Emission of coherent THz magnons in an antiferromagnetic insulator triggered by ultrafast spin-phonon interactions

SPICE Workshop - ***Terahertz spintronics: toward terahertz spin-based devices*** - **October 11th, 2023**

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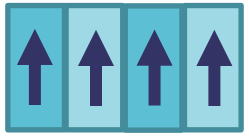
Spintronics with antiferromagnets

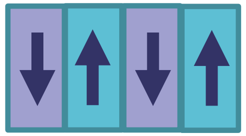
T. Jungwirth *et al.*, Nat. Nanotech. 11, 231–241 (2016)
V. Baltz *et al.*, Rev. Mod. Phys. 90, 015005 (2018)



Ferromagnet

Magnetization $\boldsymbol{M}$

Dynamics: GHz



$\boldsymbol{M} = \boldsymbol{0}$

Antiferromagnet

Néel vector $\mathbf{n} = \boldsymbol{M}_1 - \boldsymbol{M}_2$

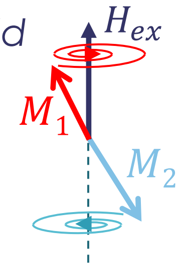
Towards THz

Exchange interaction J<0

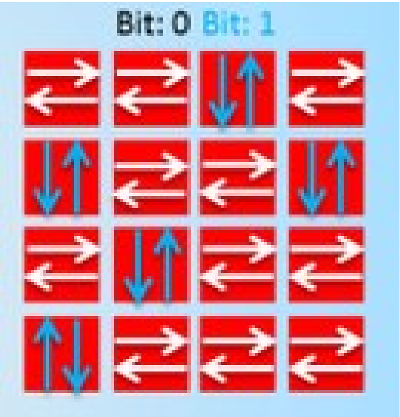
$$H_{ij} = -J\sum_{i,j} \boldsymbol{m}_i \cdot \boldsymbol{m}_j$$

High exchange field

$$B_{\mathrm{ex}} \equiv 1\,000\ \mathrm{T}$$

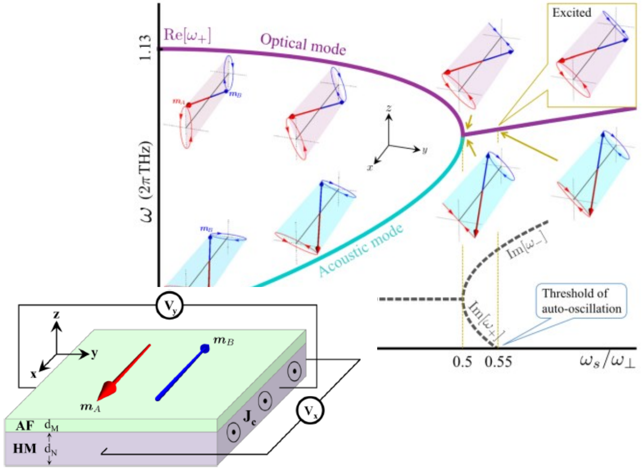


Stable magnetic memories



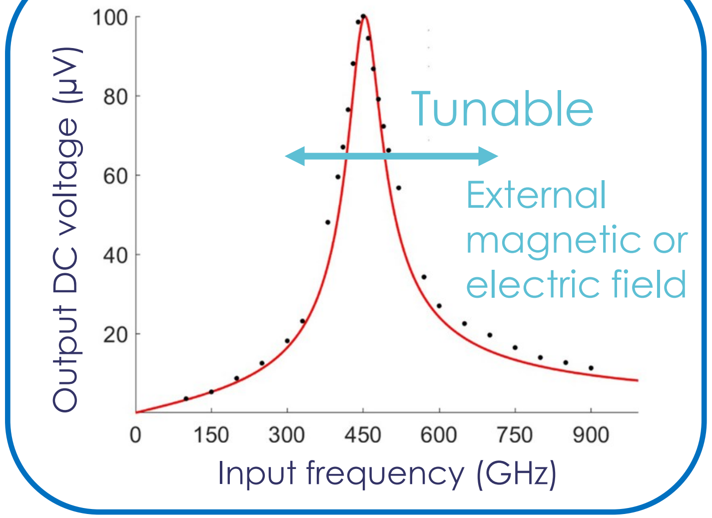
P. Wadley *et al.*, Science 351, 6273 (2016)

Spin-based oscillators



R. Cheng *et al.*, Phys. Rev. Lett., 116, 20, 207603 (2016)

THz detection



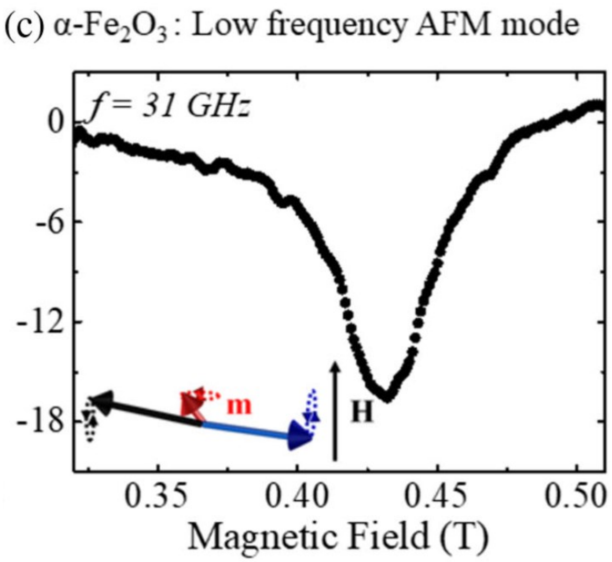
Tunable

External magnetic or electric field

A. Safin *et al.*, Appl. Phys. Lett., 117, 222411 (2020)

Antiferromagnets are platforms of choice for THz based spintronic devices

Sub-THz magnonics with canted antiferromagnets



1. AFM resonance detectable by DC spin-pumping

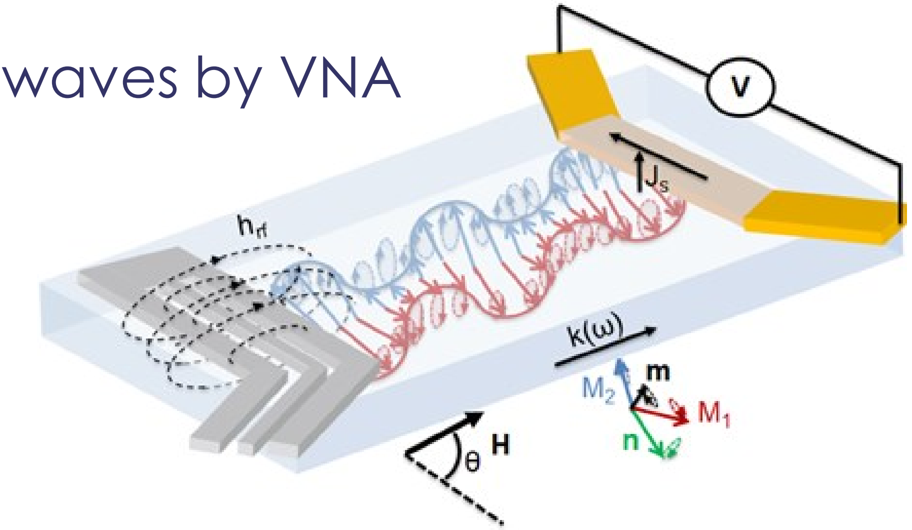
   I. Boventer, RL, *et al.*, PRL 126, 187201 (2021)

2. Larger coupling between AFM magnons and photons

   I. Boventer, RL, *et al.*, PRApplied 19, 014071 (2023)

3. Detection of ultra-fast bulk and surface spin-waves by VNA spectroscopy and DC spin-pumping

   A. El Kanj, RL, *et al.*, Science Advances 9 (32) (2023)

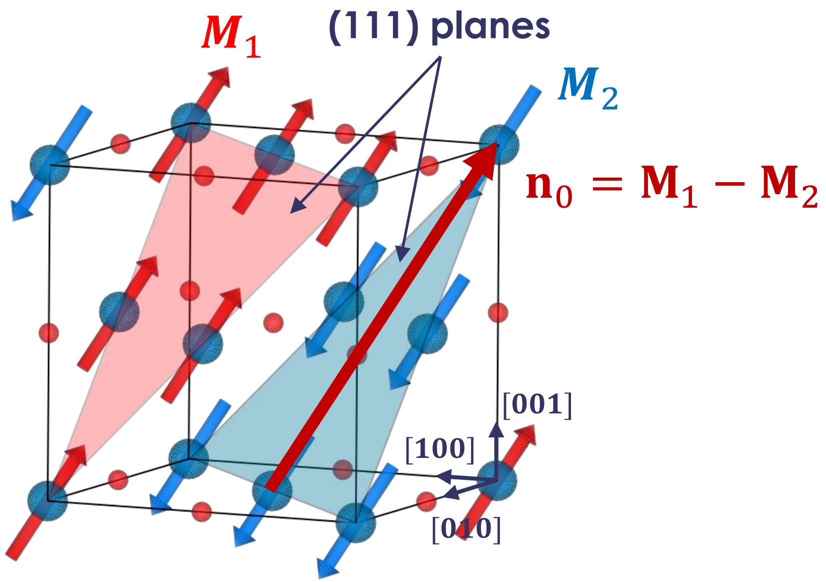


Use of antiferromagnetic materials presenting higher THz magnon modes?

Accessing AFM magnon modes: the example of NiO

Can we access the AFM magnon modes in thin films?

NiO antiferromagnet



Insulating antiferromagnet

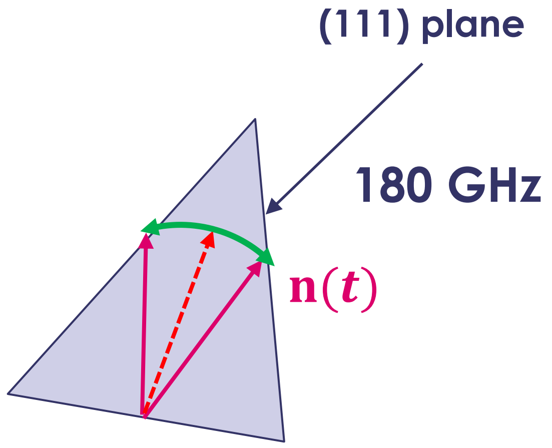
→ **Spin transport is <u>NOT</u> assured by conduction electrons**

Néel temperature T_N = 523 K

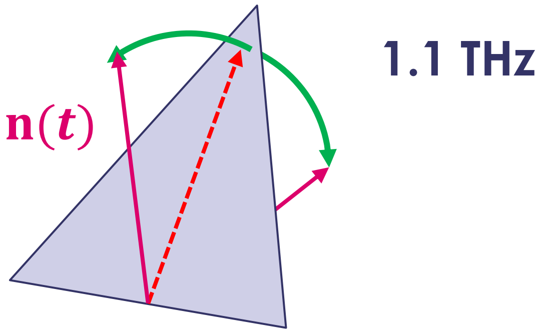
→ **Room temperature applications**

NiO modes

Low-frequency mode
« In-plane mode »



High-frequency mode
« Out-of-plane mode »

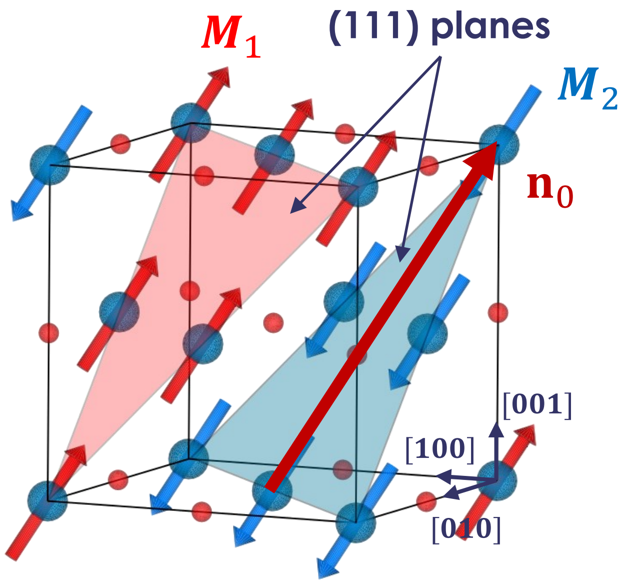


Experimentally : access to dynamical magnetization $\Delta m(t)$

Accessing AFM magnon modes: the example of NiO

Can we access the AFM magnon modes in thin films?

NiO antiferromagnet



Insulating antiferromagnet

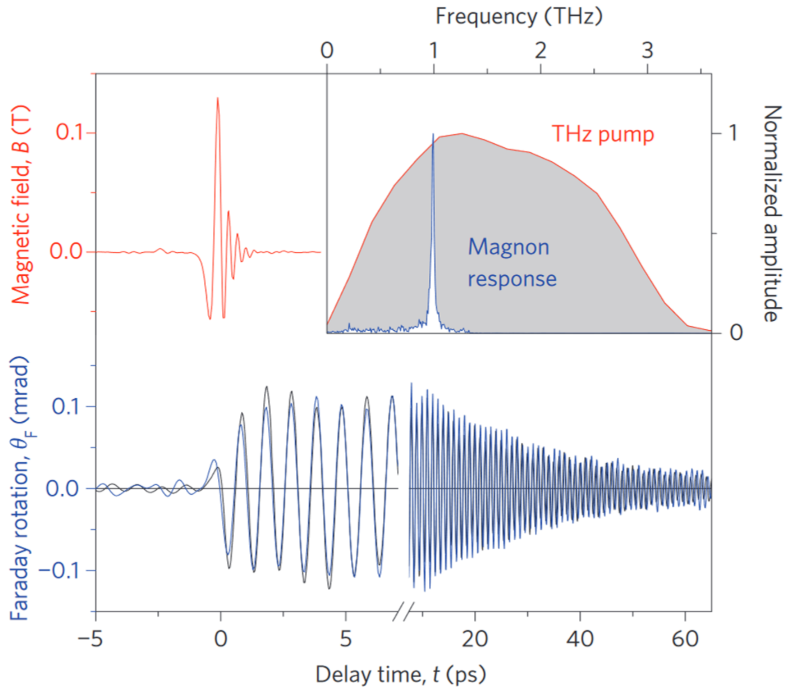
→ **Spin transport is <u>NOT</u> assured by conduction electrons**

Néel temperature T_N = 523 K

→ **Room temperature applications**

NiO bulk crystals

T. Kampfrath *et al.*, Nat. Photon. 5, 31–34 (2011)

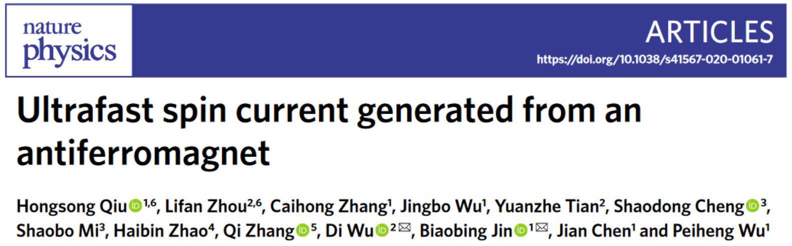


Pump-probe experiments → weak effects

Need intense THz B-field: **100 mT**

NiO thin films

H. Qiu *et al.*, Nat. Phys. 17, 388–394 (2021)

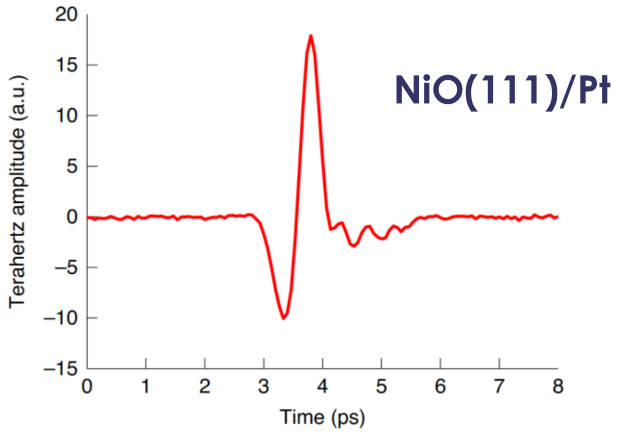

nature physics

ARTICLES

https://doi.org/10.1038/s41567-020-01061-7

Ultrafast spin current generated from an antiferromagnet

Hongsong Qiu[1,6], Lifan Zhou[2,6], Caihong Zhang[1], Jingbo Wu[1], Yuanzhe Tian[2], Shaodong Cheng[3], Shaobo Mi[3], Haibin Zhao[4], Qi Zhang[5], Di Wu[2]✉, Biaobing Jin[1]✉, Jian Chen[1] and Peiheng Wu[1]



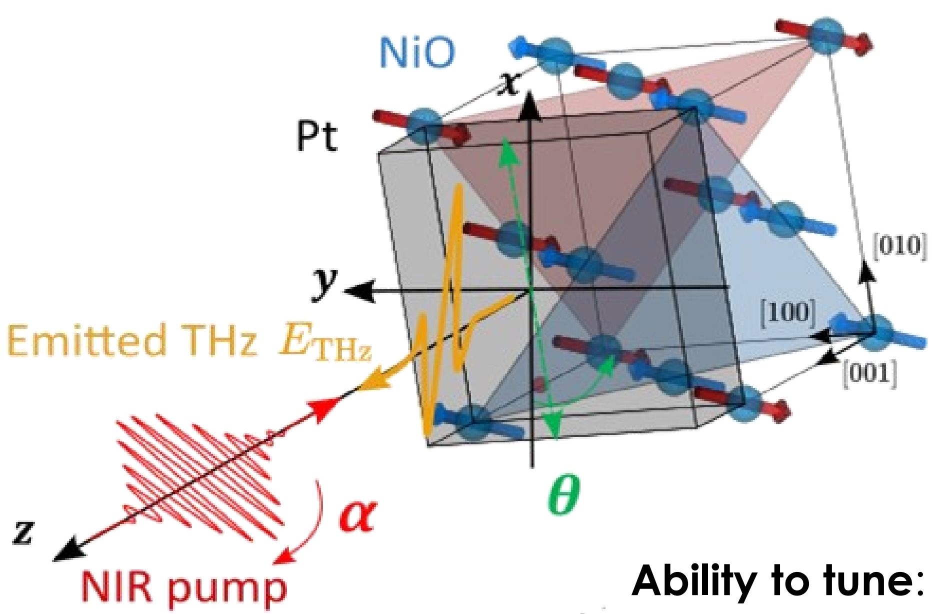
Why is there no mode at 1 THz?

Compensated AFM → accessing the magnon modes via ISHE in Pt
Is there a way to excite coherent magnons?

Accessing AFM magnon mode in easy-plane antiferromagnet NiO

Our study: excitation and detection of THz AFM magnons

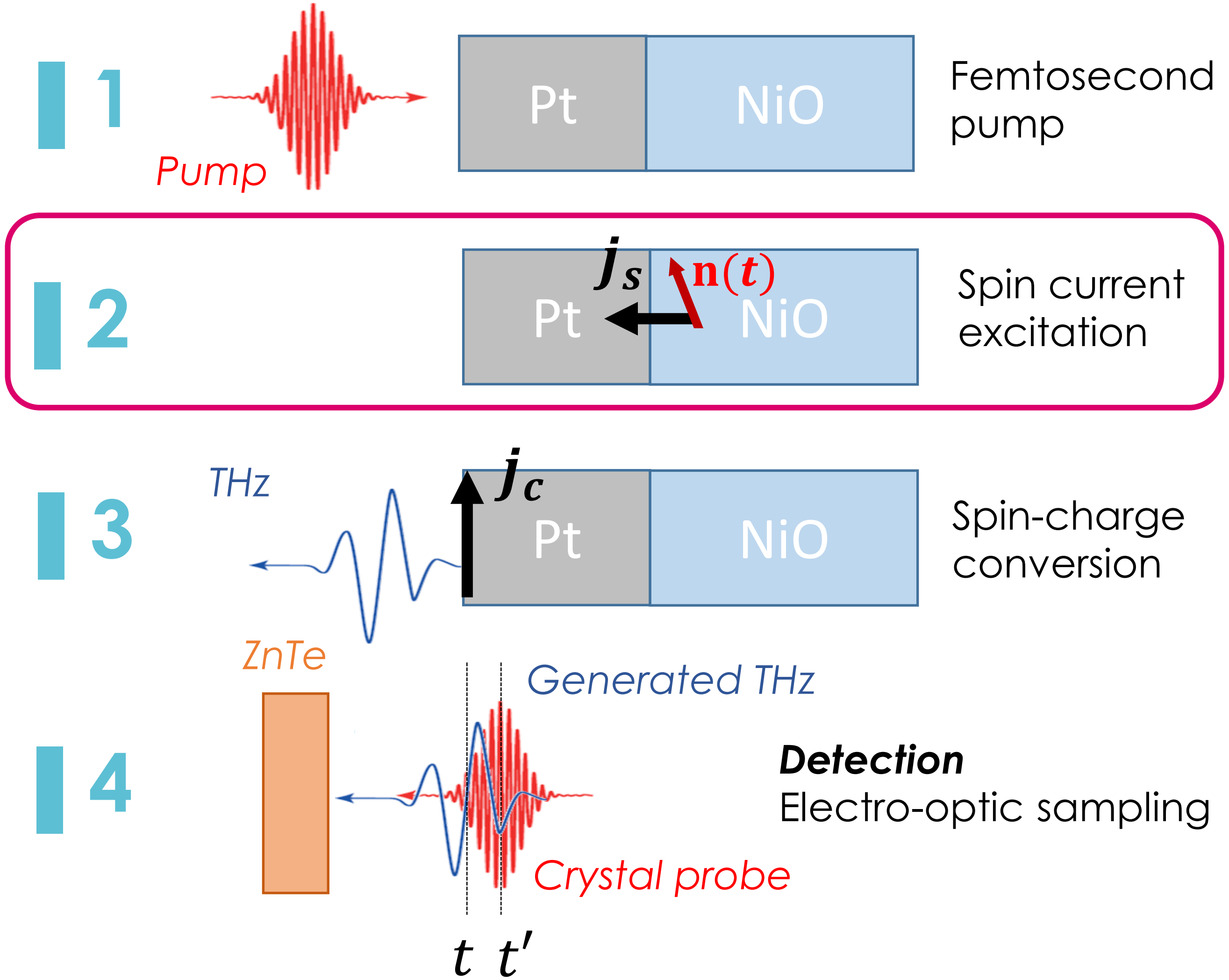
THz emission spectroscopy setup



Ability to tune:
Pump polarization α
Crystalline orientation θ

THz spin current spectroscopy by using spin-charge conversion

THz emission and detection process



1 Femtosecond pump

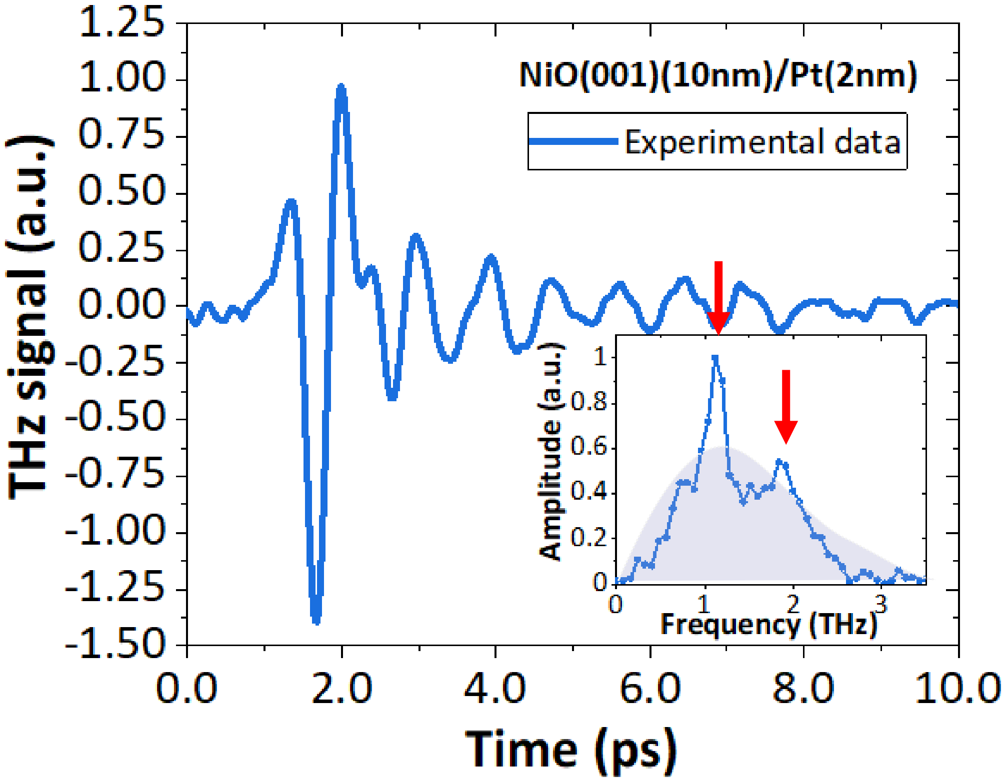
2 Spin current excitation

3 Spin-charge conversion

4 ***Detection***
Electro-optic sampling

THz narrowband emission from NiO/Pt bilayers

E. Rongione *et al.*, Nat. Commun. 14, 1818 (2023)
Collab. JGU, Tohoku Univ. for thin films growth



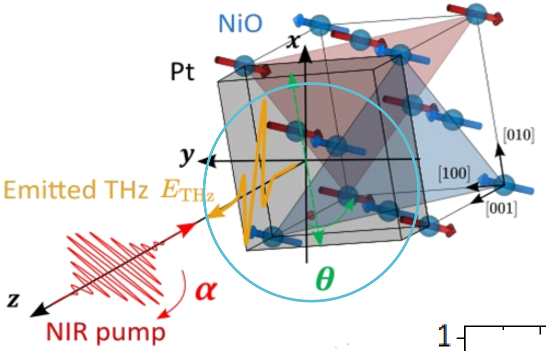
Broadband response (0.3-3 THz)
→ Uncoherent magnon bath

+

Coherent oscillations at 1.1 THz
→ High film quality
→ **Detection of the out-of-plane magnon mode**

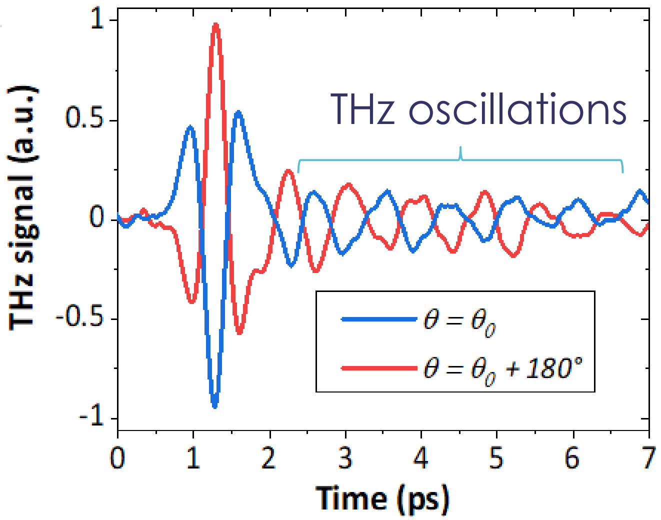
In this talk:
THz emission properties → Magnon spin current excitation mechanisms

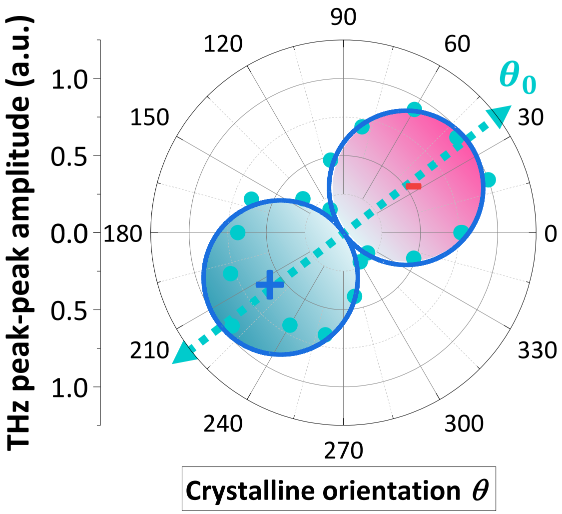
THz emission polarized along uniaxial symmetry

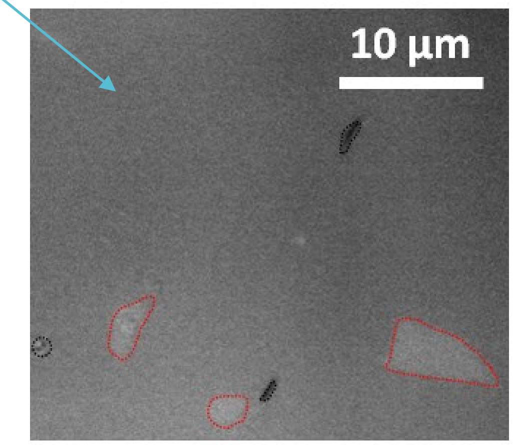


THz emission when rotating the sample (azimuthal angle θ)
Crystalline symmetries

Excited mono *T*-domain over 200 µm







Kerr imagery

F. Schreiber *et al.*, APL 117, 082401 (2020)

- → THz oscillations only in optimized thin films
  - → Thin films with low damping
- → Uniaxial emission → mono-domain orientation
  - → High film quality

THz spin current excitation linked to the Néel order

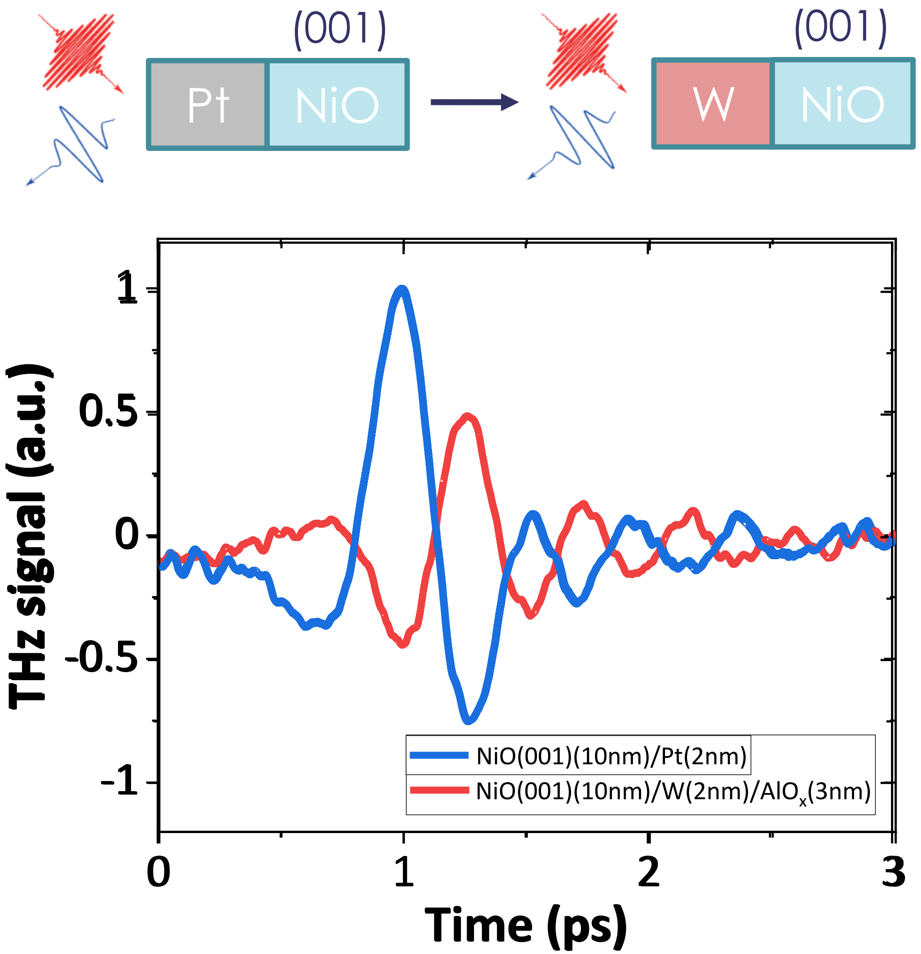
Identifying ISHE as the THz emission process

What is the THz emission mechanism?

Generated THz E-field

$$\boldsymbol{E}_{\mathrm{THz}} \propto \frac{\partial \boldsymbol{j_c}}{\partial t} \propto \theta_{\mathrm{SHE}}(\boldsymbol{j_s} \times \boldsymbol{\sigma})$$

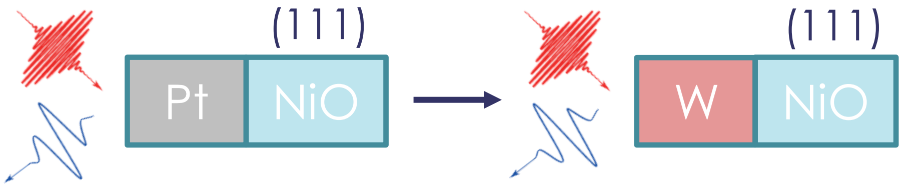
Spin Hall angle $\theta_{\mathrm{SHE}} \rightarrow -\theta_{\mathrm{SHE}}$
→ Reversed phase = ISHE

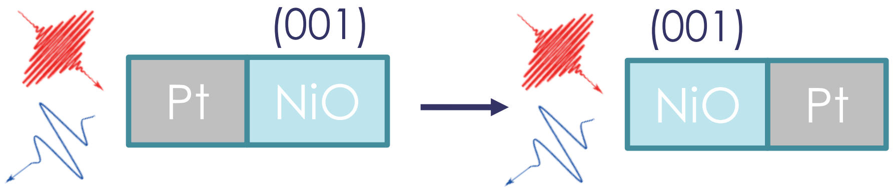


Emission process happens via inverse spin Hall effect in the heavy metal

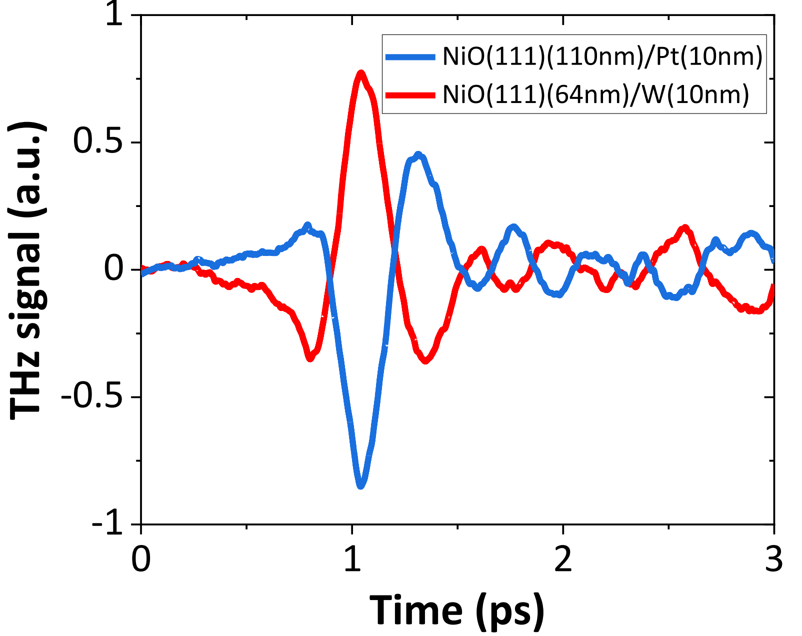
Details about the electric dipolar emission in NiO/Pt bilayers

What is the THz emission mechanism?

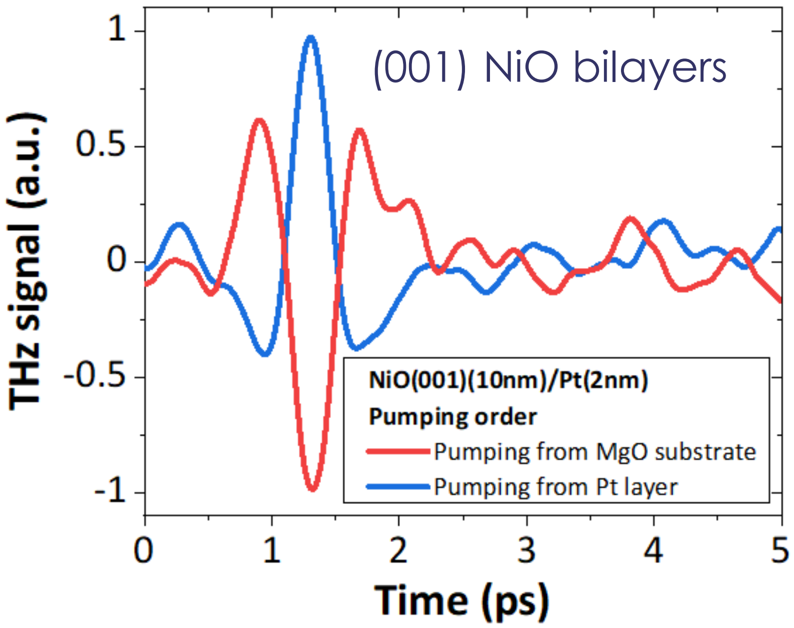




ISHE in (111) NiO bilayers



Electric dipolar radiation



Emission process follows electric dipolar radiation → inverse spin Hall in the heavy metal

What have we demonstrated up to now?

Reporting THz emission from NiO/Pt bilayers

Broadband + narrowband 1.1 THz contributions

↓

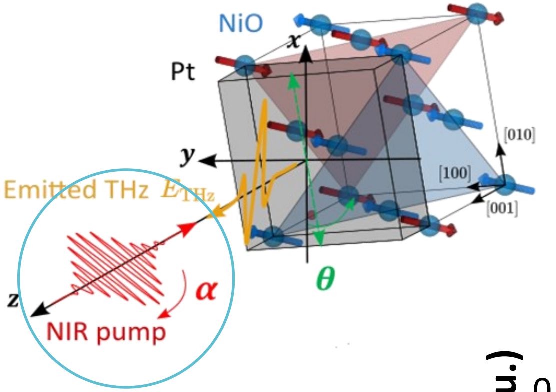
THz emission is: linked to the AFM magnon modes ✓

mediated via ISHE spin-charge conversion ✓

↓

What are the spin current excitation mechanisms?

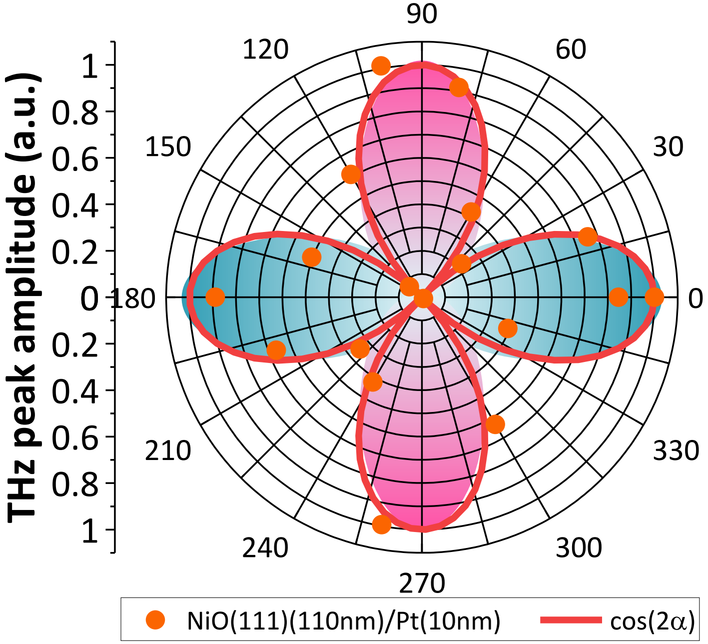
Identification of THz excitation process in (111) NiO thin films



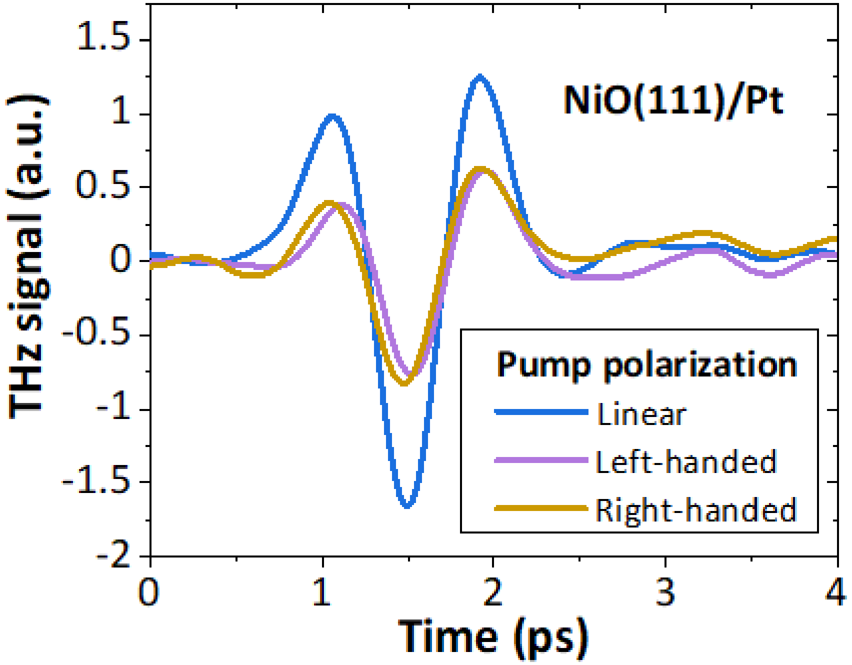
NiO(111)/Pt

Effect of the pump polarization on the THz emission

Linear pump polarization



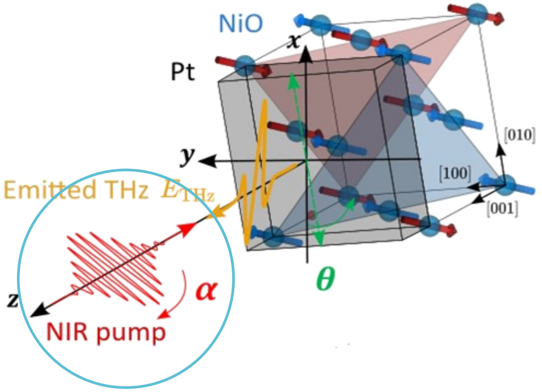
Circular pumping



Polarization-dependent excitation → *Inverse Cotton-Mouton effect*

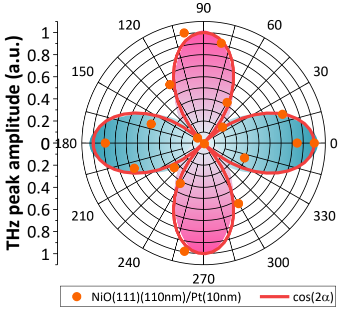
H. Qiu *et al.*, Nat. Phys. 17, 388–394 (2021)
C. Tzschaschel *et al.*, Phys. Rev. B 95, 174407 (2017)

Identification of THz excitation process in (001) NiO thin films

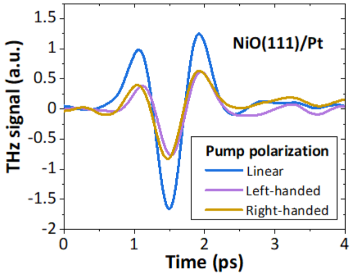


Effect of the pump polarization on the THz emission

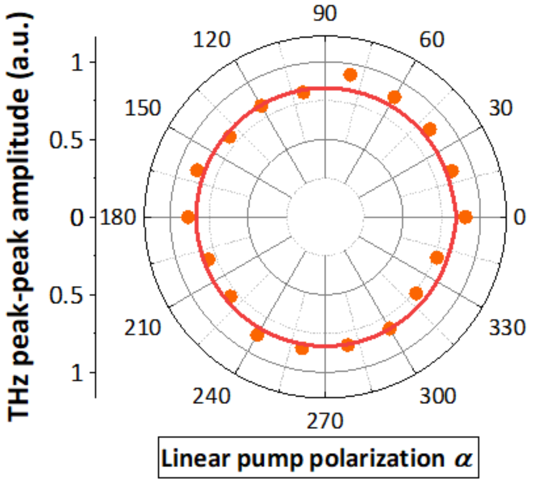
Linear pump polarization

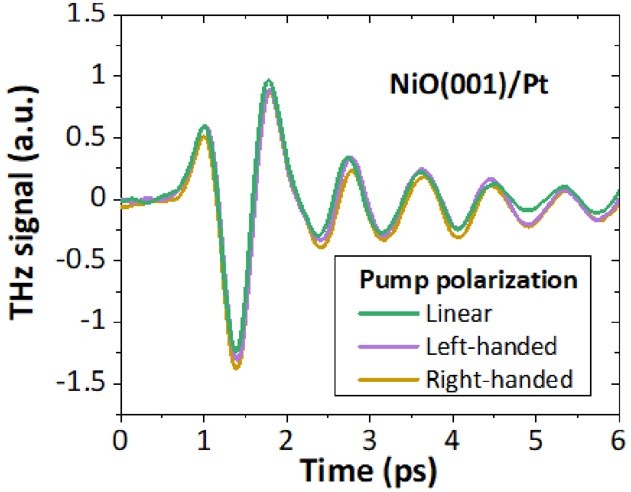


Circular pumping



NiO(111)/Pt





NiO(001)/Pt

Polarization-independent signals → thermally-mediated effects?

Different physics on (001) and (111) samples
→ Do we map different spin current excitation mechanisms?

Spin current dynamics in THz spintronic emitters

THz emission spectroscopy → possibility to extract the spin current dynamics

Collab. O. Gueckstock, T. Seifert,
T. Kampfrath (Freie Universität Berlin)

Metallic spintronic THz emitter W/CoFeB/Pt

Electron-spin relaxation time <200 fs

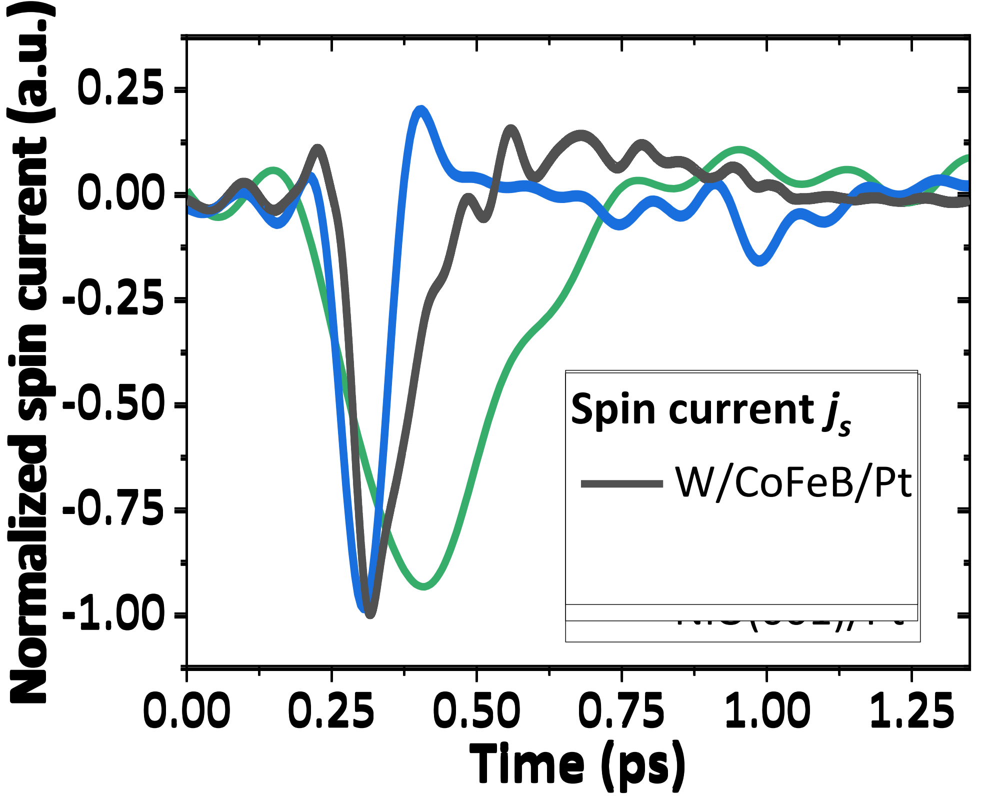
T. Seifert *et al.*, Nature Photon. 10, 483–488 (2016)

Insulating AFM THz emitters:

- **Magneto-optical excitation for (111) films**
  Timescale <80 fs

- **Thermal excitation mechanism for (001) films**
  Timescale >200 fs (build-up) - 300 fs (relaxation)

$$E_{\mathrm{THz}}(\omega) \longrightarrow j_c(\omega) \longrightarrow j_s(\omega)$$



Drasticallly different timescales for optical and thermal excitations
Origin of the thermally-mediated excitation in (001) thin films?

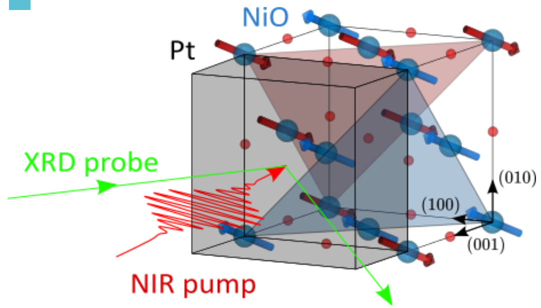
Phononic excitation in NiO(001) thin films

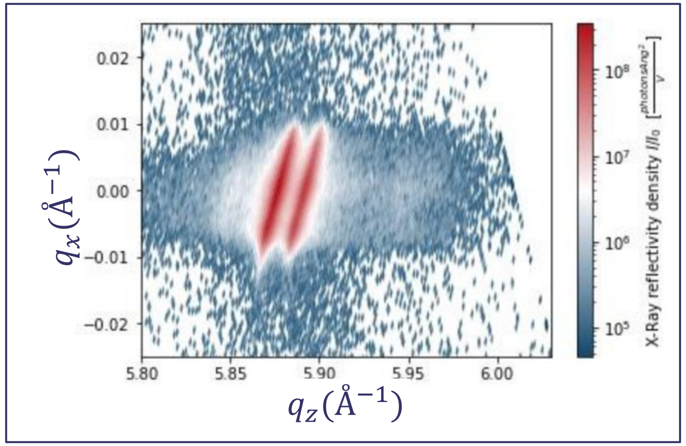
Transport in insulators → lattice dynamics

H. Meer *et al.*, Nano Lett. 21, 1, 114–119 (2021)

Collab. M. Mattern, M. Bargheer (University of Potsdam)

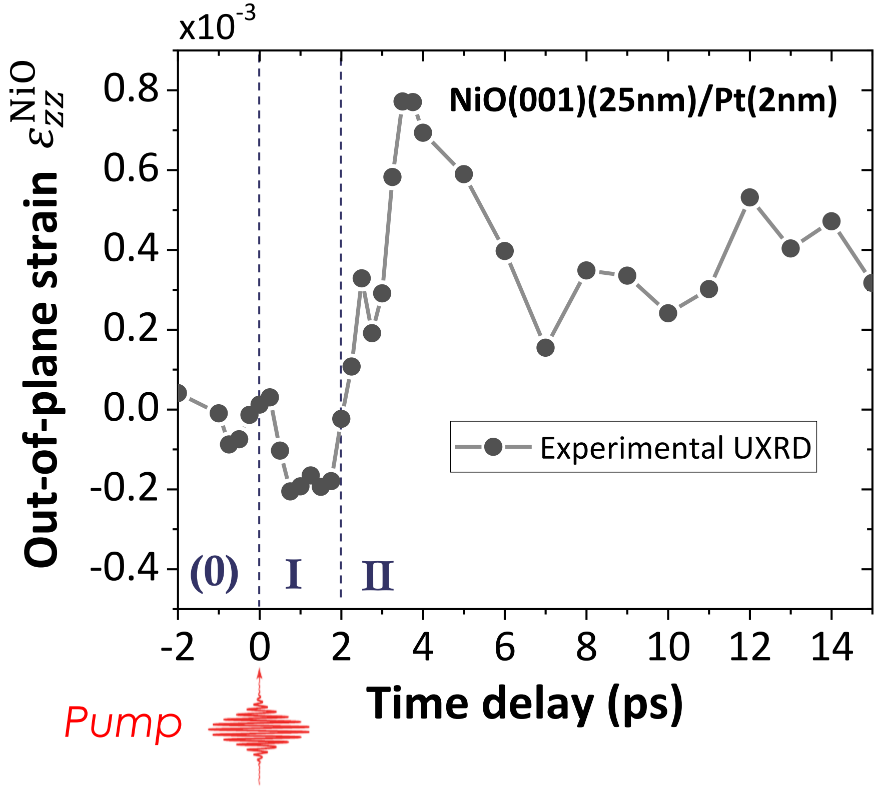
Ultrafast strain wave → Magneto-striction → Néel vector dynamics

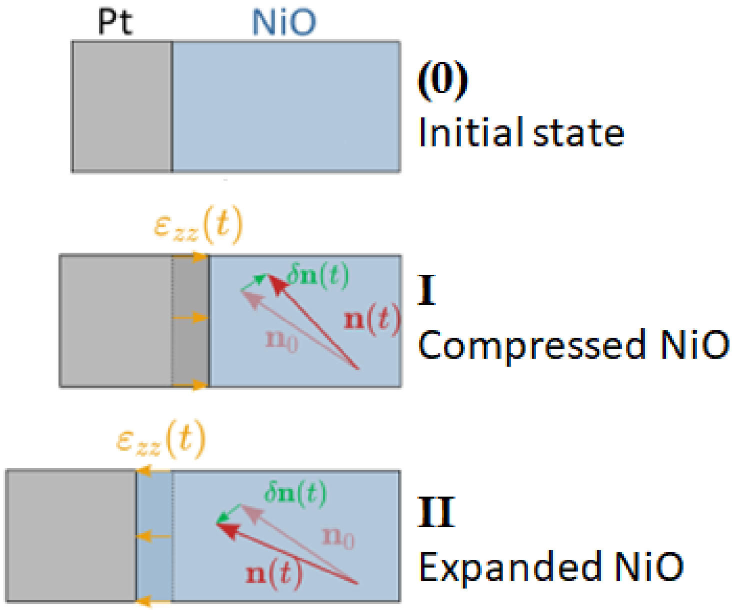




NiO(004) peak tracking

Time-resolved XRD





Out-of-plane strain $\varepsilon_{zz}(t)$ is sufficient to induce reduction of the Néel vector

Understanding the phonon-spin interactions in the THz regime

O. Gomonay and D. Bossini, J. Phys. D: Appl. Phys. 54 374004 (2021)
E. Rongione *et al.*, Nat. Commun. 14, 1818 (2023)
Collab. O. Gomonay (JGU)

Two potential sources of excitation Γ

Acoustic strain $\propto \varepsilon_{zz}(d,t)$
- Coherent phonon propagation

→ Allow AFM order reduction via **magneto-striction**

M. Deb *et al.*, Phys. Rev. B 103, 024411 (2021)
H. Meer *et al.*, Nano Lett. 21, 1, 114–119 (2021)

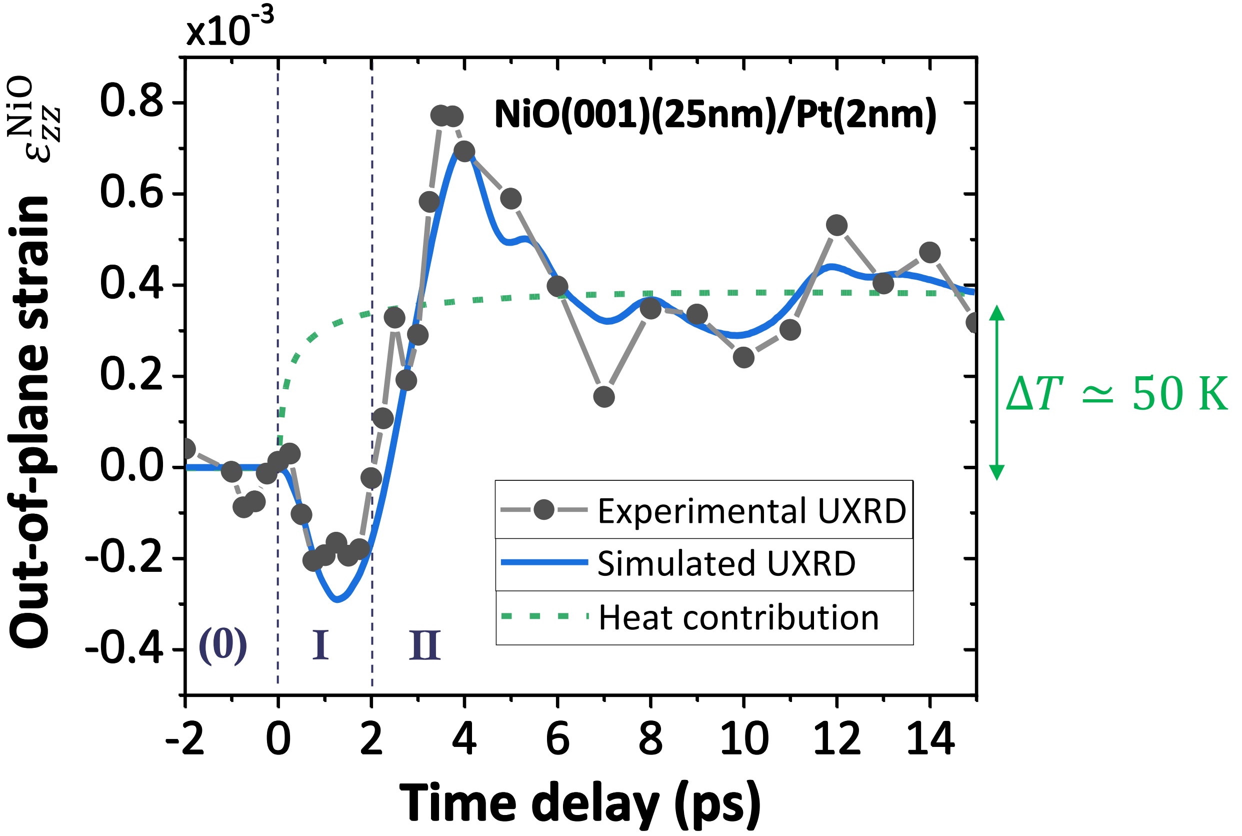
Heat contribution $\propto \Delta T(d,t)$
- Incoherent phonon propagation

→ Temperature imbalance → **spin-Seebeck effects**

T. Seifert *et al.*, Nat. Commun. 9, 2899 (2018)
F. N. Kholid *et al.*, Nat. Commun. 14, 538 (2023)

- Anisotropy changes

S. Zeuschner *et al.*, Phys. Rev. B 106, 134401 (2022)



Two contributions to the Néel vector dynamics: coherent strain wave and incoherent heat contribution

Understanding the phonon-spin interactions in the THz regime

O. Gomonay and D. Bossini, J. Phys. D: Appl. Phys. 54 374004 (2021)
E. Rongione *et al.*, Nat. Commun. 14, 1818 (2023)

Collab. O. Gomonay (JGU)

Damping **System anisotropies** **Excitation**

$$\boldsymbol{n} \times (\ddot{\boldsymbol{n}} + 2\alpha_{\mathrm{AF}}\dot{\boldsymbol{n}} - c^2\Delta\boldsymbol{n} + \gamma^2 H_{\mathrm{ex}}H_{\mathrm{an}}) \propto (\boldsymbol{n} \times \boldsymbol{\Gamma})$$

THz dynamics modelling

Acoustic strain $\propto \varepsilon_{zz}(d,t)$

- Coherent phonon propagation

→ Allow AFM order reduction via **magneto-striction**

M. Deb *et al.*, Phys. Rev. B 103, 024411 (2021)
H. Meer *et al.*, Nano Lett. 21, 1, 114–119 (2021)

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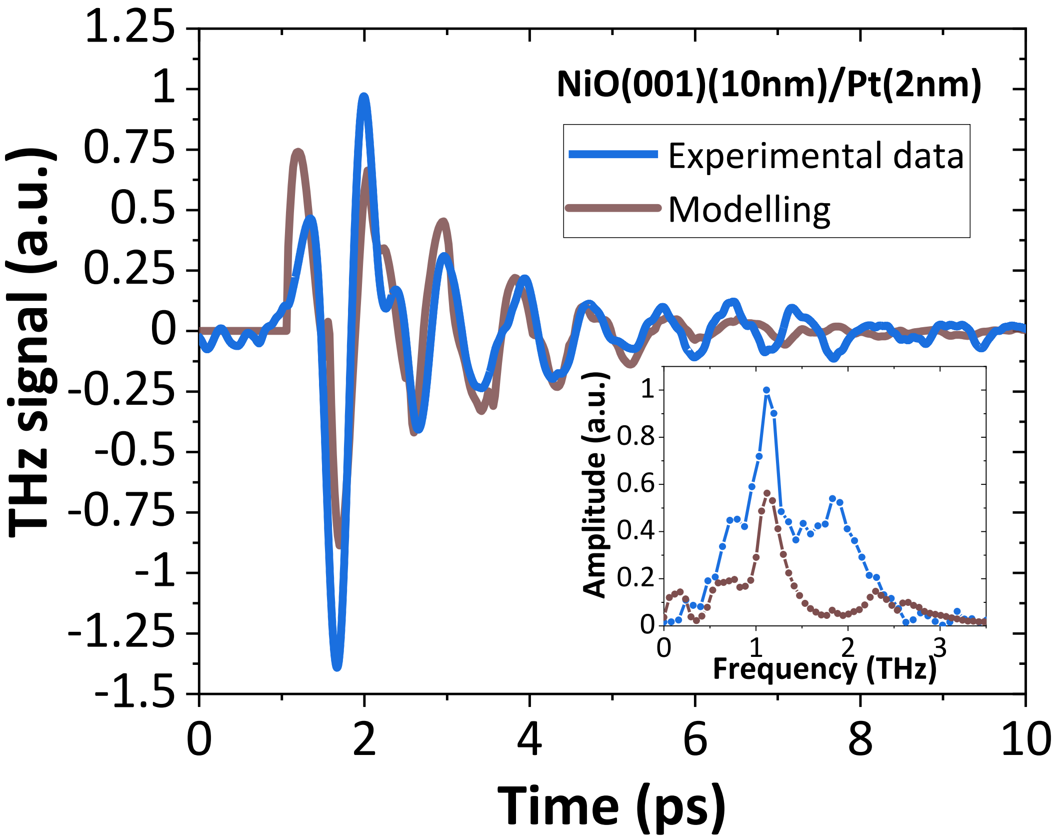
Qualitative agreement



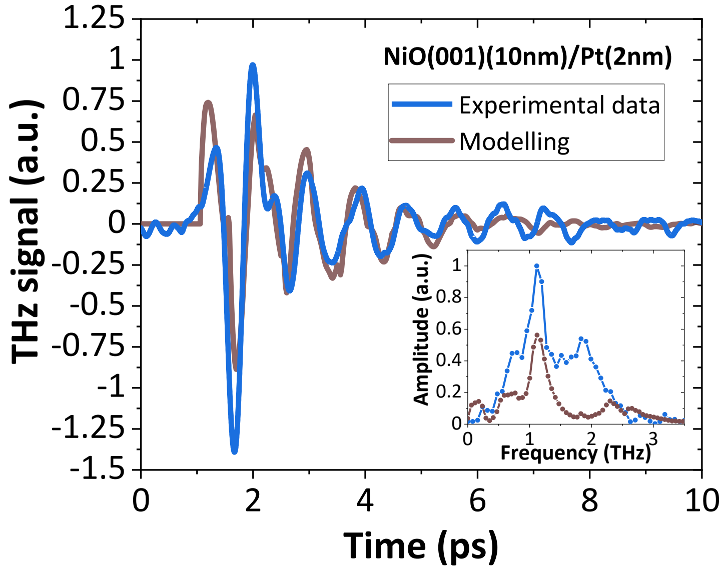
Dynamical reduction of the Néel vector → magnon pumping → THz emission

Conclusions and perspectives

First observation of narrowband 1.1 THz emission in AFM thin films

THz emission from spin-charge conversion
THz excitation of AFM modes by two main mechanisms:

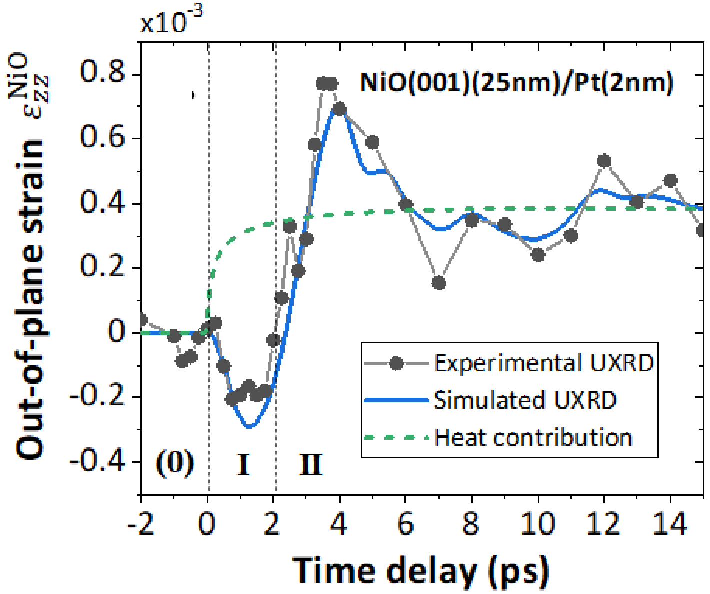
- **Off-resonant optical-spin torque** in (111) orientation
- **Ultrafast spin-phonon interactions** in (001) orientation



Take-home message

Dynamical reduction of the Néel vector by spin-phonon interactions → THz magnon pumping → coherent THz free-space emission

Perspectives
Opto-magnonics + joined optical and spin-orbit excitations



Link to the article:
E. Rongione *et al.*, Nat. Commun., 14, 1818 (2023)

Acknowledgments

THz emission spectroscopy

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S. Massabeau, N. Reyren, F. Godel, S. Collin
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Thank you for your attention!