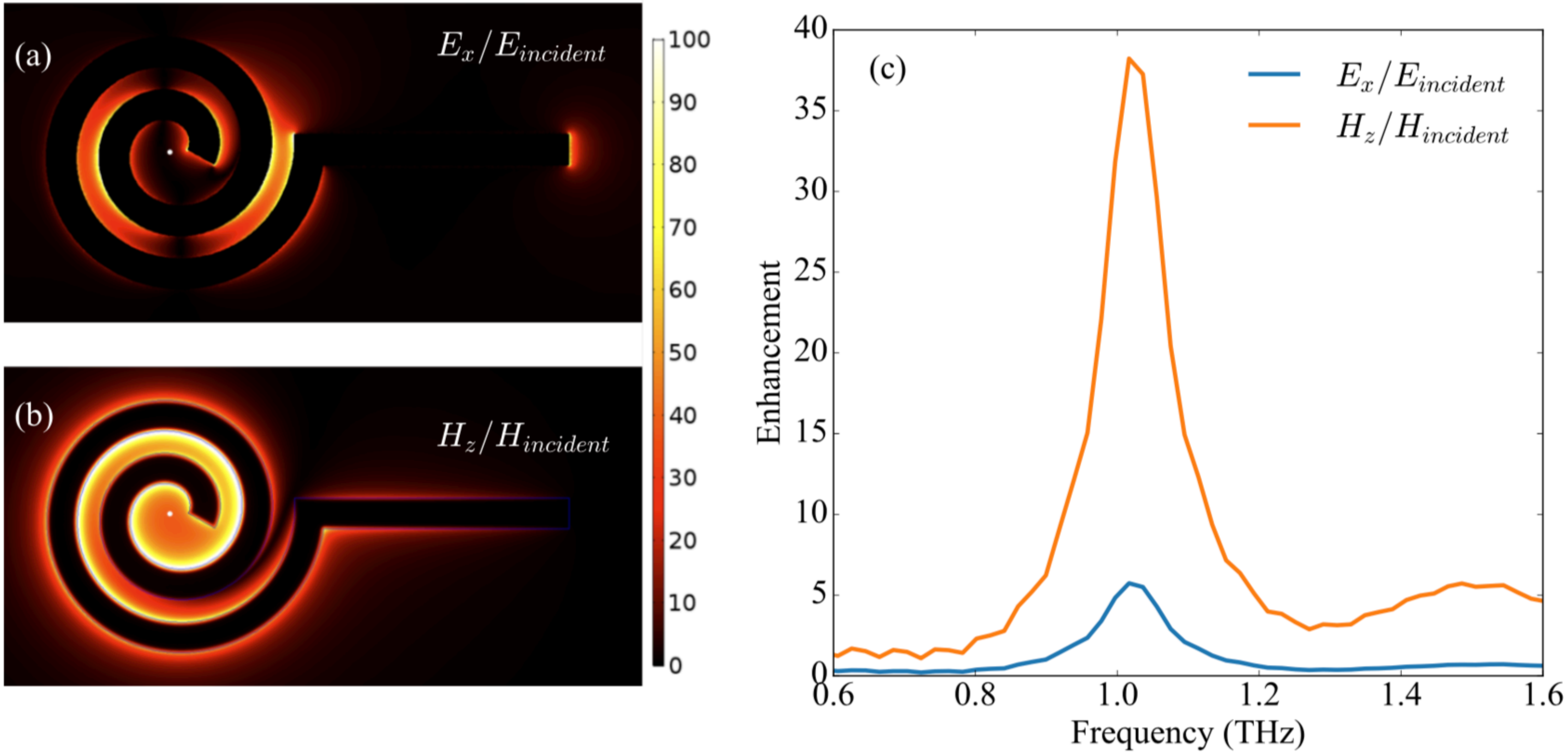
Work Plane

"Lollipop"

D. Polley, M. Pancaldi, M. Hudl, P. Vavassori, S. Urazhdin, **S. Bonetti**, *Journal of Physics D: Applied Physics* **51**, 084001 (2018)

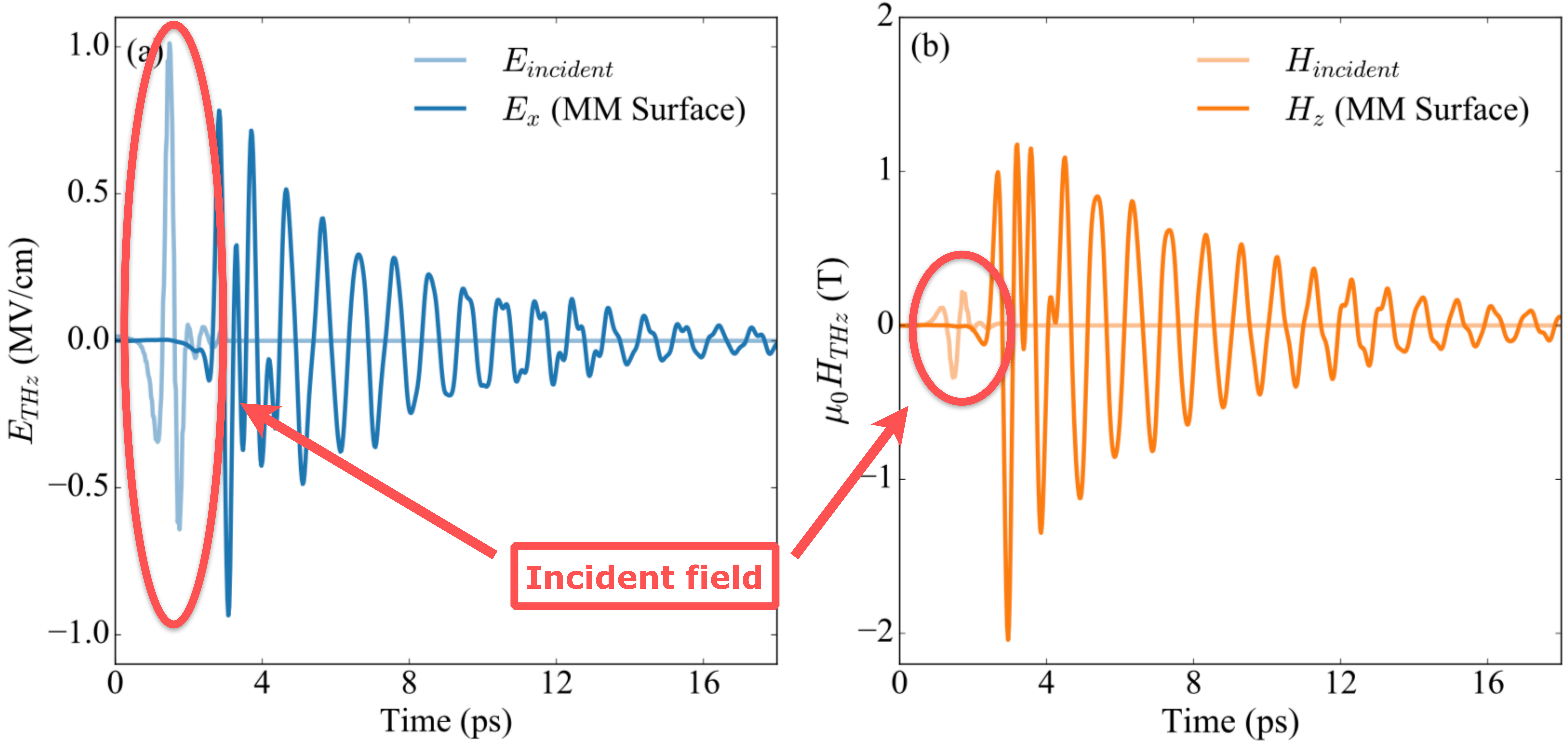
D. Polley, N. Z. Hagström, C. von Korff Schmising, S. Eisebitt, **S. Bonetti**, *Journal of Physics B: Atomic Molecular and Optical Physics* **51**, 224001 (2018),

Full 3D FEM simulations of field enhancement: *frequency domain*



D. Polley, N. Z. Hagström, C. von Korff Schmising, S. Eisebitt, **S. Bonetti**, *Journal of Physics B: Atomic Molecular and Optical Physics* **51**, 224001 (2018),

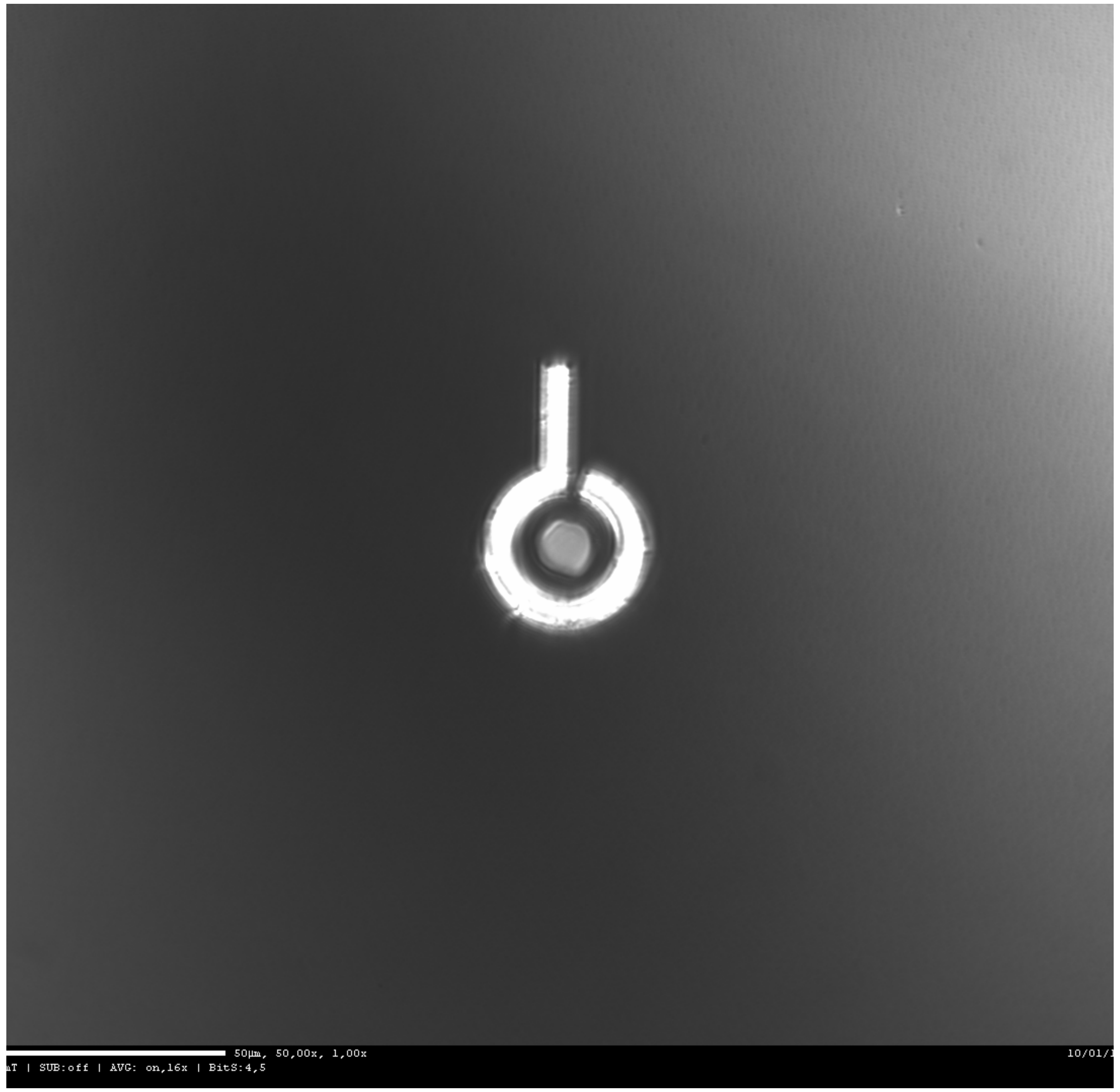
Full 3D FEM simulations of field enhancement: *time domain*



H. Qiu et al, Opt. Lett., **43**, 1658 (2018)

D. Polley, N. Z. Hagström, C. von Korff Schmising, S. Eisebitt, **S. Bonetti**, *Journal of Physics B: Atomic Molecular and Optical Physics* **51**, 224001 (2018),

Next step: let’s try this experimentally



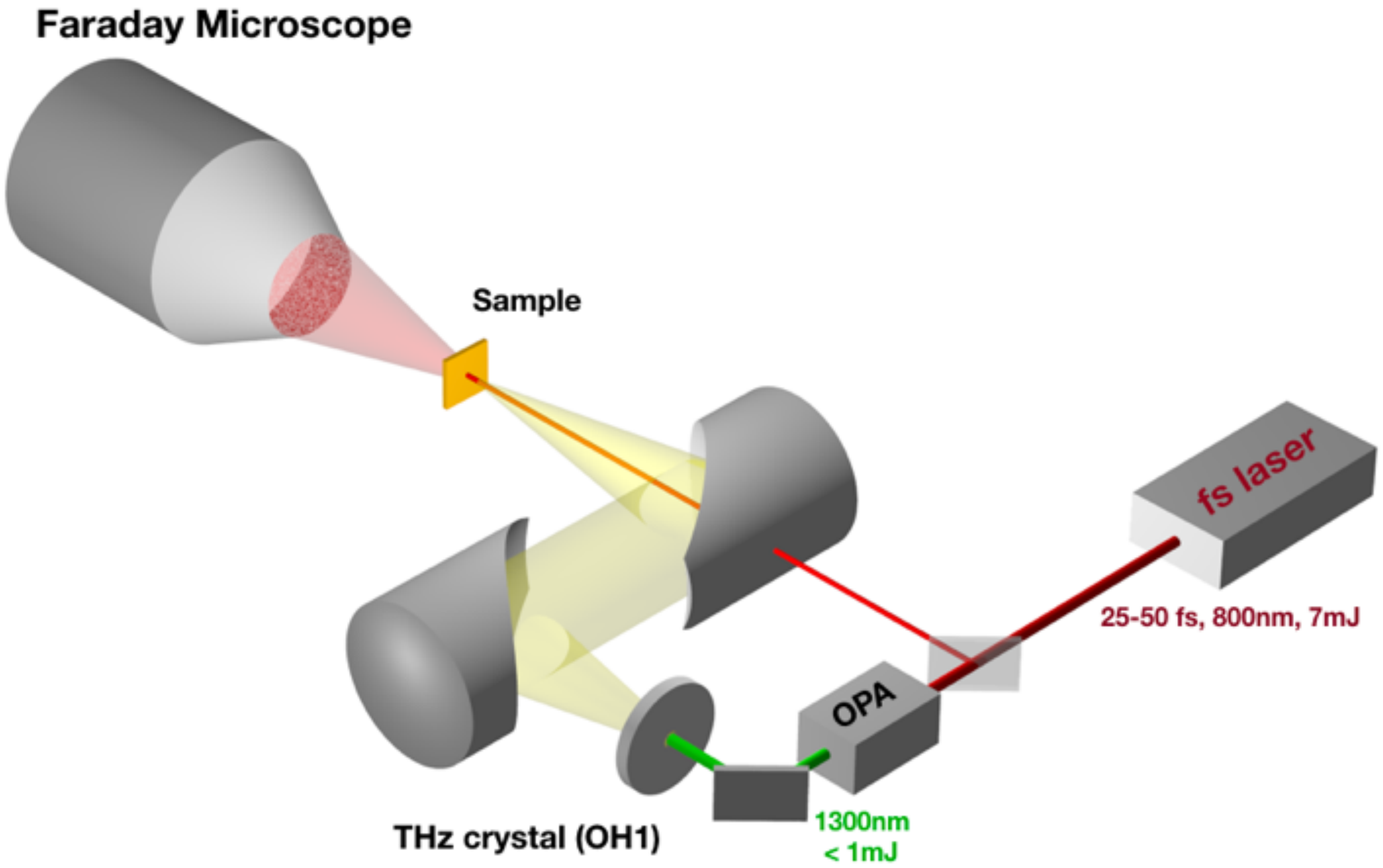


Table-top, large amplitude, ultrafast magnetization dynamics and switching in ferromagnets possible?

Take-home messages

- Strong THz fields “see” properties of materials hidden to higher energy photons
- Nutation in ferromagnets observed
- Magnetic metamaterials can realize Tesla-field THz pulses

Thank you for your attention!