

Coherent terahertz control of antiferromagnetic spin waves

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Outline

Background

- ❑ What applications can arise from the control of antiferromagnetic spin waves
- ❑ Why have antiferromagnetic spin wave dynamics remain elusive up until this publication
- ❑ Why is the electromagnetic terahertz regime utilized in these experiments
 - ❑ Short aside about terahertz radiation
- ❑ Archetypal antiferromagnetic material: NiO

This work

- ❑ Dynamical detection of spin waves through Faraday rotation
- ❑ Multi-THz pulse control of spin wave dynamics

What followed

- ❑ What mechanisms are at play
 - ❑ Magnetic Difference Frequency Generation (DFG), Inverse Faraday Effect (IFE), Inverse Cotton Mouton Effect (ICME)

Emerging Field of Spintronics Focused on Antiferromagnetic Materials

- ❑ Show promise in the application of robust memory devices due to their resistance to stray fields from the lack of net magnetization of the material
- ❑ Fast THZ response compared to the 3 order of magnitude slower GHz response inherent to ferromagnetic materials

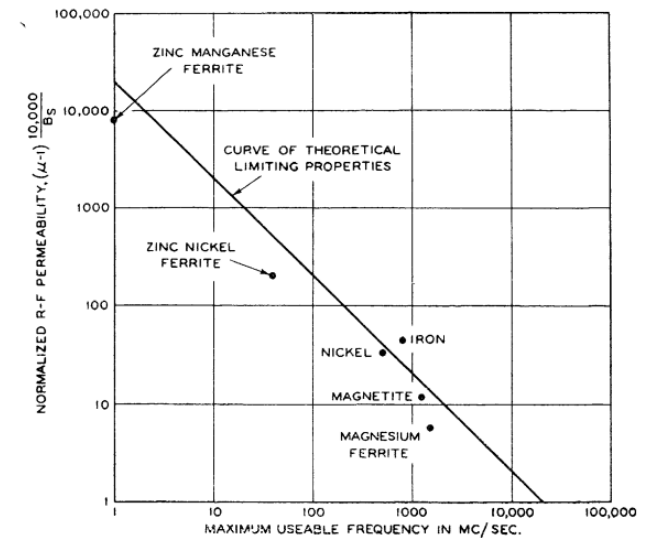
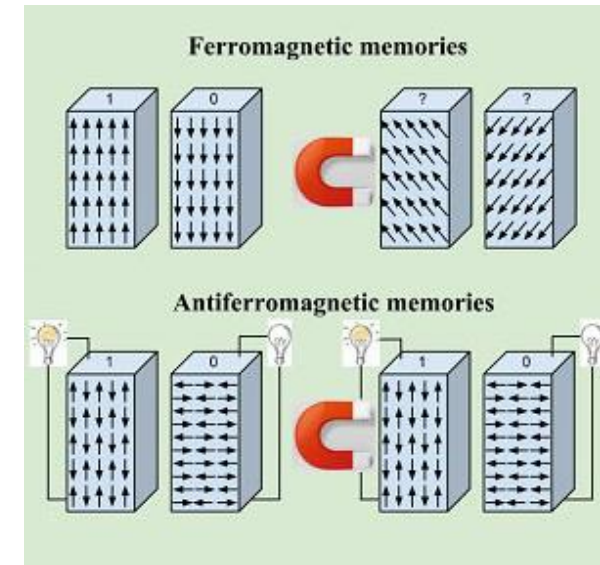


Fig. 10. — Plot for several materials of maximum usable frequency vs. effective high frequency permeability.

Difficulties associated with control of AFM spin waves

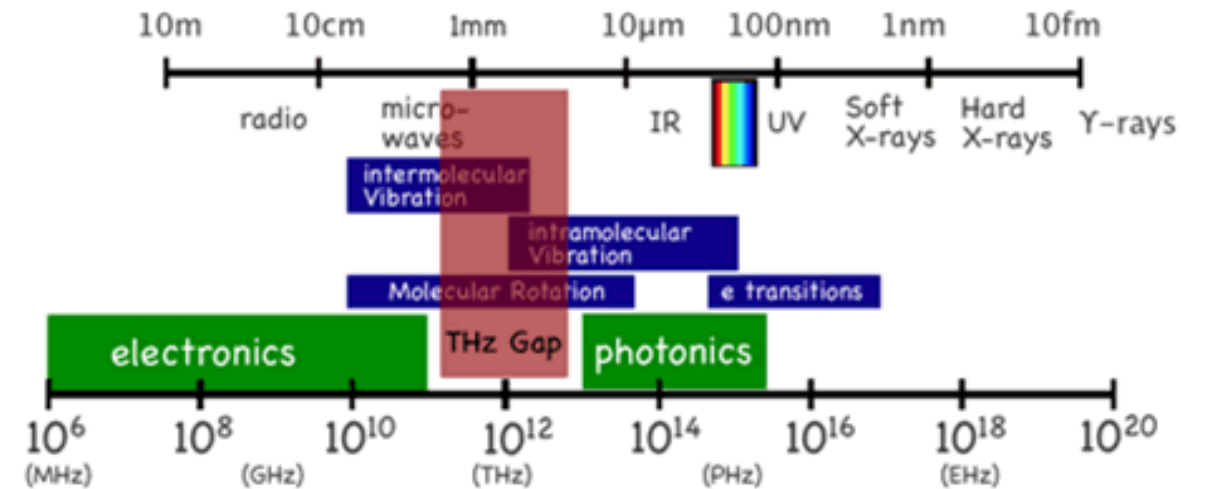
Although AFM materials have these very promising qualities, it is these very properties that make both control and detection of the spin wave dynamics difficult.

- ❑ Magnetic field switching sources in the GHz range are widely available, but the lack of materials with magnetic switching in the THz range makes it difficult to match the resonance of AFM materials
- ❑ The lack of net magnetization, makes direct measurement (such as pulsed inductive magnetometer PIMM) difficult and therefore, indirect measurements such as detection of Faraday rotations (to be described later) need to be employed

Terahertz Electromagnetic Radiation

One way to circumvent the lack of fast magnets to excite AFM materials is the use of the THz EM spectrum

- ❑ Exists in the technology gap between electronics and photonics
- ❑ Elementary processes such as lattice vibrations, plasma oscillations, and superconducting quasiparticles can be accessed in this frequency regime
- ❑ Although these are mediated by electric dipole interactions, this work demonstrates the ability to control spin waves through magnetic dipole interactions



Terahertz Generation

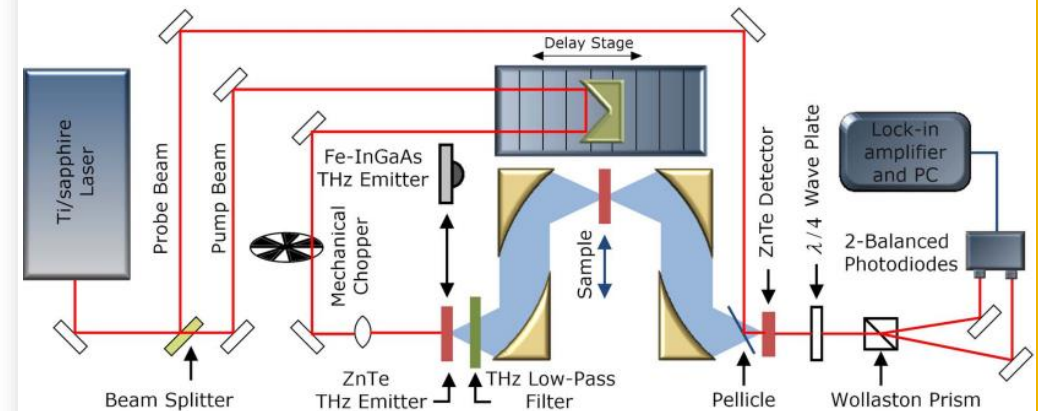
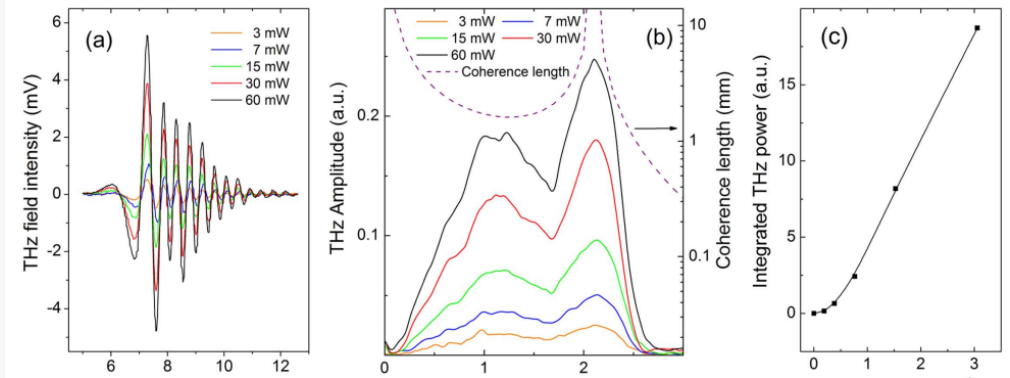
- Generation of THz waves by optical rectification in nonlinear crystals involves a difference-frequency mixing process between all possible frequency modes included in the spectrum of the incident laser pulse
- The difference frequency mixing induces electric dipoles oscillating and emitting electromagnetic waves at the beating frequencies in the THz range. The oscillating dipoles in the optical rectification process can be in electronic and acoustic phonons traveling inside the crystal at certain quantized energy states

Peak emission of terahertz waves from (110)-oriented ZnTe by interacting phase-matched phonon resonances

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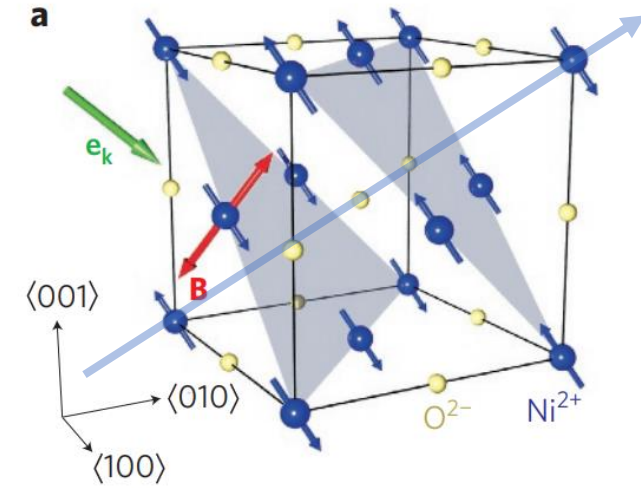


Choosing the Right Material

- ❑ Antiferromagnetic (of course) – AFM materials display THz spin wave resonance
 - ❑ Insulating – Utilizing EM radiation for as a magnetic pulse makes the electric field unavoidable. Therefore, insulating materials should be considered
 - ❑ Nonthermal mechanism for spin wave excitation – Although prior to this work, the mechanisms of spin wave excitation were not fully understood, the process was observed to be nonthermal allowing for faster response
-

Nickle Oxide (Magnetic Structure)

- NiO is an AFM material with FM planes stacked along the $[111]$ direction. In the figure shown, one of the possible spin orientations are depicted. The spins in one plane align in the $[11\bar{2}]$ direction and the adjacent planes point anti parallel in the $[\bar{1}\bar{1}2]$ direction.
- There are 3 possible spin orientations for each T-domain giving rise to 12 orientational domains



	<i>S</i>		<i>T</i>	
	$T_1[1, 1, 1]$	$T_2[1, 1, \bar{1}]$	$T_3[1, \bar{1}, 1]$	$T_4[\bar{1}, 1, 1]$
S_1	$[1, 1, \bar{2}]$	$[1, 1, 2]$	$[1, 2, 1]$	$[2, 1, 1]$
S_2	$[\bar{2}, 1, 1]$	$[1, \bar{2}, \bar{1}]$	$[\bar{2}, \bar{1}, 1]$	$[\bar{1}, 1, \bar{2}]$
S_3	$[1, \bar{2}, 1]$	$[\bar{2}, 1, \bar{1}]$	$[1, \bar{1}, \bar{2}]$	$[\bar{1}, \bar{2}, 1]$

Distribution of antiferromagnetic spin and twin domains in NiO

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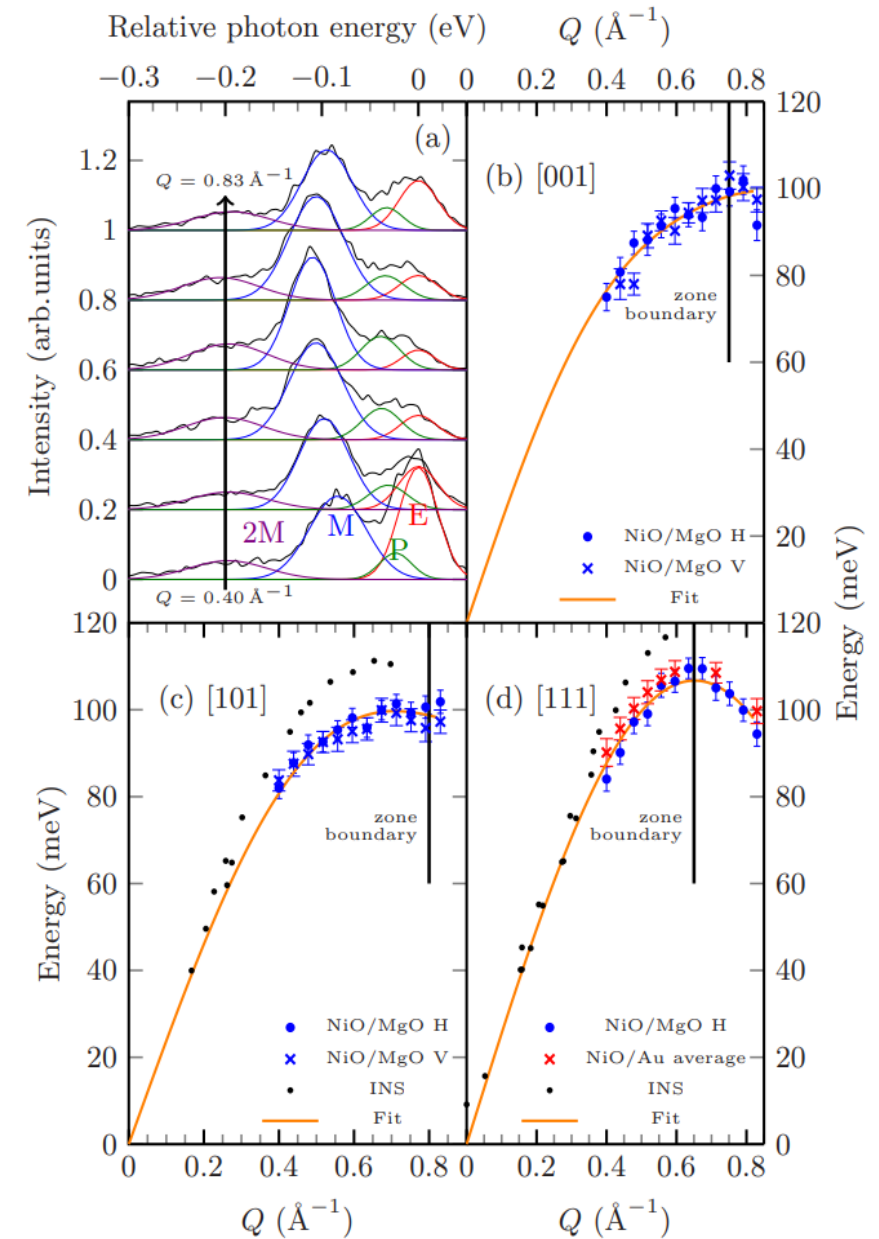
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Nickle Oxide Antiferromagnetism

Linear dispersion of magnons is indicative of AFM materials. Common ways to probe dispersion curves use inelastic neutron scattering or more recently resonant inelastic x-ray scattering

FIG. 5. (a) Low-energy features of the spectra in Fig. 3(a) with the decomposition into four Gaussians corresponding to elastic peak, phonons, magnon, and double magnon. They are labeled E, P, M, and 2M, respectively, on the lowest spectrum. (b) Spin wave dispersion along the [001] direction. (c) Spin wave dispersion along the [101] direction. (d) Spin wave dispersion along the [111] direction. The fits result from linear spin wave theory, as explained in the text. The INS data are taken from Ref. [10].



Three-dimensional dispersion of spin waves measured in NiO by resonant inelastic x-ray scattering

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Nickel Oxide Magnetic Resonance in the THz range

To take advantage of the fast THz switching speed of AFM materials, the resonance peak needs to be identified

- Far infrared inelastic spectroscopy was employed to locate the AFM resonance peak at different temperatures

Antiferromagnetic Resonance in NiO in Far-infrared Region

By Hisamoto KONDOH
 Department of Physics
 University of Osaka Prefecture
 (Received June 23, 1960)

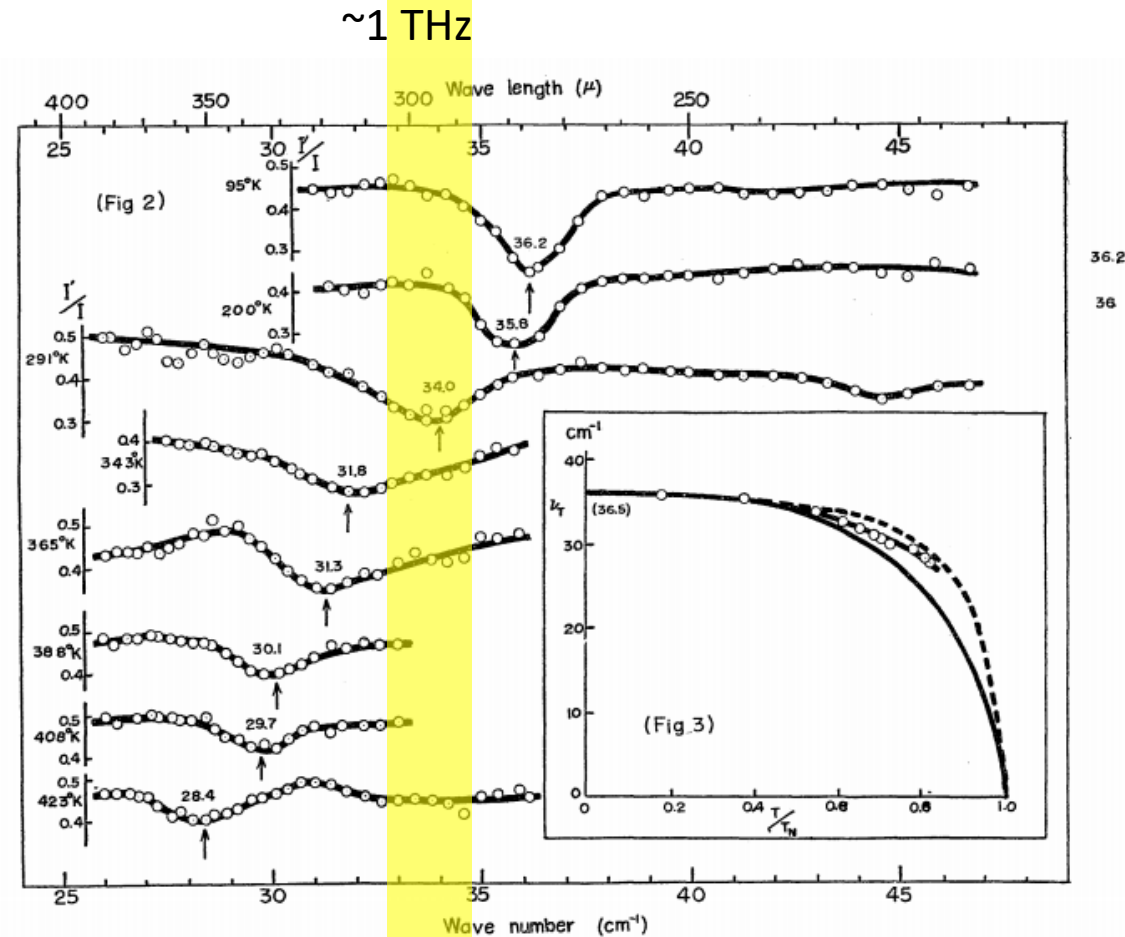


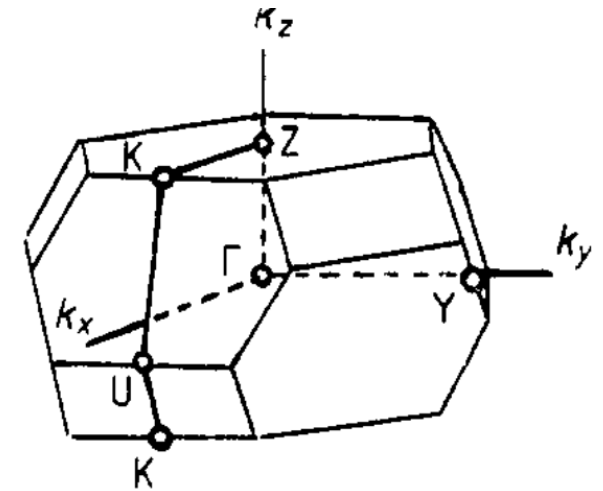
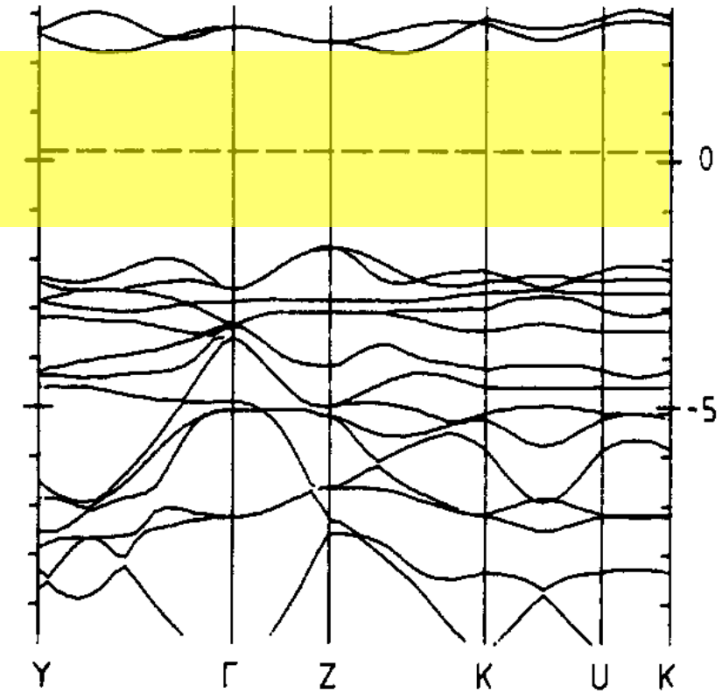
Fig. 2. Transmission rate, I'/I , through a sample having a dozen crystals, at various temperature. Incident beam is perpendicular to the (111) surfaces of the crystals.

Fig. 3. Temperature dependence of the frequency of the absorption maximum. Experimental points are plotted from the data of Fig. 2. A thin curve is the Brillouin curve with $S=1$ and a dotted curve the variation of sublattice magnetizations with temperature obtained from the neutron diffraction experiment by Roth. These two curves are so normalized as to coincide with our experimental value of wave number at 0°K and at room temperature, respectively.

Nickel Oxide (Insulating)

- Nickel Oxide (NiO) is often referred as the prototypic AFM material since its properties are well understood.
- Since EM radiation is being used, and therefore the electric field can not be avoided, it is important to use an insulating material (such as NiO) rendering the electric field effects negligible
- In NiO free carrier and lattice absorption are negligible

Large Band Gap of NiO



Band-structure description of Mott insulators (NiO, MnO, FeO, CoO)

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Nonthermal Response

Raman scattering in nanosized nickel oxide NiO

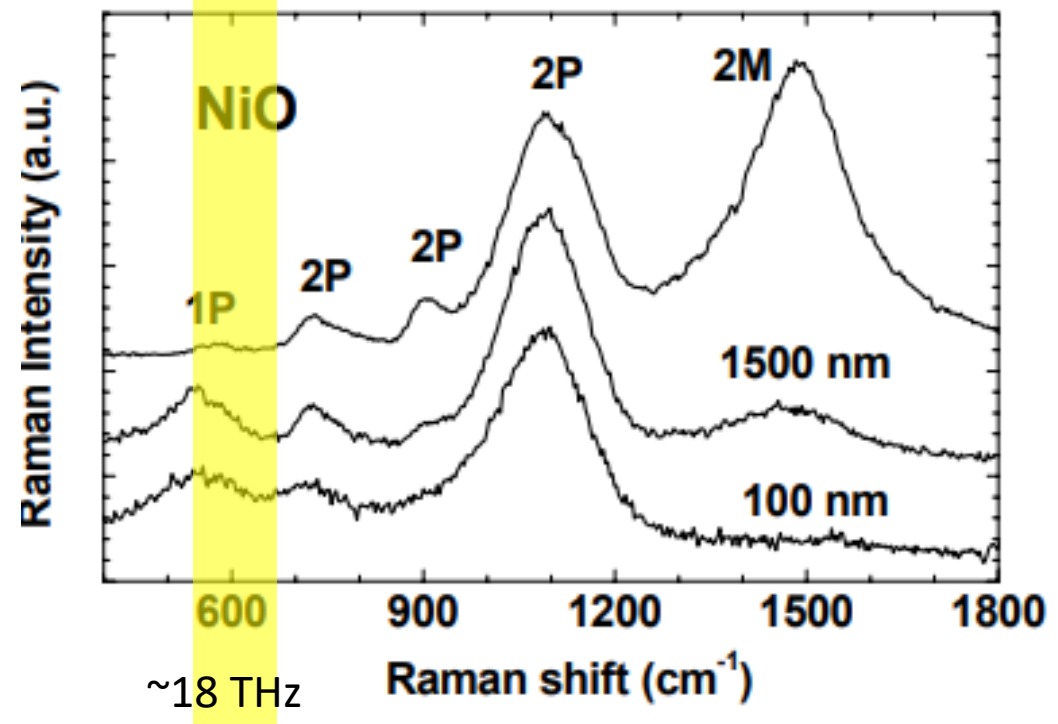
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- As stated previously, free carriers and lattice absorptions are negligible due to NiO's insulating properties
- All optical phonon resonances are located above 12 THz, above the excitation frequency used in this experiment

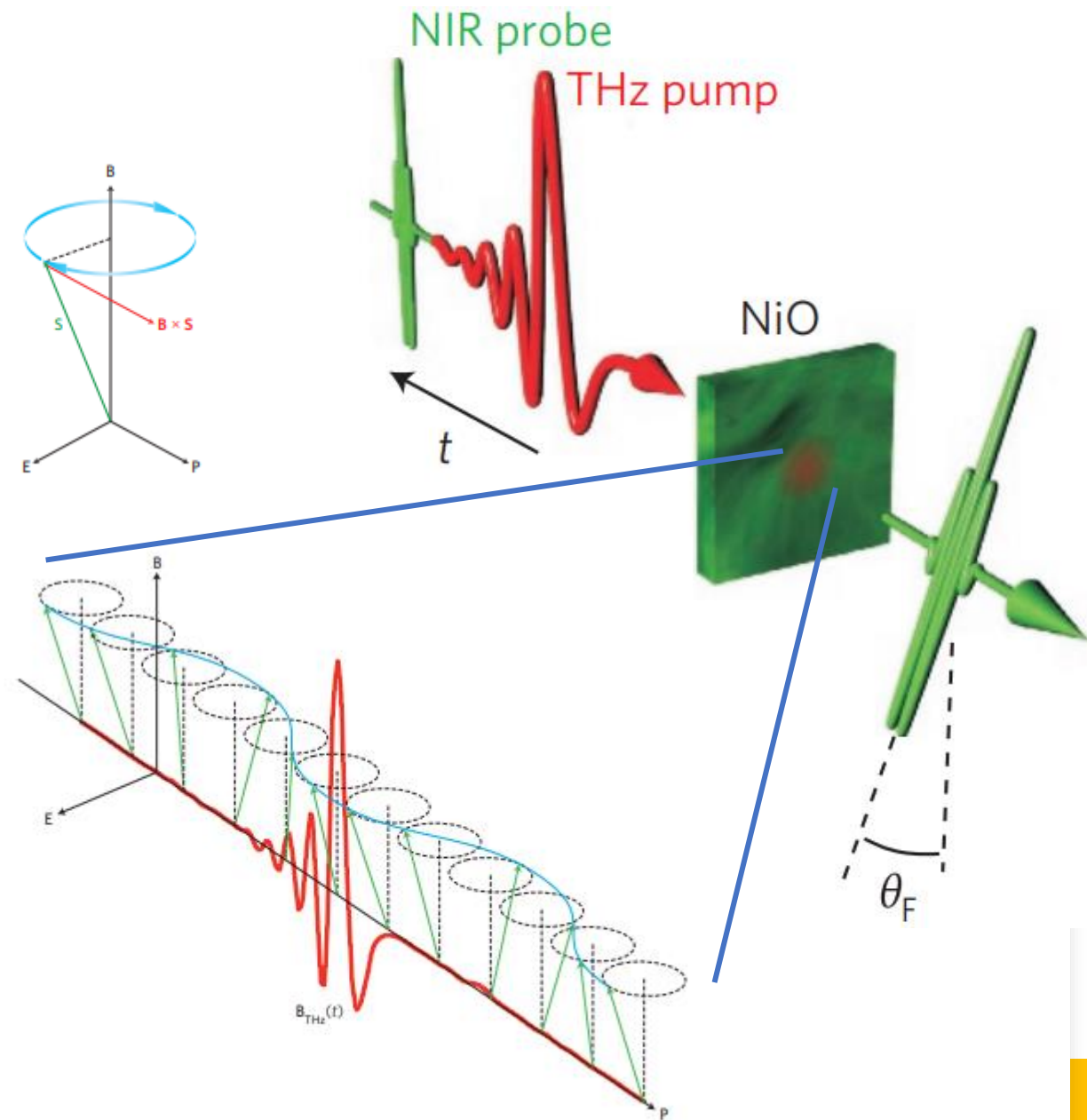


Let's Get to
the Paper!!

- Experimental Design
- Theoretical Model
- Results

Experimental Design

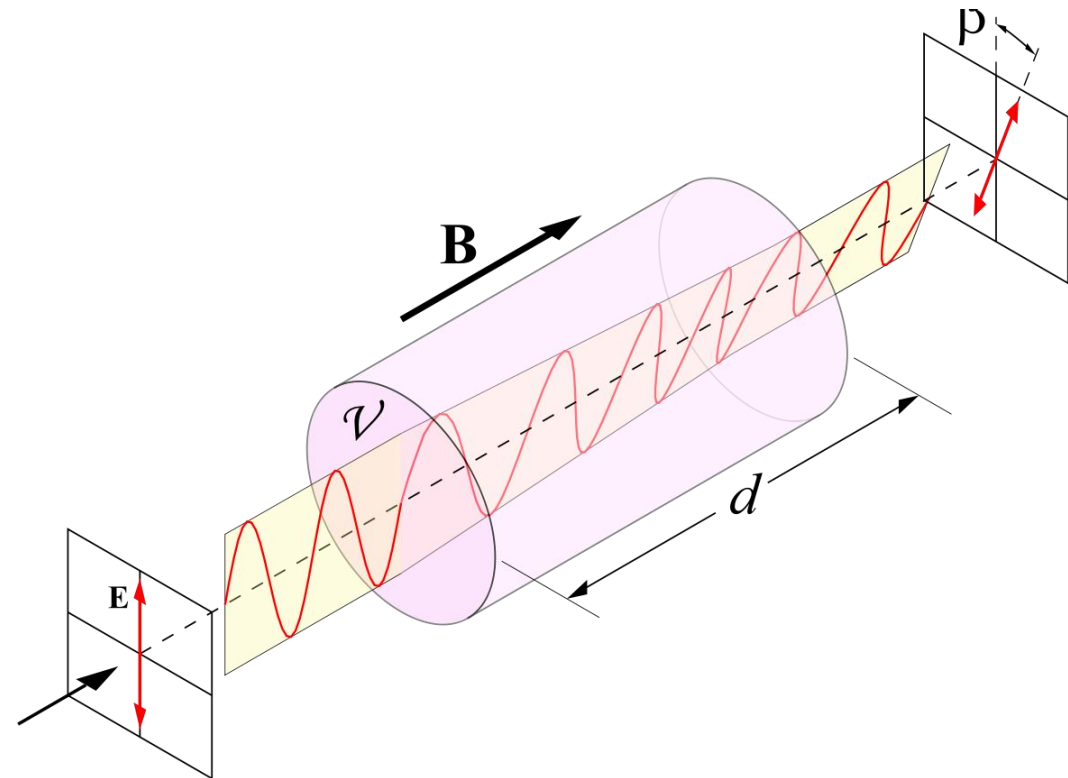
- ❑ Single-cycle THz pump NIR probe spectroscopy was employed to observe the AFM spin wave dynamics within NiO single crystals by controlling the delay between the pump and probe pulse.
- ❑ Monitoring the rotation of the linearly polarized probe pulse caused by the Faraday effect allows for the observation of the spin dynamics on the picosecond timescale



Faraday Rotation

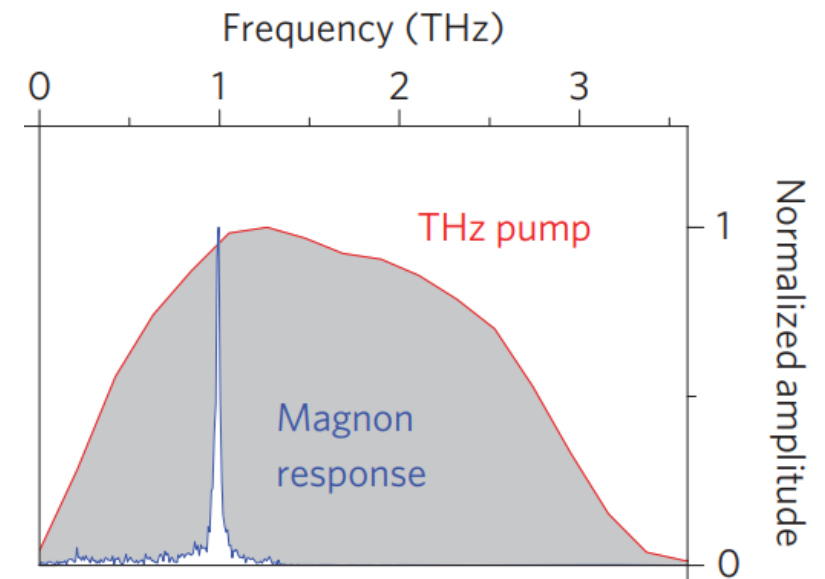
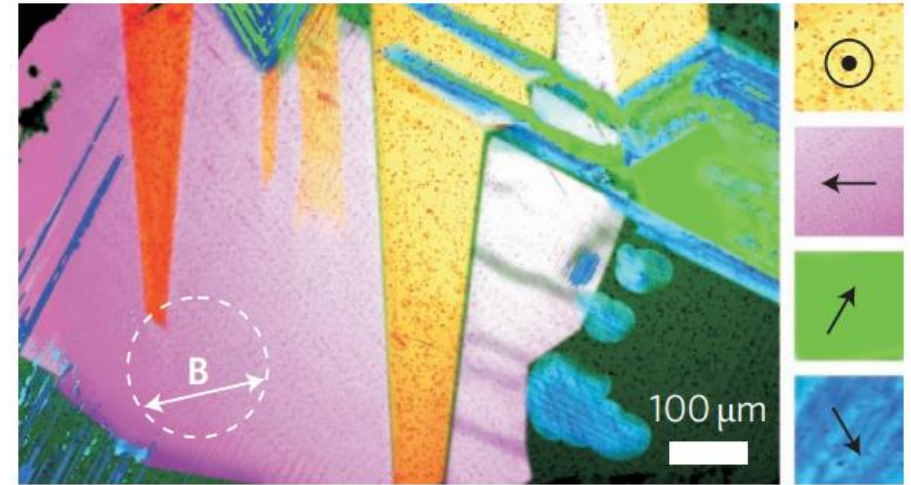
Faraday rotation causes a polarization rotation which is proportional to the projection of the magnetic field along the direction of the light propagation

$$\theta_F(t) = Vd \langle \mathbf{e}_k \cdot \mathbf{M}(t) \rangle$$



Experimental Design

- ❑ To ensure only one T-domain was being influenced, polarized microscopy was implemented to identify the orientations of the domains within the crystal
- ❑ The spectral bandwidth of the THz pulse ranged from 1-3 THz which ensures the overlap with the high frequency magnon resonance



Theoretical Description of Spin Waves in NiO

The electromagnetic waves interact through a pure magnetic interaction by exerting a Zeeman torque on each spin

$$\mathbf{G} = \gamma \mathbf{S} \times \mathbf{B}$$

The Hamiltonian of the system can be written as follows

$$H = -J\mathbf{S}_1 \cdot \mathbf{S}_2 + \sum_{i=1}^2 [D_x S_{ix}^2 + D_y S_{iy}^2] + \gamma \mathbf{B}(t) \cdot \sum_{i=1}^2 \mathbf{S}_i$$

Spin-spin exchange interaction between the two sublattices. The interaction coefficient is negative to account for the AFM

Accounts for the anisotropy mediated by the spin-spin dipolar interaction and the previously mentioned rhombohedral distortion caused by the spin orbit interaction

Zeeman energy due to the applied magnetic field interacting with the individual spins

Theoretical Description of Spin Waves in NiO

We can convert to the Heisenberg picture using the following equation

$$\frac{d}{dt} (S_i(t))_H = \frac{i}{\hbar} [H, S] + \frac{d}{dt} (S_i(t))_S$$

We obtain a system of Heisenberg equations

$$\frac{d}{dt} \mathbf{S}_i = -\frac{\gamma}{1+\alpha^2} \left[\mathbf{S}_i \times \mathbf{B}_i^{\text{eff}} - \frac{\alpha}{S_i} \mathbf{S}_i \times (\mathbf{S}_i \times \mathbf{B}_i^{\text{eff}}) \right]$$

Where $\mathbf{B}_i^{\text{eff}} = \mathbf{B}(t) + (-JS_{3-i} + D_x S_{ix} \mathbf{e}_x + D_y S_{iy} \mathbf{e}_y) / \gamma$ And $\gamma = g\mu_B / \hbar$

The set of coupled equations can be solved numerically for S_1 and S_2 and using

$$\mathbf{M} = n\gamma\hbar(\mathbf{S}_1 + \mathbf{S}_2) \quad \text{Allows one to calculate the magnetization}$$



Theoretical Description of Spin Waves in NiO

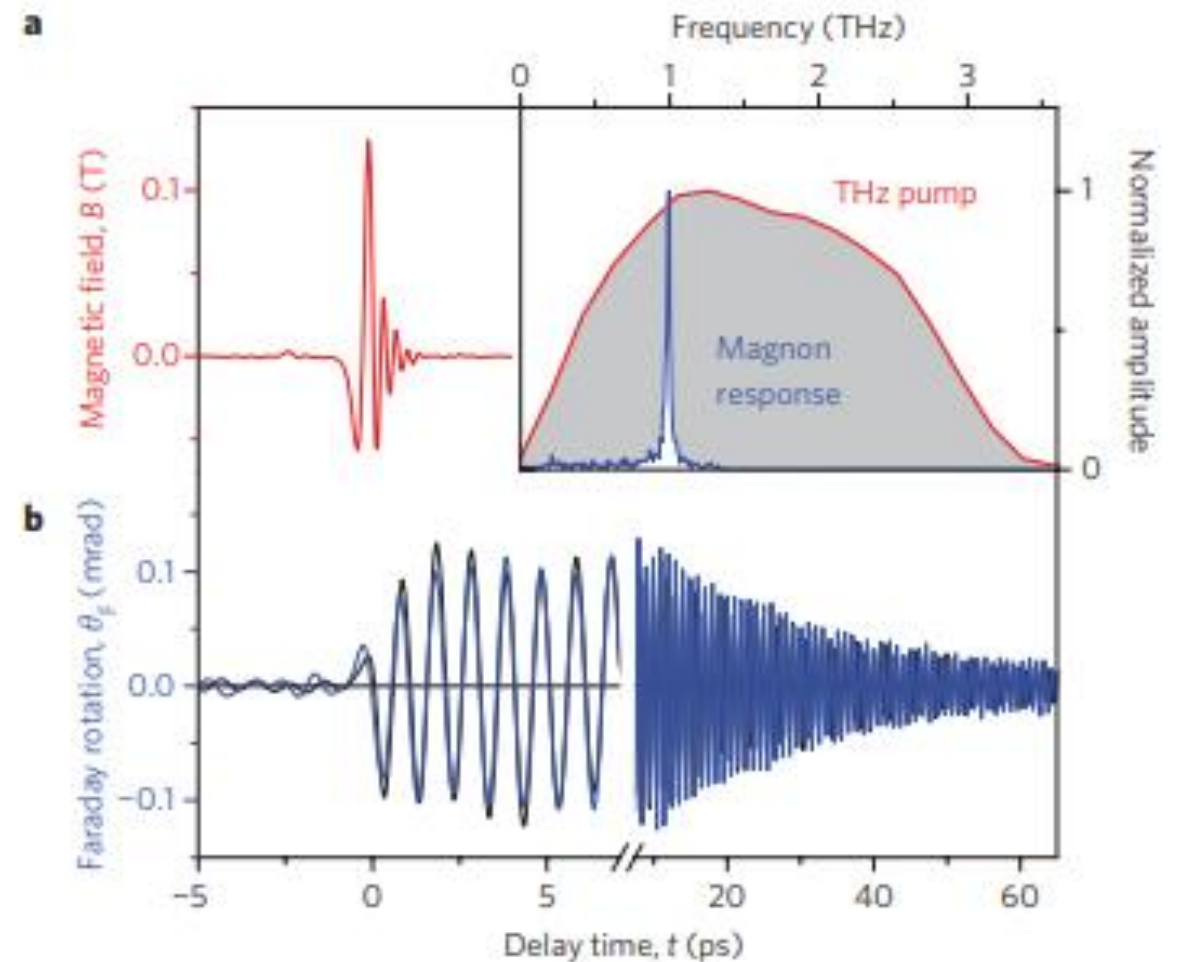
Lastly, the Faraday rotation can be calculated with $\theta_F(t) = Vd\langle \mathbf{e}_k \cdot \mathbf{M}(t) \rangle$

Where V is the magneto-optic Verdet constant, d is the thickness of the sample, and the angle brackets indicate an average of the irradiated volume

For each T-domain, an average over the 3 S-domains was performed

Measuring the dynamics of spin waves

- Period of oscillation measured to be 1 ps
- Reaches max amplitude in 3 ps
- Fourier transform of Faraday rotation reveals sharp peak at 1 THz
- Exponential decay with a time constant of 29 ps
- Black curve represents simulated spin precession based on the model described above
- Blue curve represents experimental data



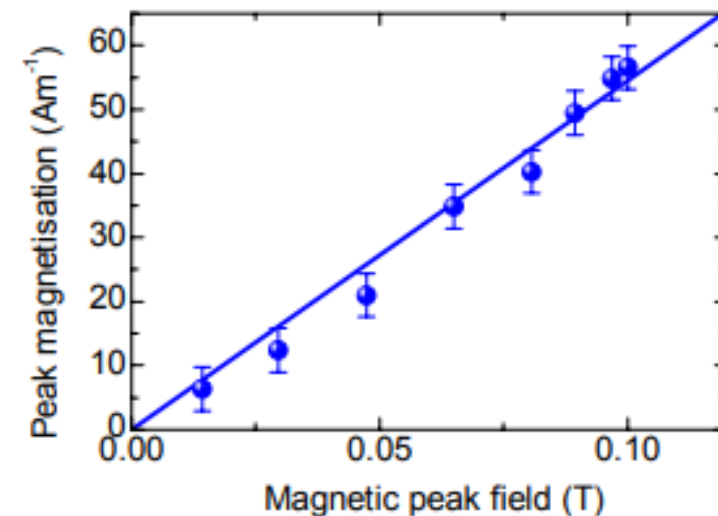
Ruling out Magneto-Electric Effect

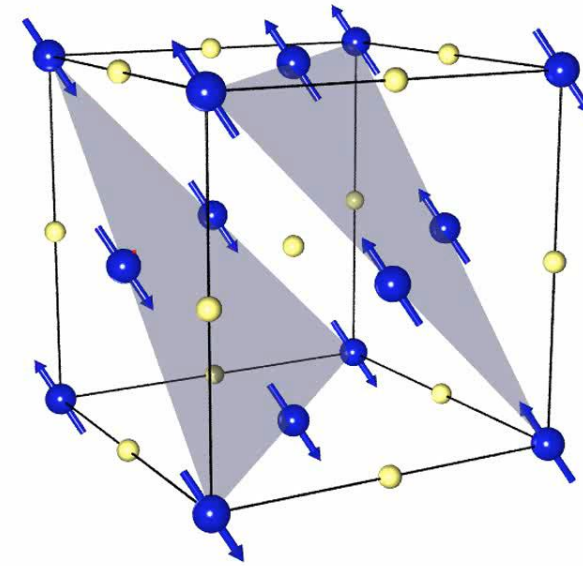
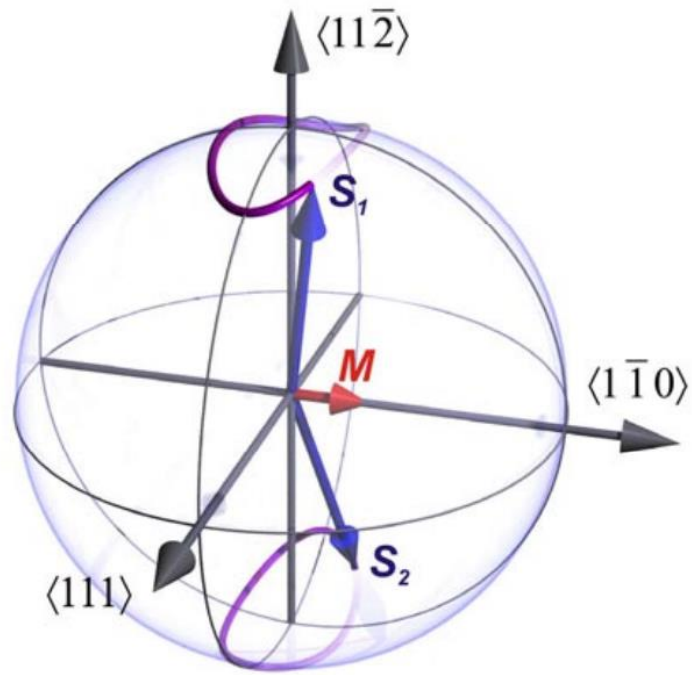
Some evidence of THz irradiation having a pure magnetic interaction with NiO lies in the linear response of the magnetization with respect the magnetic field strength

Multiferroic and Magnetoelectric Materials

Wolfgang Kleemann and Christian Binek

The linear ME effect was first verified on the rhombohedral antiferromagnet chromia, $\alpha\text{-Cr}_2\text{O}_3$, [4] and theoretically explored by Landau and Lifshitz [15]. They found that quite stringent symmetry properties must be fulfilled, namely time and spatial inversion symmetry, T and I , respectively, have to be broken.





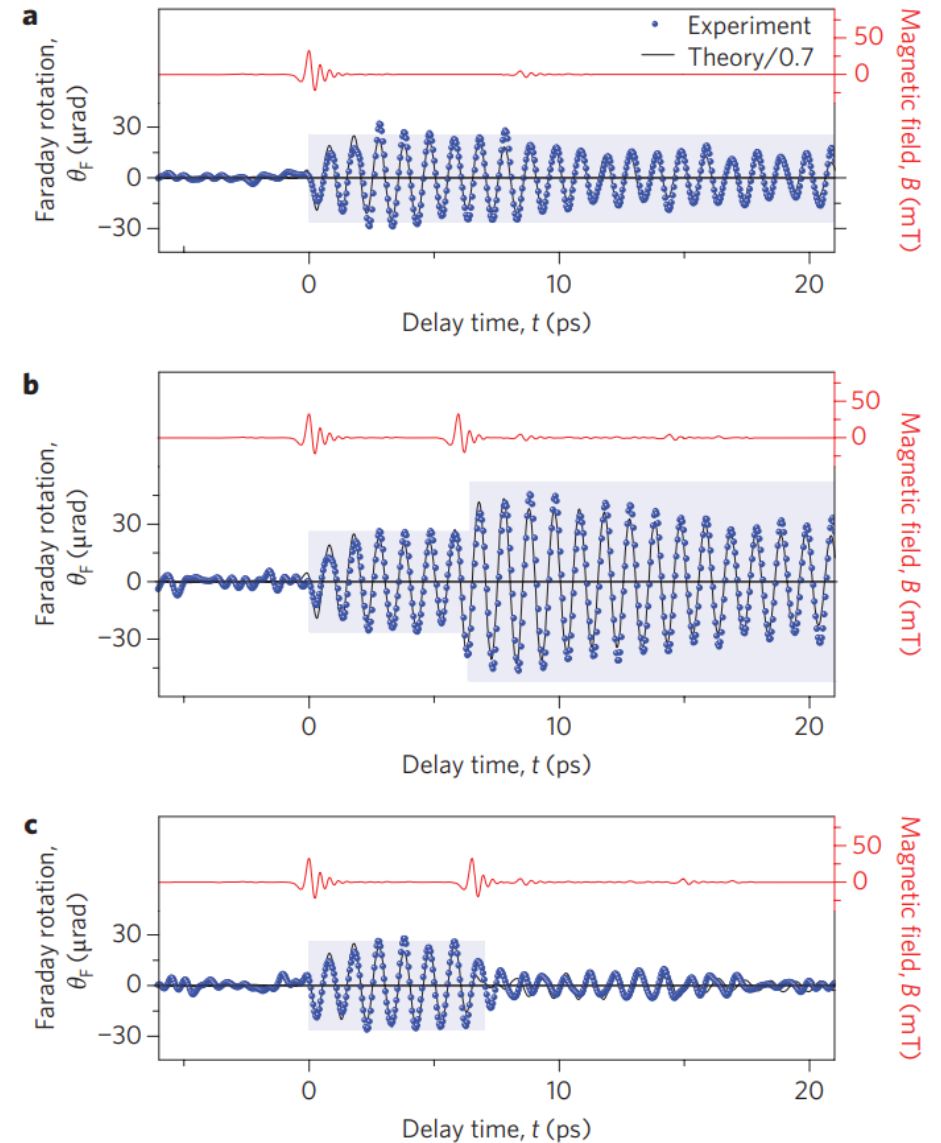
Simulation of spin precession

- ❑ Out of plane component oscillates anti-parallel in each plane
- ❑ In plane component contributes to a net transient magnetization

Control over spin wave dynamics

With the delivery of multiple pump THz pulses, control over the spin wave dynamics was demonstrated

- With a delay of 6 ps a doubling of the faraday rotation amplitude was observed
 - This induces a torque in phase with the spin wave precession
- With a delay of 6.5 ps, the spin dynamics are effectively switched off
 - This induces a torque out of phase with the spin wave precession



What came next

Mechanistic Determination

- ❑ Magnetic Difference Frequency Generation
- ❑ Inverse Faraday Effect – A static magnetization is induced by an external oscillating electrical field (i.e. EM radiation)
- ❑ Inverse Cotton Mouton Effect – magnetization induced in a medium by a non-resonant linearly polarized light propagating in the presence of a transverse magnetic field

Selection Rules for Light-Induced Magnetization of a Crystal with Threefold Symmetry: The Case of Antiferromagnetic NiO

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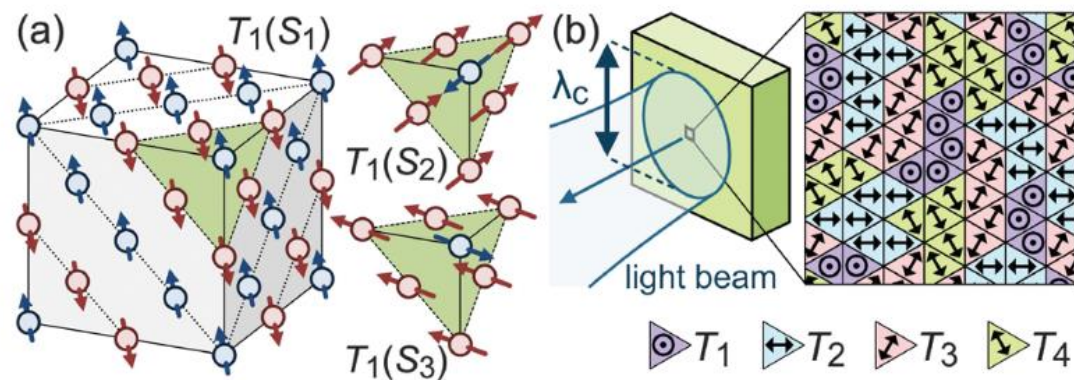
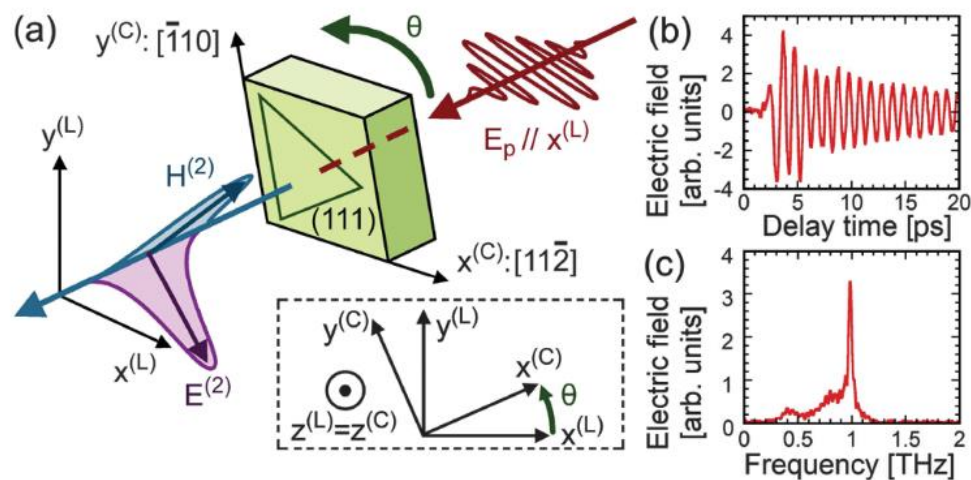
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We propose Raman-induced collinear difference-frequency generation (DFG) as a method to manipulate dynamical magnetization. When a fundamental beam propagates along a threefold rotational axis, this coherent second-order optical process is permitted by angular momentum conservation through the rotational analogue of the umklapp process. As a demonstration, we experimentally obtained polarization properties of collinear magnetic DFG along a [111] axis of a single crystal of antiferromagnetic NiO with micromultidomain structure, which excellently agreed with the theoretical prediction.

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PACS numbers: 78.20.Ls, 42.65.-k, 76.50.+g



Coherent control of terahertz radiation from antiferromagnetic magnons in NiO excited by optical laser pulses

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(Received 4 November 2011; revised manuscript received 5 March 2012; published 30 May 2012)

We investigated terahertz (THz) radiation from twinned NiO(110) single crystals excited by optical laser pulses with various polarization states. We found that the polarity of THz radiation from optically excited coherent antiferromagnetic (AFM) magnons can be controlled by switching the helicity of circular polarization and by rotating the direction of linear polarization of the laser pulses. The dependence of THz radiation on the polarization of the laser pulses suggests that AFM magnons are excited by the inverse Faraday effect in a twinned crystal with linear magnetic birefringence.

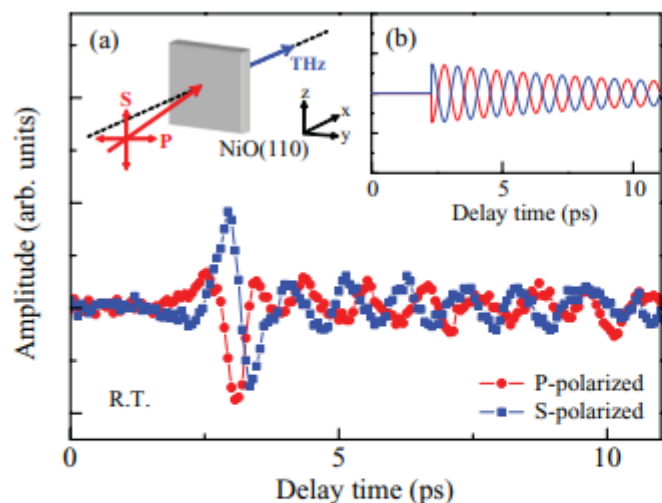


FIG. 2. (Color online) (a) Waveforms of THz radiation from NiO(110) irradiated with linearly polarized laser pulses. Circles (squares) show the case using P (S)-polarized laser pulses schematically presented in the inset. (b) Calculated waveforms in the case of linearly polarized excitation.

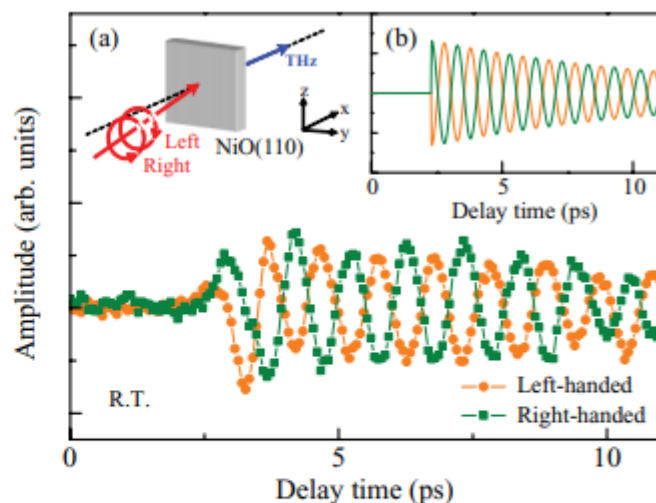


FIG. 3. (Color online) (a) Waveforms of THz radiation from NiO(110) irradiated with circularly polarized laser pulses. Circles (squares) show the case using left (right)-handed circularly polarized laser pulses schematically presented in the inset. (b) Calculated waveforms in the case of circularly polarized excitation.

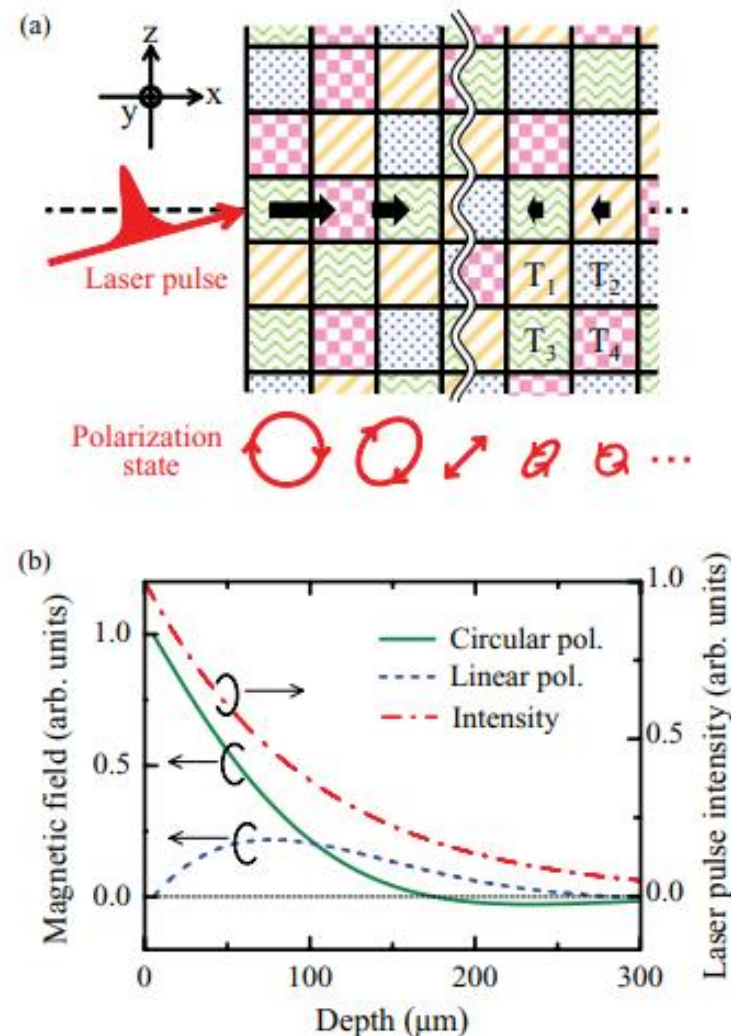


FIG. 4. (Color online) (a) Schematic of polarization states of the laser pulses in a twinned single crystal of NiO. Arrows presented on the sample show the direction and magnitude of magnetic field pulses induced by optical laser pulses. (b) Solid (dotted) curve displays the peak amplitude of the magnetic field pulse induced by circularly (linearly) polarized laser pulses as a function of the depth from the sample surface. Dashed-dotted curve shows the decay in the intensity of the incident laser pulses in the sample with a penetration depth of $\sim 100 \mu\text{m}$.

Ultrafast optical excitation of coherent magnons in antiferromagnetic NiO

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Stefan Günther,¹ Manfred Fiebig,¹ and Takuya Satoh^{1,2,4}

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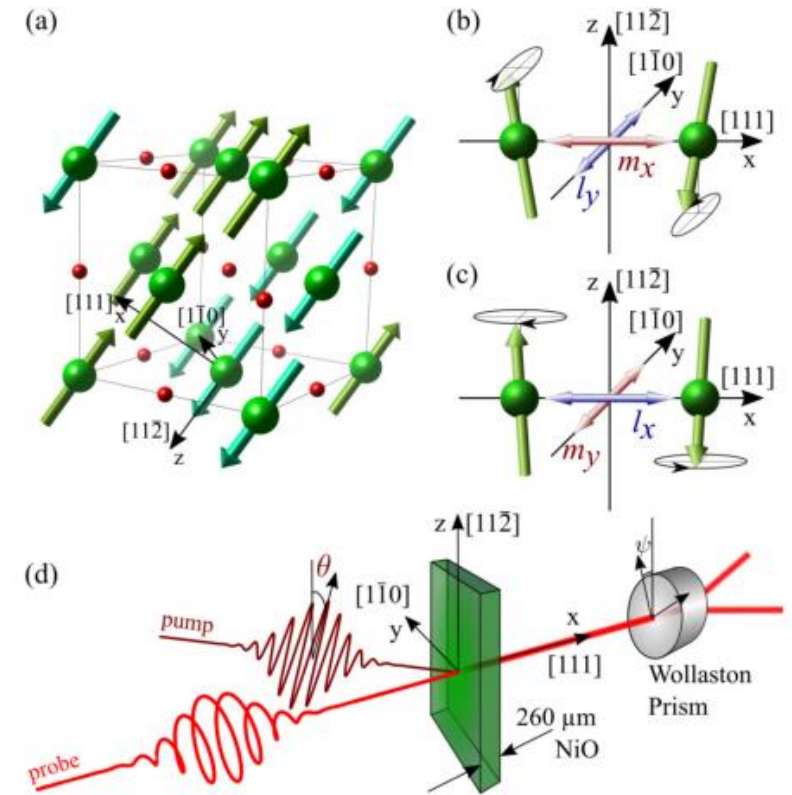
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In experiment and theory, we resolve the mechanism of ultrafast optical magnon excitation in antiferromagnetic NiO. We employ time-resolved optical two-color pump-probe measurements to study the coherent nonthermal spin dynamics. Optical pumping and probing with linearly and circularly polarized light along the optic axis of the NiO crystal scrutinizes the mechanism behind the ultrafast magnon excitation. A phenomenological symmetry-based theory links these experimental results to expressions for the optically induced magnetization via the inverse Faraday effect and the inverse Cotton-Mouton effect. We obtain striking agreement between experiment and theory that, furthermore, allows us to extract information about the spin domain distribution. We also find that in NiO the energy transfer into the magnon mode via the inverse Cotton-Mouton effect is about three orders of magnitude more efficient than via the inverse Faraday effect.



Optimizing ZnTe THz Generation by Phonon-Polariton Phase Matching

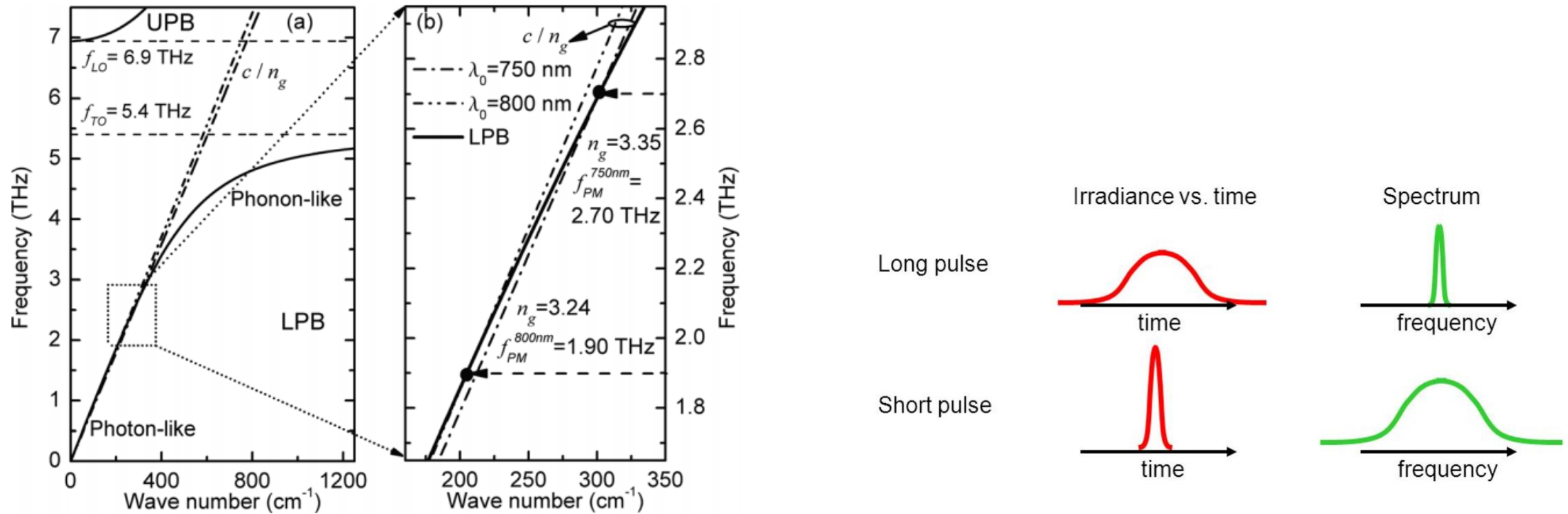


FIG. 3. (a) Phonon-polariton dispersion of ZnTe. (b) The detail of the LPB in middle region. The light lines representing optical pulses of $\lambda_0 = 750$ nm and $\lambda_0 = 800$ nm intersect the LPB, and the intersections represent the phase-matched frequencies of $f_{PM}^{750\text{nm}} = 2.70$ THz and $f_{PM}^{800\text{nm}} = 1.90$ THz, respectively.