Probing optical responses of chiral phonons using continuous-wave terahertz spectroscopy

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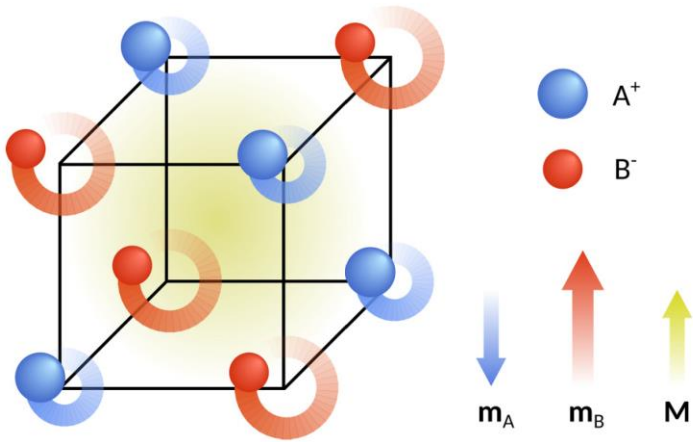
Outline

- Motivation
- Experimental Setup and Method
- Results
- Future Plan

Dynamical multiferroicity



Phys. Rev. Mater. 1, 014401 (2017)

- Magnetization is induced by time-dependent polarization $\partial_t \boldsymbol{P} \rightarrow \boldsymbol{M}$
- Phonons can carry orbital angular momentum
- Circular ionic motion can generate effective magnetization

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Orbital magnetic moments of phonons

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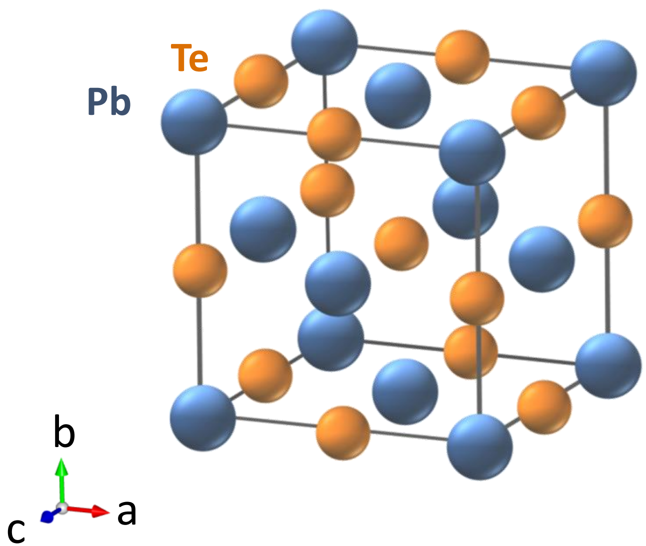
Compound	ν_0	Z	d/d_0	N	μ_{ph}	M	$\Delta\Omega/\Omega_0$
Rocksalt structure[a]							
BaO	3.0	0.7	9	6	0.15	1.0	0.0002
CsF	3.6	0.3	3	1	0.06	0.1	0.00008
CsH	11.9	1.1	25	15	1.12	16.8	0.0005
LiI	4.8	0.5	7	3	0.18	0.5	0.0002
MgO	11.7	0.6	4	5	0.04	0.2	0.00002
PbO[b]	7.7	1.2	7	16	0.13	2.1	0.00008
PbS	2.2	0.8	8	8	0.12	1.0	0.0003
PbSe	1.6	0.6	4	5	0.04	0.2	0.0001
PbTe	1.4	0.7	3	5	0.02	0.1	0.00006
SnTe	1.0	1.0	5	13	0.004	0.1	0.00002

PbTe

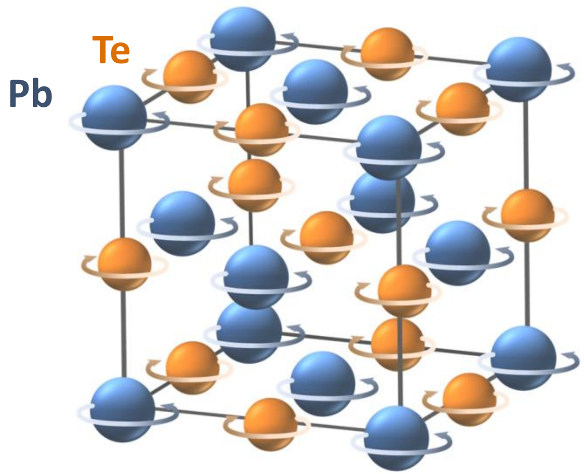
PbTe crystal structure



- Infrared-active transverse optical phonon modes are triply degenerate.
- The circular motion of ions carries orbital angular momentum.
- Phonon Zeeman splitting and magnetic circular dichroism.

PbTe

PbTe crystal structure



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Soft chiral phonons in PbTe

PHYSICAL REVIEW LETTERS **128**, 075901 (2022)

Magnetic Control of Soft Chiral Phonons in PbTe

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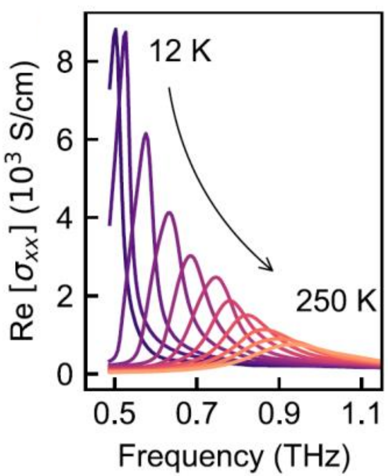
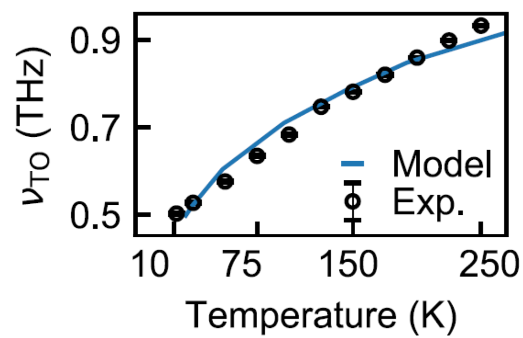
- TO phonons probed using THz time-domain spectroscopy
- TO peak shifts to higher frequency as temperature increases
  → phonon softening
- Strong quartic anharmonicity in PbTe leads to mode softening.

Soft TO phonon mode

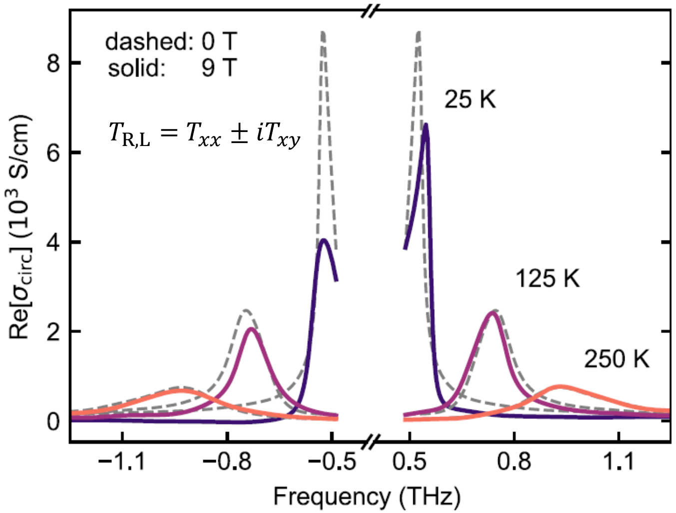


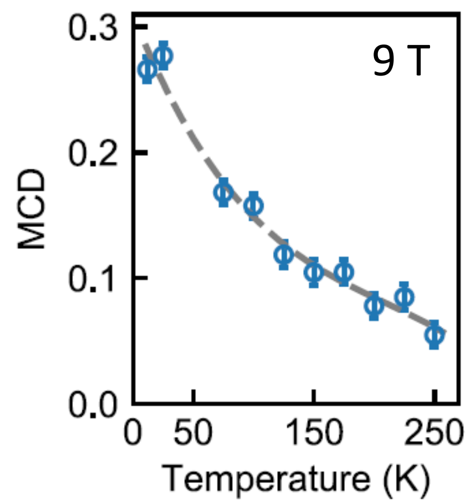


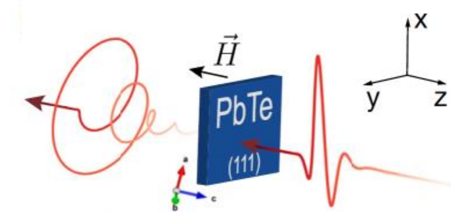
Anharmonic phonon Hamiltonian

$$\mathcal{H}_{\rm ph} = \frac{P^2}{2m} + \frac{1}{2}kX^2 + \frac{1}{4}\lambda X^4$$

Magnetic Circular Dichroism in PbTe







- Extracted left- and right-circularly polarized responses from THz transmission

- TO phonons show differential absorption for LCP and RCP light → Magnetic circular dichroism

Magnetic circular dichroism

$$\mathrm{MCD} = \frac{\sigma_R - \sigma_L}{\sigma_R + \sigma_L}$$

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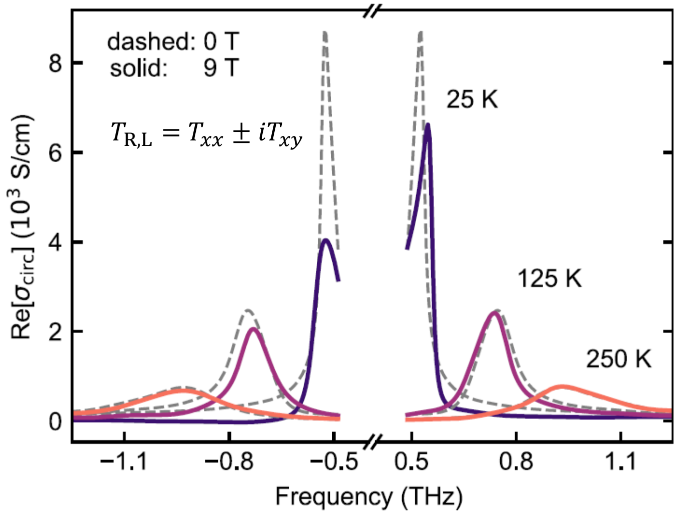
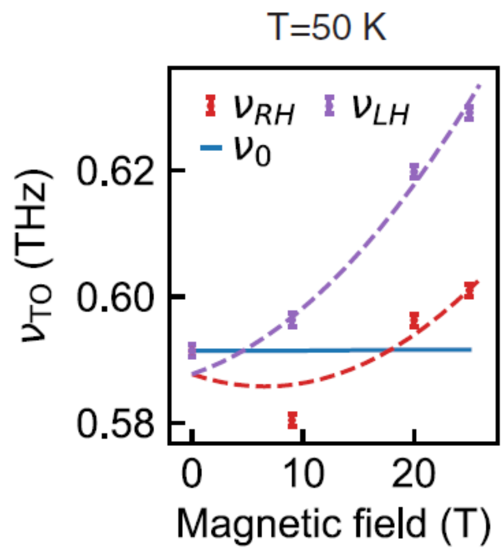
Phonon Zeeman Splitting in PbTe





- Phonon peaks split and shift under magnetic field
  → Zeeman splitting
- g-factor is larger than harmonic DFT prediction

TO phonon energy $E_{\pm}(H_z) = E_0 \pm g^* \mu_B \mu_0 H_z + \sigma_{dia} \mu_0^2 H_z^2$

Zeeman effect Diamagnetic shift

$\boldsymbol{g^* \sim 4 \times 10^{-2}} > 6 \times 10^{-5}$

9 T with anharmonic phonons

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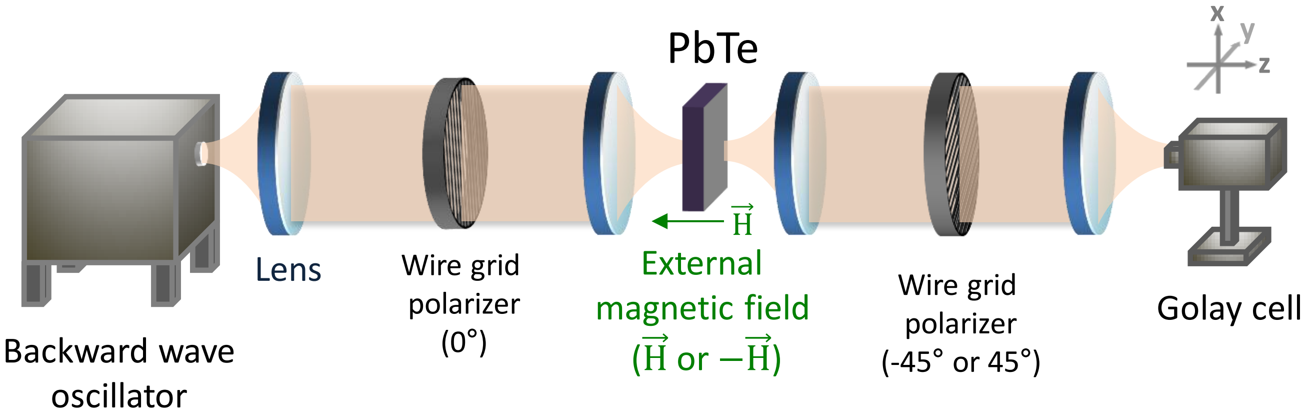
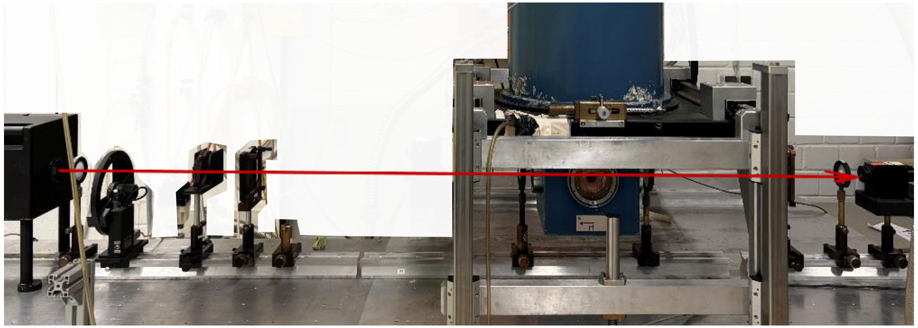
Experimental Setup

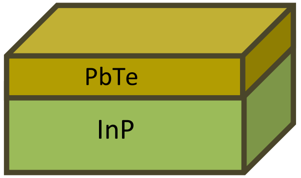
Frequency-domain THz spectroscopy



Frequency range: 1 – 40 cm^{-1}
Temperature range: 2 – 300 K
Magnetic field: up to 7 T



PbTe thin films



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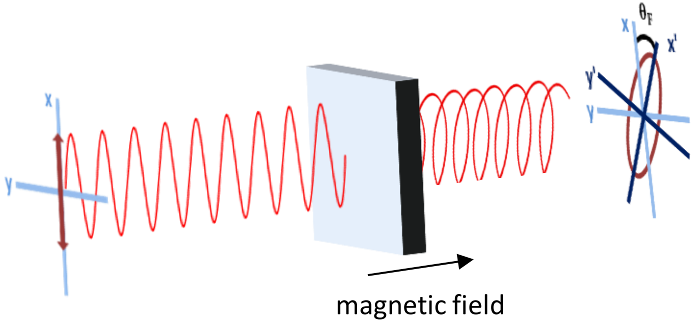
Faraday rotation

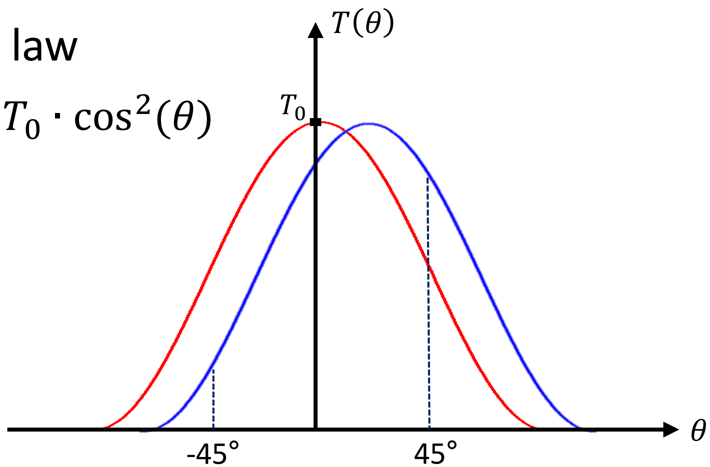
Faraday rotation



$$\theta_F(\omega, H) = \tan^{-1}\left(\frac{\sqrt{T_{-45^\circ}(\omega, H)} - \sqrt{T_{45^\circ}(\omega, H)}}{\sqrt{T_{-45^\circ}(\omega, H)} + \sqrt{T_{45^\circ}(\omega, H)}}\right)$$

Malus's law

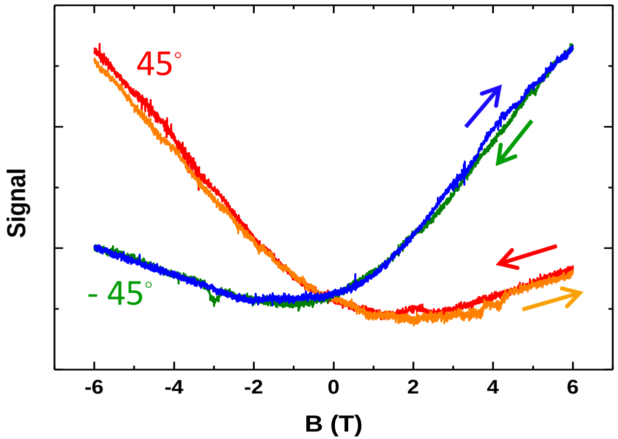
$$T(\theta) = T_0 \cdot \cos^2(\theta)$$



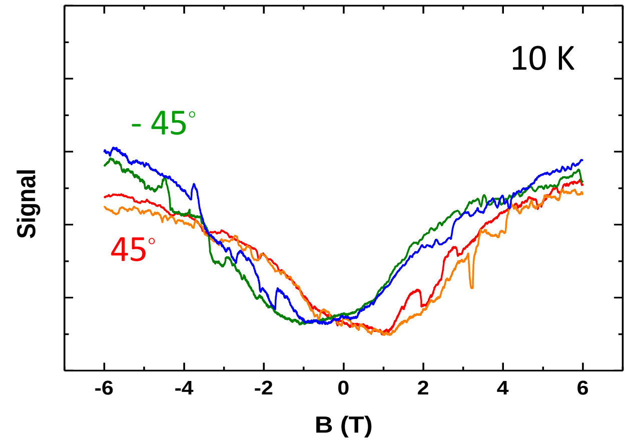
- Faraday angle extracted from intensity at +45° and –45°
- ±45° maximizes sensitivity to small Faraday angle

Experimental Data

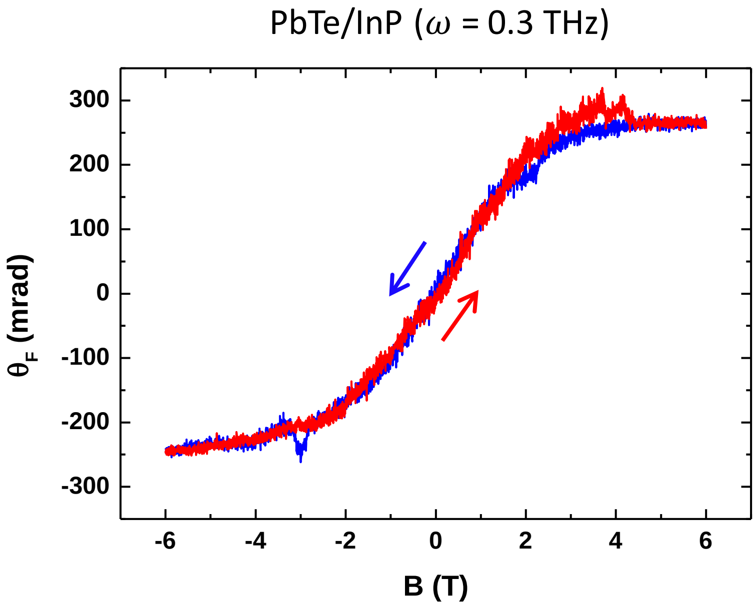
PbTe/InP (ω = 0.3 THz)

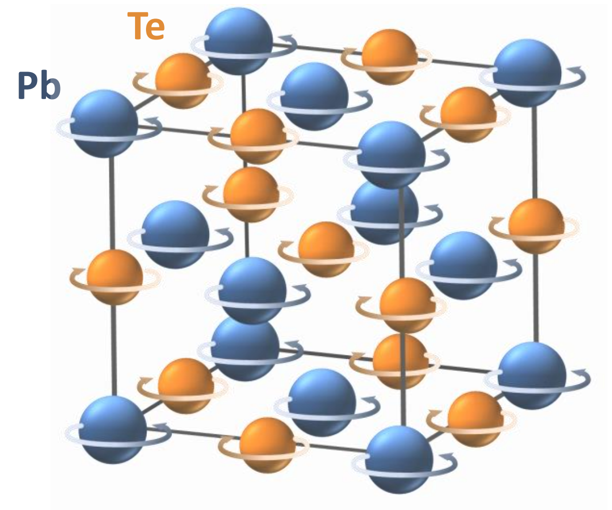


InP (ω = 0.3 THz)



Faraday Rotation

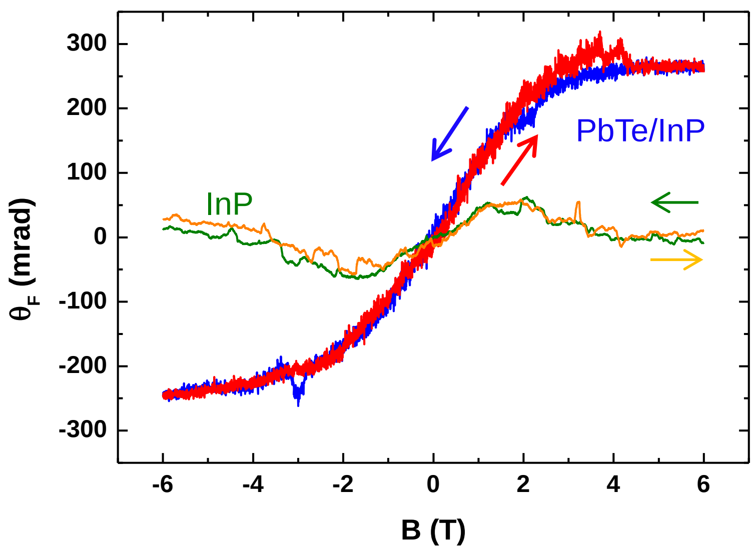


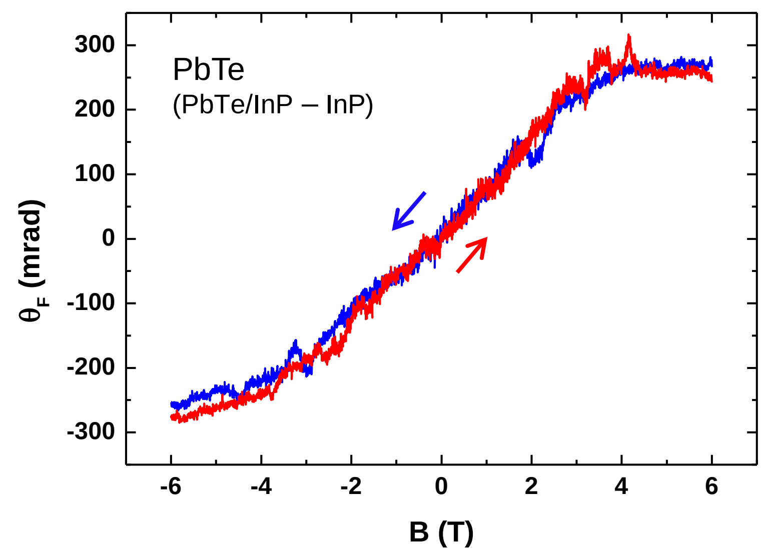


- The magneto-optical response shows no hysteresis and exhibits strong Faraday rotation under magnetic field.
- This behavior is possibly related to chiral phonon.

Faraday Rotation

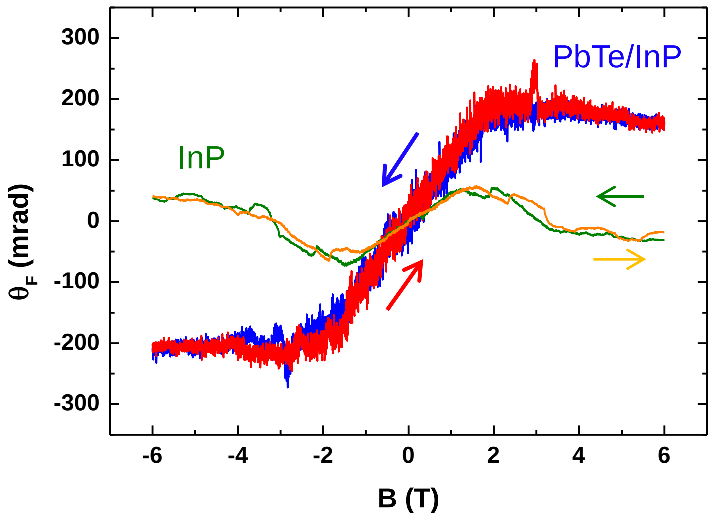
PbTe/InP (ω = 0.3 THz)

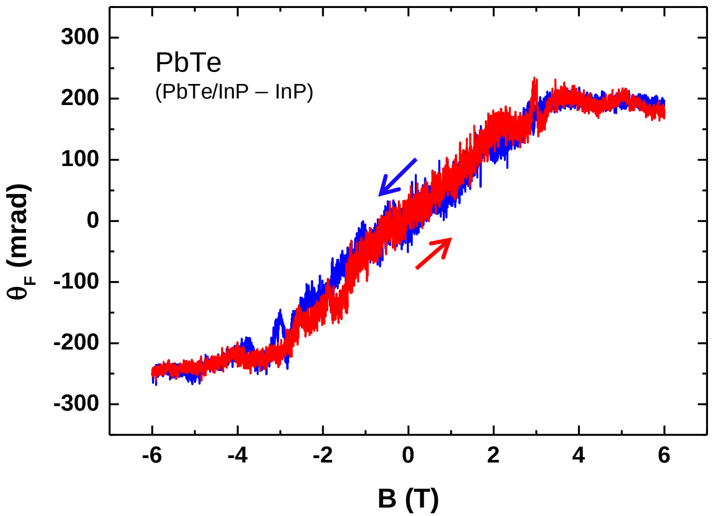




Faraday Rotation

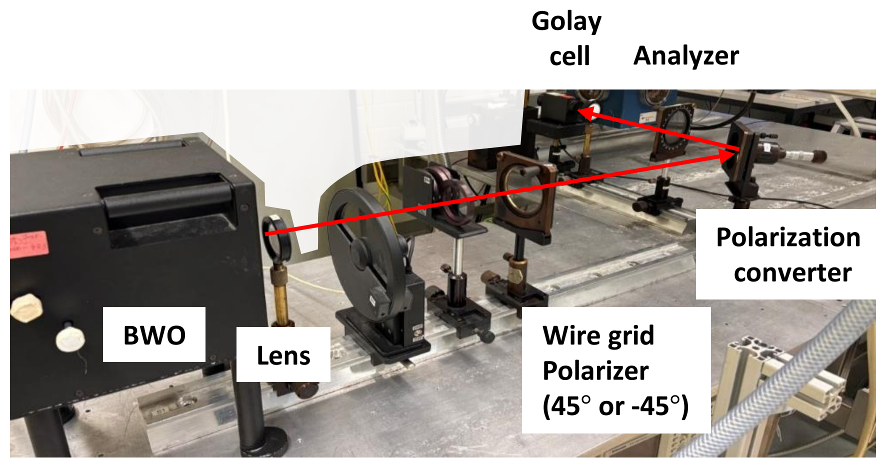
PbTe/InP (ω = 0.316 THz)

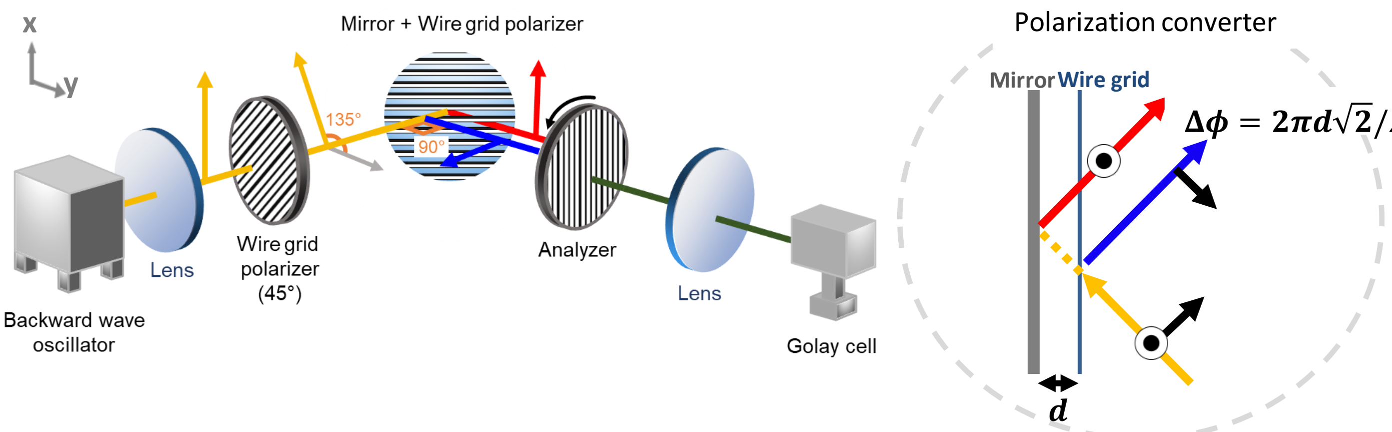


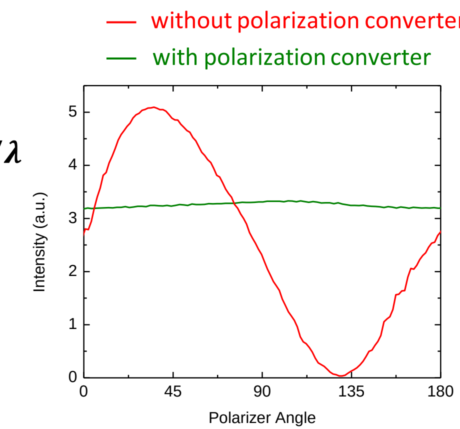


Future Plan

- Change InP substrates with THz-transparent substrates.
- Measure phonon absorption difference using circularly polarized THz radiation
  → Circular dichroism from chiral phonons







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Prof. Martin Dressel

Dr. Artem Pronin

Luca Eisele

Prof. Erik Bakkers

Pim Lueb

Summary

- We measured Faraday rotation in PbTe using THz frequency-domain spectroscopy.
- Strong Faraday rotation was observed in PbTe, but no hysteresis was observed.
- We suggest that the chiral phonons contribute to the magneto-optical response in PbTe.

PbTe/InP (ω = 0.3 THz)