THz emission in topological materials

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TERAHERTZ SPINTRONICS: TOWARD TERAHERTZ SPIN-BASED DEVICES

Workshop October 10th - 12th, 2023
WASEM Monastery, Ingelheim, Germany

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Part 1:

Ferromagnet/2D TMDC interface:

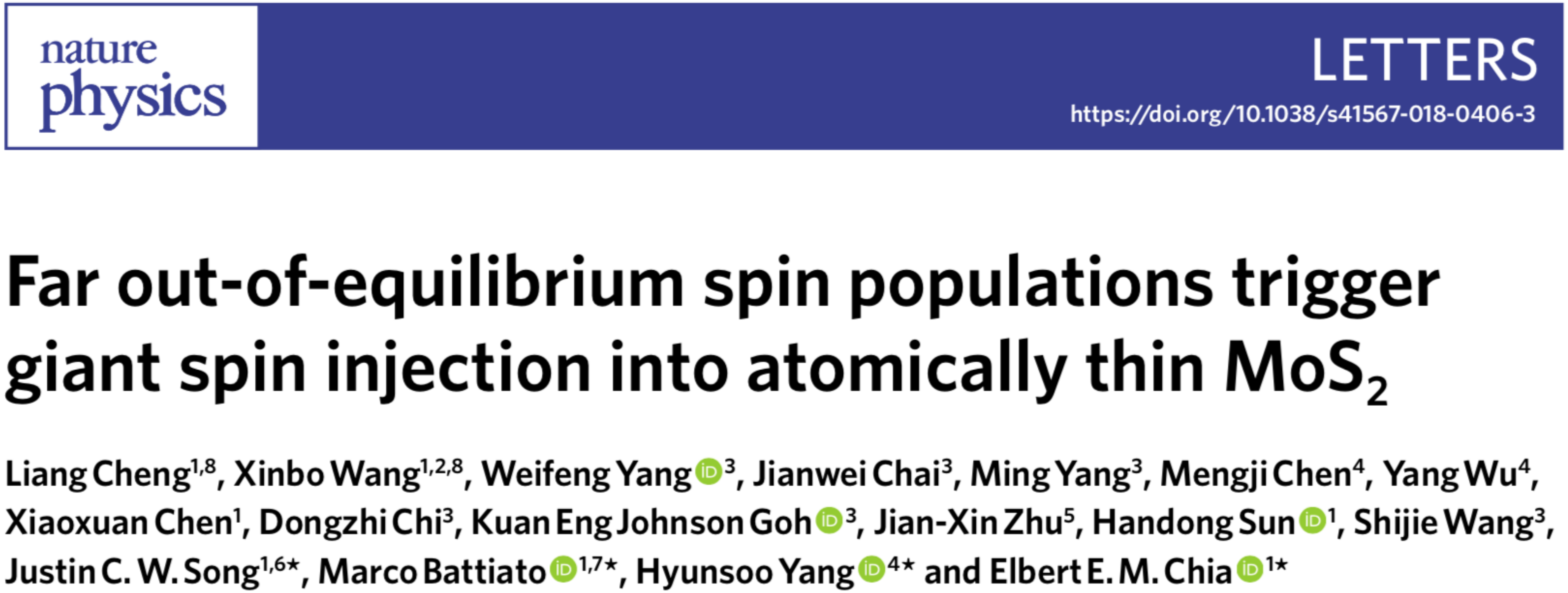
Efficient spin injection into an atomically-thin
semiconductor via
out-of-equilibrium spin populations

Part 2:

Centrosymmetric Dirac semimetal:

Giant photon momentum locked THz emission

Part 1: Ferromagnet/Semiconductor (Co/1L-MoS_2)



nature physics

LETTERS

https://doi.org/10.1038/s41567-018-0406-3

Far out-of-equilibrium spin populations trigger giant spin injection into atomically thin MoS_2

Liang Cheng[1,8], Xinbo Wang[1,2,8], Weifeng Yang[3], Jianwei Chai[3], Ming Yang[3], Mengji Chen[4], Yang Wu[4], Xiaoxuan Chen[1], Dongzhi Chi[3], Kuan Eng Johnson Goh[3], Jian-Xin Zhu[5], Handong Sun[1], Shijie Wang[3], Justin C. W. Song[1,6]*, Marco Battiato[1,7]*, Hyunsoo Yang[4]* and Elbert E. M. Chia[1]*

Nature Physics, DOI:10.1038/s41567-018-0406-3 (2019)

Spin injection into semiconductors

Challenge in spintronics: Efficient spin injection into a semiconductor

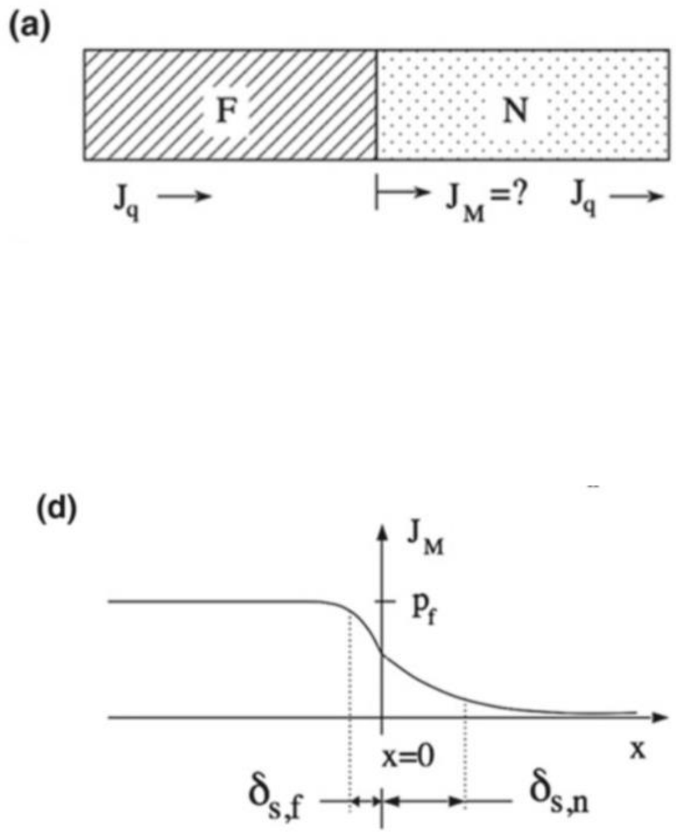
- Traditional method:
  - Get spin-polarized electrons by passing charge current through a ferromagnet/ semiconductor (FM/SC) interface.
  - Neglecting interface resistance,

$$J_M = p_F \frac{\mu_B}{e} J_q \frac{1}{1+(r_N/r_F)\,(1-p_F^2)}, \quad p_F \sim 0.4 \text{ for FM.}$$

- Difficulty:
  - Conductance mismatch:

    $\frac{r_N}{r_F} \gg 1$ for FM(r_F)/SC(r_N) $\Rightarrow$ small J_M.

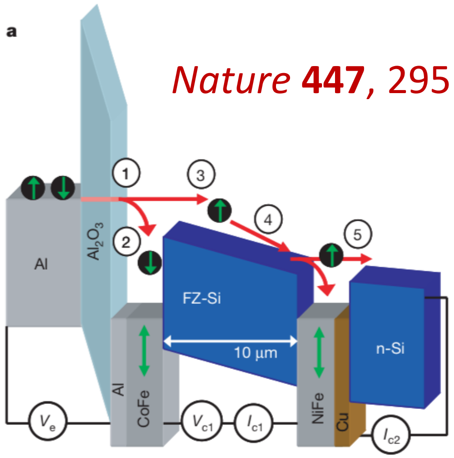
    Predicted efficiency of spin injection $\sim 1\%$ for FM/SC .
  - Efficiency can be optimized to $20-50\%$ by adjusting interface (contact) resistance → complicated heterostructure fabrication



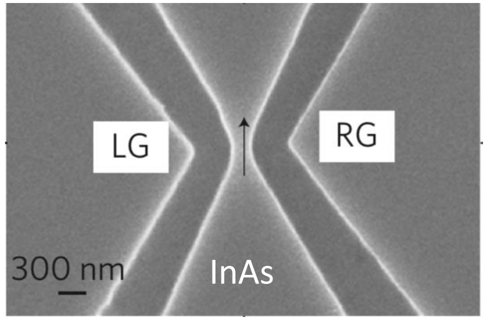
M. Dyakonov, *Spin physics in Semiconductors*, 2nd edition, 2017.

Novel spin injection methods

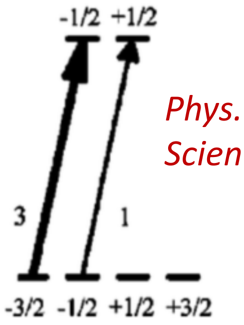
- Tunneling effect
  - Hot electrons emitted from tunnel junction become spin-polarized by going through a FM layer and then injected into SC. Emission current ($\sim 1\ \mathrm{mA}$), spin current ($\sim 100\ \mathrm{nA}$) → ~0.1% efficiency. Does not need large spin-orbit coupling (SOC).
- Quantum point contact (in spin-FET)
  - A short quantum wire made of SC with strong SOC. The spin injection efficiency is $\sim 100\%$. Low T only.
- Optical spin injection
  - Spin-polarized electron excited by circular light pump in SC with strong SOC. The spin injection efficiency is up to $\sim 50\%$.



Nature **447**, 295 (2007)



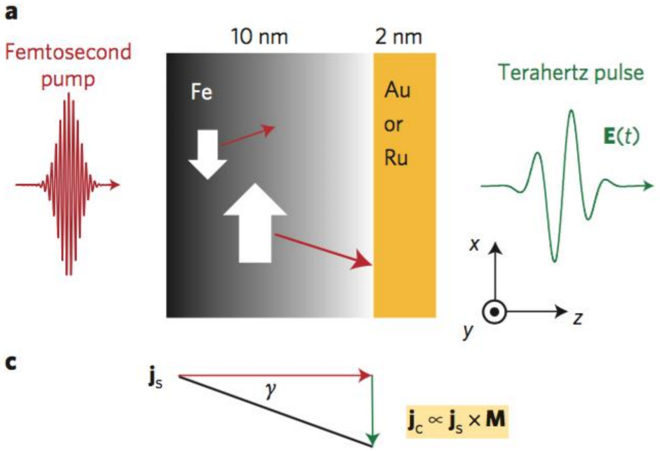
Nature Nanotech. **4**, 759 (2009)
Nature Nanotech. **10**, 35 (2014)



Phys. Rev. B **71**, 035209 (2005)
Science **330**, 1801 (2010)

Motivation: Ultrafast spin injection in ferromagnet/heavy-metal heterostructure

- Spin current generation:
  - Diffusion of spin polarized electrons to heavy-metal layer generated by ultrafast laser excitation in FM layer.
- Spin current detection in non-magnetic heavy metal with SOC:
  - Ultrafast spin current ⇒ transient charge current (ISHE) ⇒ THz emission (detected by THz spectrometer).



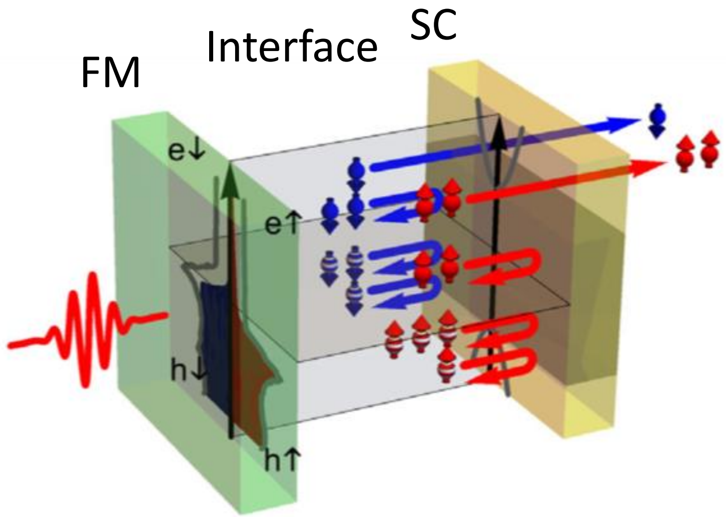
T. Kampfrath *et al.*, *Nat. Nanotech.* **8**, 256 (2013)

Question:

Can we use this method to inject spins into a **semiconductor**?

Ultrafast spin injection in FM/SC

- Bandgap block:
  - Only high-energy electrons can be injected to SC (details in next slide)
- Advantages:
  - High spin-injection efficiency
  - Large spin current
  - SOC is unnecessary
  - Simple device fabrication
  - Temperature-insensitive

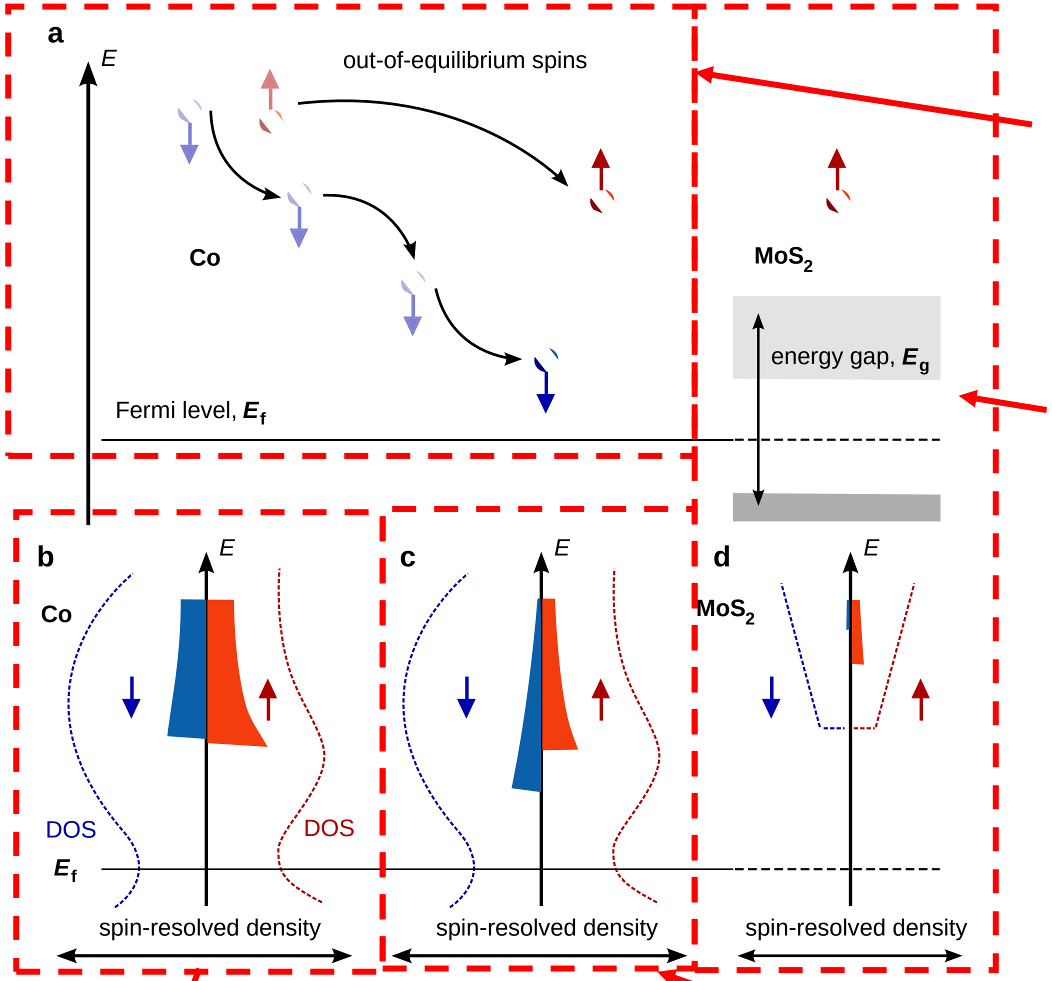


Theoretical prediction: Spin injection from FM to semiconductor (Si). Low-energy electrons are blocked by the SC bandgap.

M. Battiato et al., *Phys. Rev. Lett.* **116**, 196601 (2016)

Marco Battiato (NTU)

Using high-energy electrons to achieve a fully spin-polarized population



As the out-of-equilibrium excited carriers diffuse, they relax in energy.

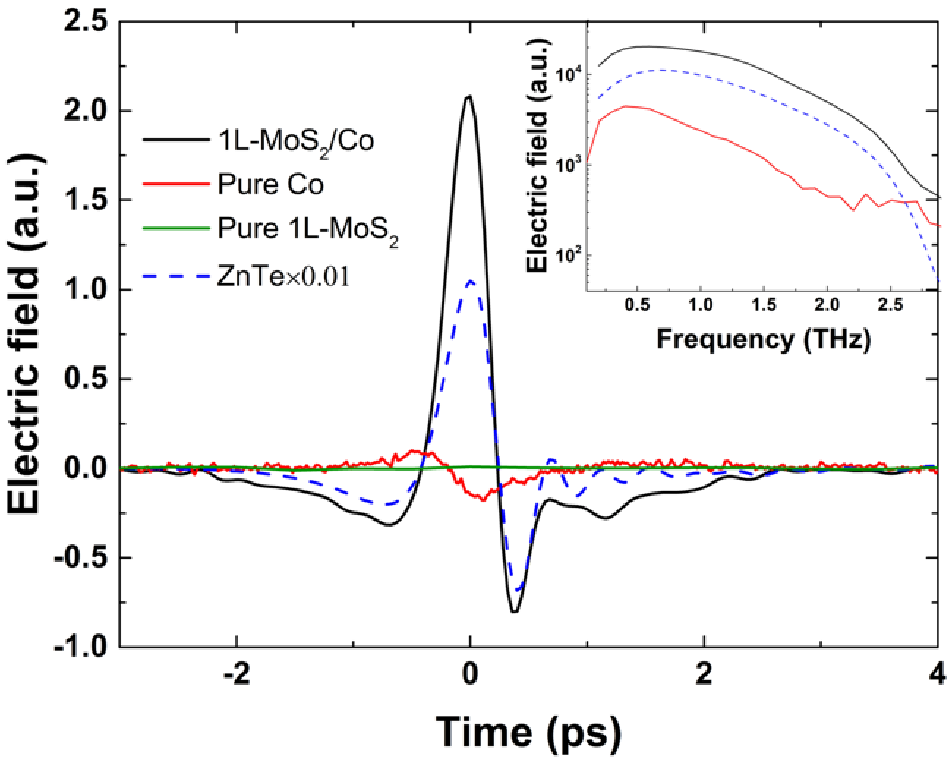
Only those with energy higher than the SC conduction band minimum can cross into the SC layer
→ SC bandgap filters only the high-energy carriers where the population is almost fully spin polarized

→ extremely-high spin polarization of the current injected into the SC

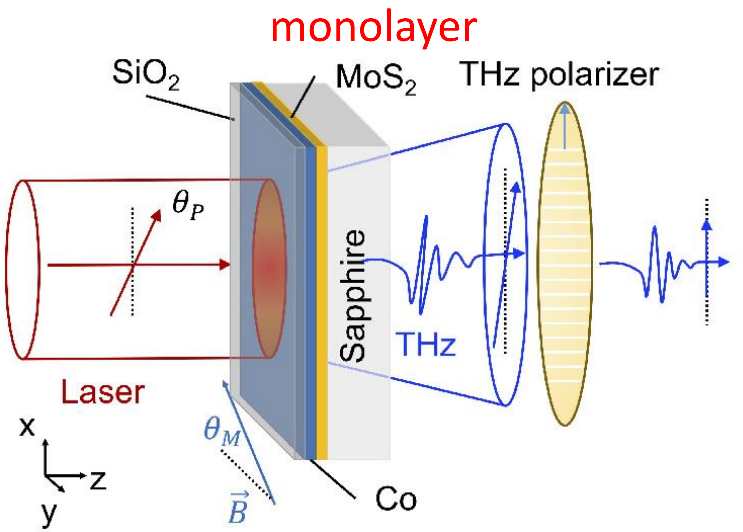
Strong spin asymmetry of the e-e scattering lifetimes:
→ minority spin electrons quickly thermalize
→ most majority spin electrons persist for a longer time at high energies

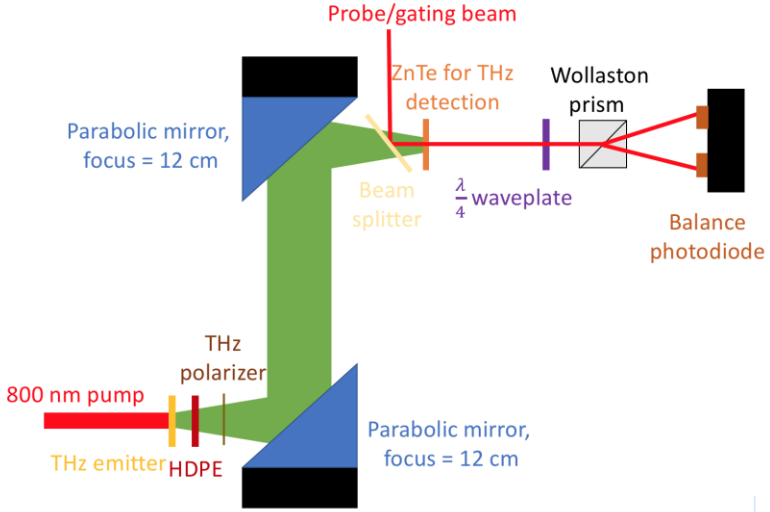
Strongly out-of-equilibrium carrier distribution generated in the FM layer by a fs laser excitation

Experimental setup



$Co/MoS_2 \gg Co \gg MoS_2 \approx 0$





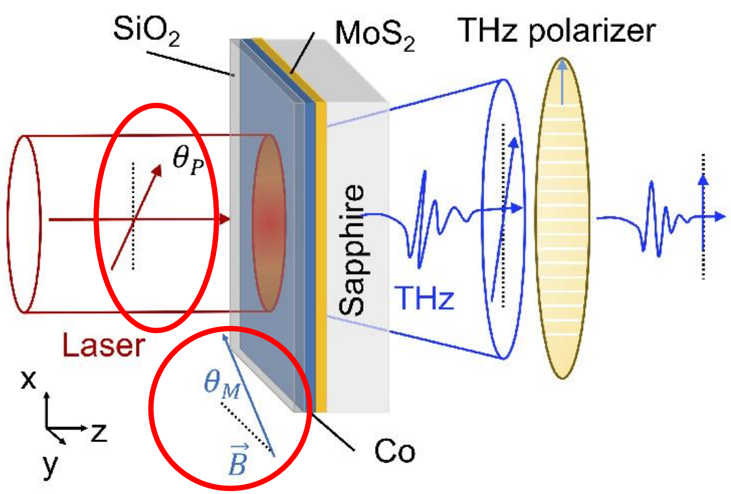
electro-optic detection

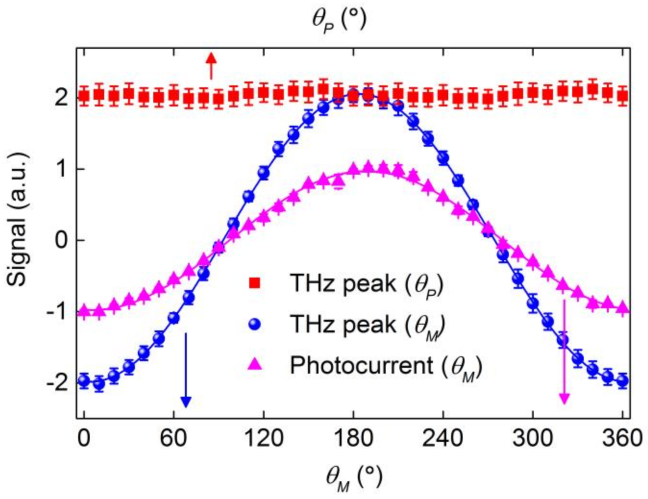
THz emission from Co/MoS_2

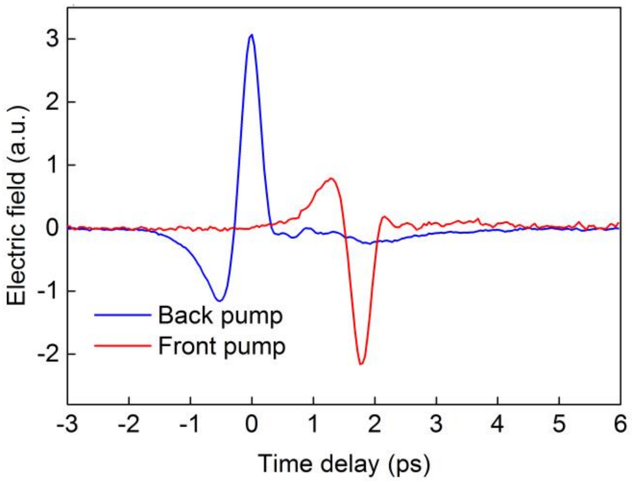
$\vec{J}_C \propto \vec{J}_S \times \vec{M}$ ⇒ spin-to-charge conversion (SCC) origin of THz emission

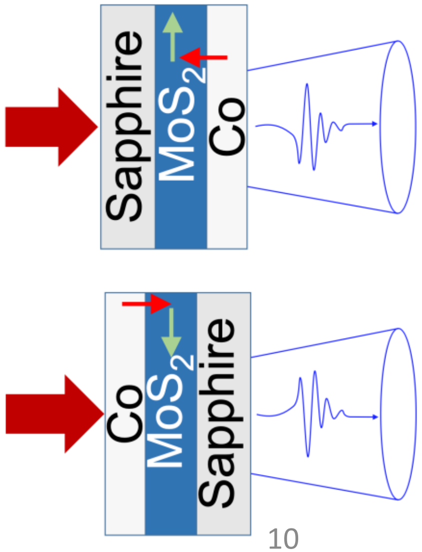
Dependence of THz emission:

- No dependence on θ_P
- 360° symmetry on θ_M
- Polarity of THz emission flips when pump direction is flipped $\Longrightarrow$ THz signal is not from demagnetization in Co layer





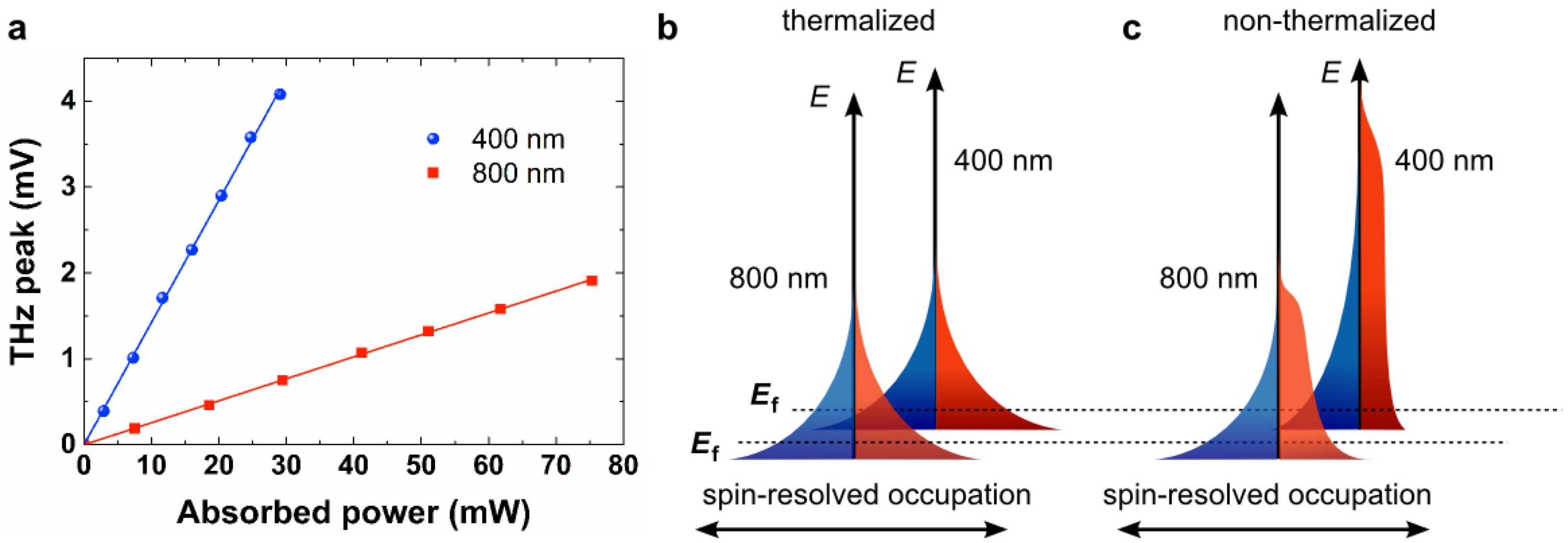




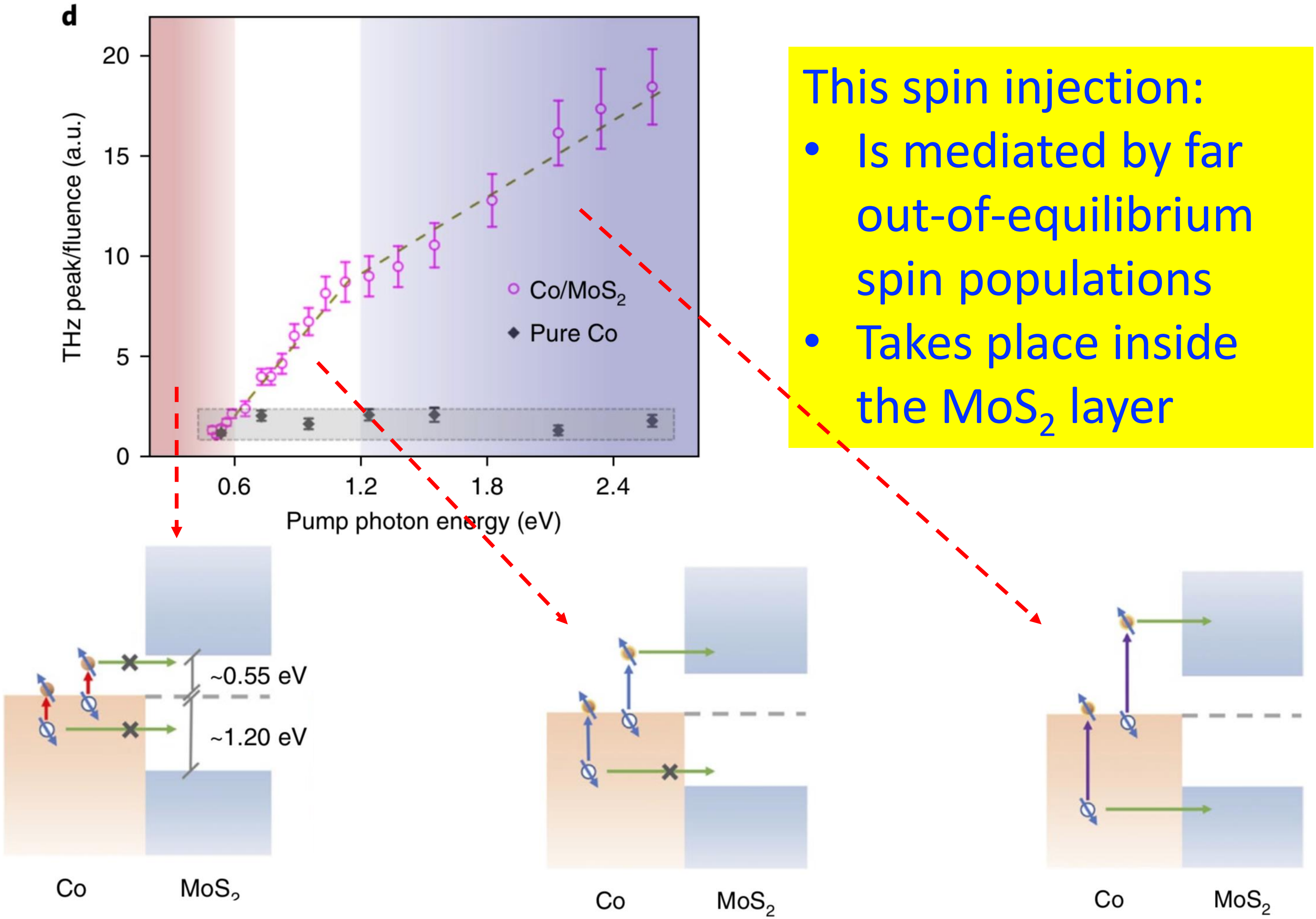
Pump power & wavelength dependence

- Linear dependence on pump power (consistent with previous ISHE-based THz emission in FM/NM heterostructures) → has not reached saturation regime
- At same absorbed pump power, THz emission amplitude with 400-nm pump is larger (5X more) than with 800-nm pump ⇒ far-from-equilibrium effect

Higher energy of injected electrons
⇒ higher injection efficiency



Pump-photon energy dependence



This spin injection:

- Is mediated by far out-of-equilibrium spin populations
- Takes place inside the MoS$_2$ layer

How large is this spin injection?

From:

- Peak THz E-field strength from 1-mm ZnTe emitter
- Emitted THz E-field peak from Co/MoS_2 is 2% that of 1-mm ZnTe
- SCC efficiency $\theta_{eff} = 0.14$ [Nano Lett. **16,** 7514 (2016)]

- Get $\boxed{J_S = 4 \times 10^6 \mathrm{A/cm^2}}$

Gigantic spin injection

- Spin current densities obtained from other techniques & sample systems:

  Spin pumping into metals: $\boxed{J_S \sim 10^2 - 10^3 \mathrm{A/cm^2}}$

  [Co_2FeAl/Mo: PRB **97,** 064420 (2018); CoFeB/Bi_2Se_3 Nano Lett. **15,** 7126 (2015)]

  Spin pumping into semiconductors: $\boxed{J_S \sim 10^2\ \mathrm{A/cm^2}}$

  [$Ni_{81}Fe_{19}$/p-GaAs: Nat. Mater. **10,** 655 (2011)]

  Tunnel junctions (semiconductor): $\boxed{J_S \sim 10^{-1} - 10^0 \mathrm{A/cm^2}}$

  Si/Al_2O_3/Fe junction: Nat. Phys. **3**, 542–546 (2007);

  Si/Al_2O_3/$Ni_{80}Fe_{20}$ junction: Nature **462**, 491–494 (2009)

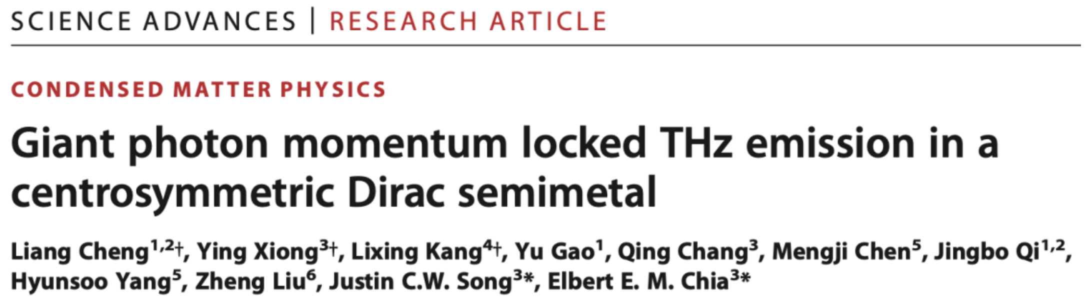
Conclusions of Part 1

Efficient spin injection into a semiconductor is possible

- Overcame impedance mismatch problem
- Mediated by far out-of-equilibrium electrons
- Giant spin current density: $J_S^{peak} = 4 \times 10^6 \mathrm{A/cm^2}$
- Spin-to-charge conversion based THz emission in Co/1L-MoS_2 heterostructure was observed.

Nature Physics **15**, 347-351 (2019)

Part 2:
Giant photon momentum locked THz emission in a centrosymmetric Dirac semimetal


SCIENCE ADVANCES | RESEARCH ARTICLE

CONDENSED MATTER PHYSICS

Giant photon momentum locked THz emission in a centrosymmetric Dirac semimetal

Liang Cheng[1,2]†, Ying Xiong[3]†, Lixing Kang[4]†, Yu Gao[1], Qing Chang[3], Mengji Chen[5], Jingbo Qi[1,2], Hyunsoo Yang[5], Zheng Liu[6], Justin C.W. Song[3]*, Elbert E. M. Chia[3]*

Science Advances **9,** eadd7856 (2023)

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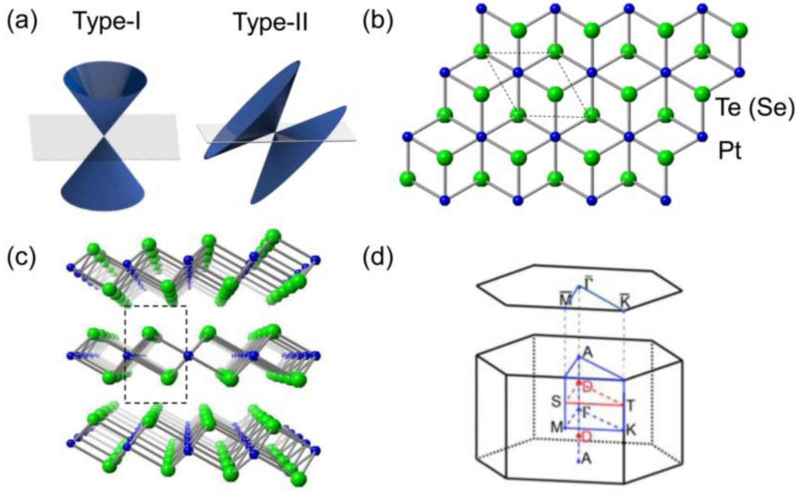
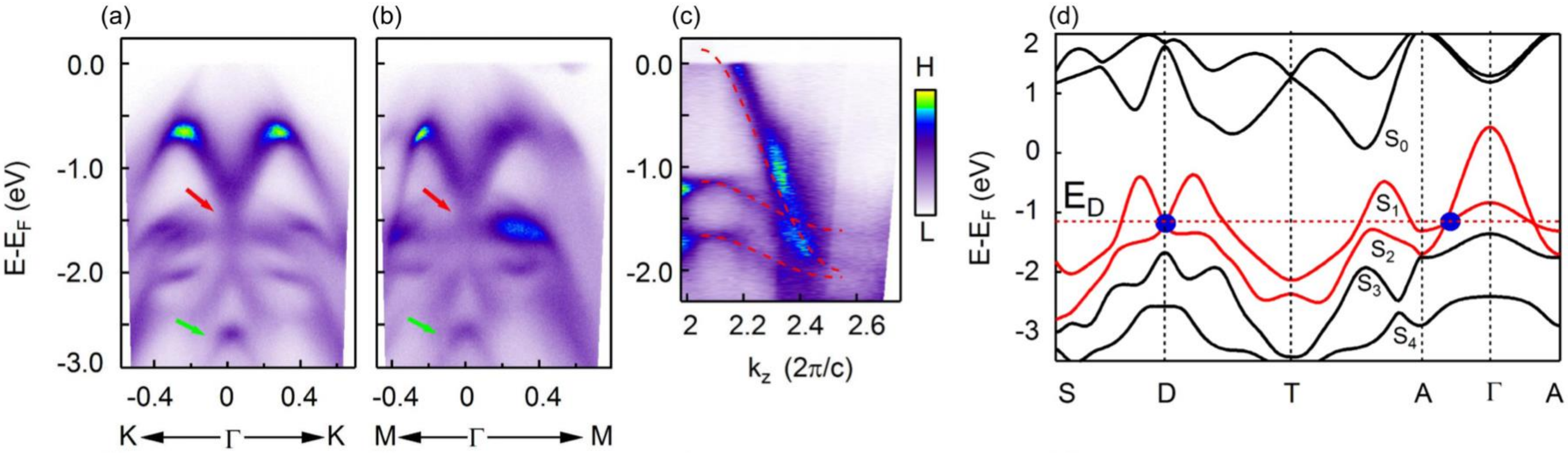
$PtSe_2$: Centrosymmetric, Dirac semimetal



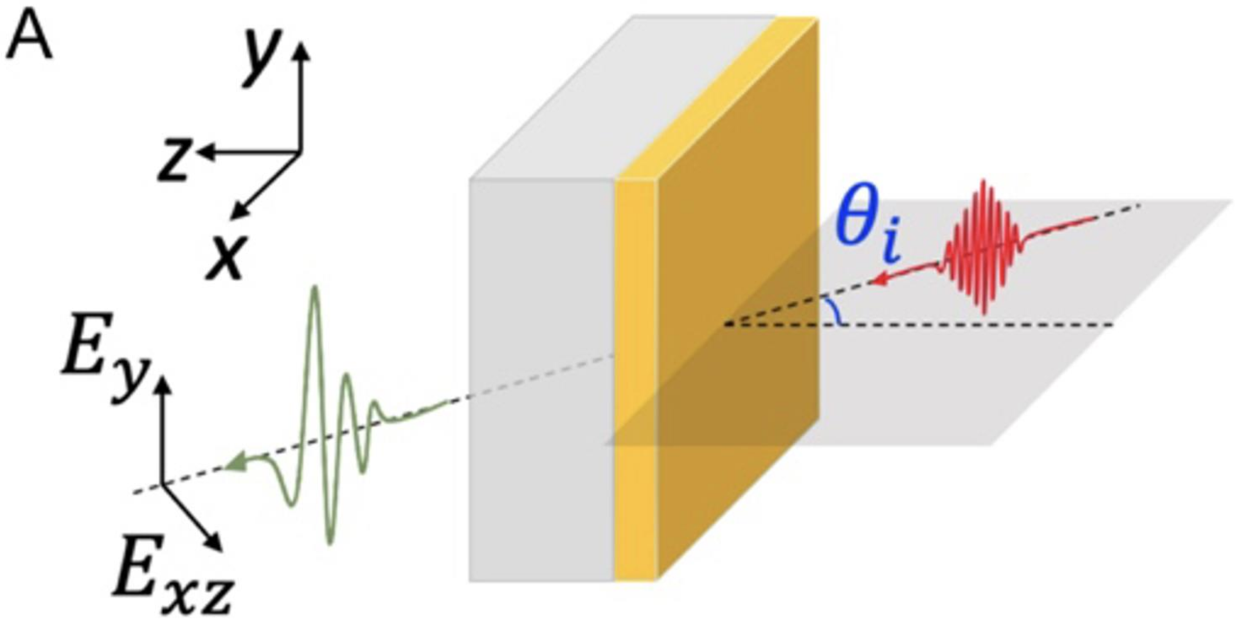
- Type II Dirac semimetal
  - Dirac point is 1.2 eV below Fermi level
- Symmetry:
  - D_{3d} group (i.e. centrosymmetric), no second order nonlinear optical effects (such as photogalvanic effects and SHG) expected.



K. Zhang et al., *Phys. Rev.* B **96,** 125102 (2017)

M. S. Bahramy et al., *Nat. Mater.* 17 21 (2018)

Experimental setup: THz emission



PtSe$_2$ polycrystalline thin film

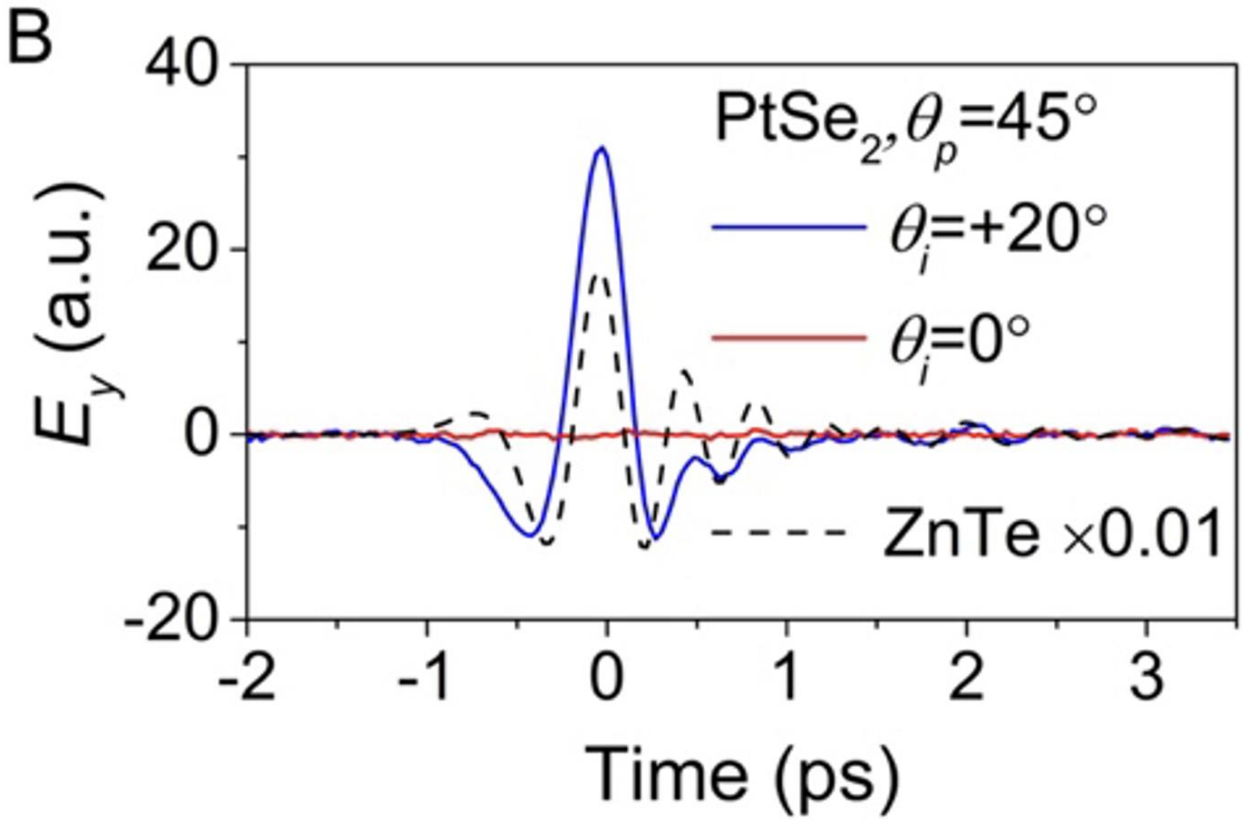
Pump laser: 800-nm wavelength, 80-fs pulse duration, 1-kHz repetition rate, 1-mm dimeter, 2.5-mW power

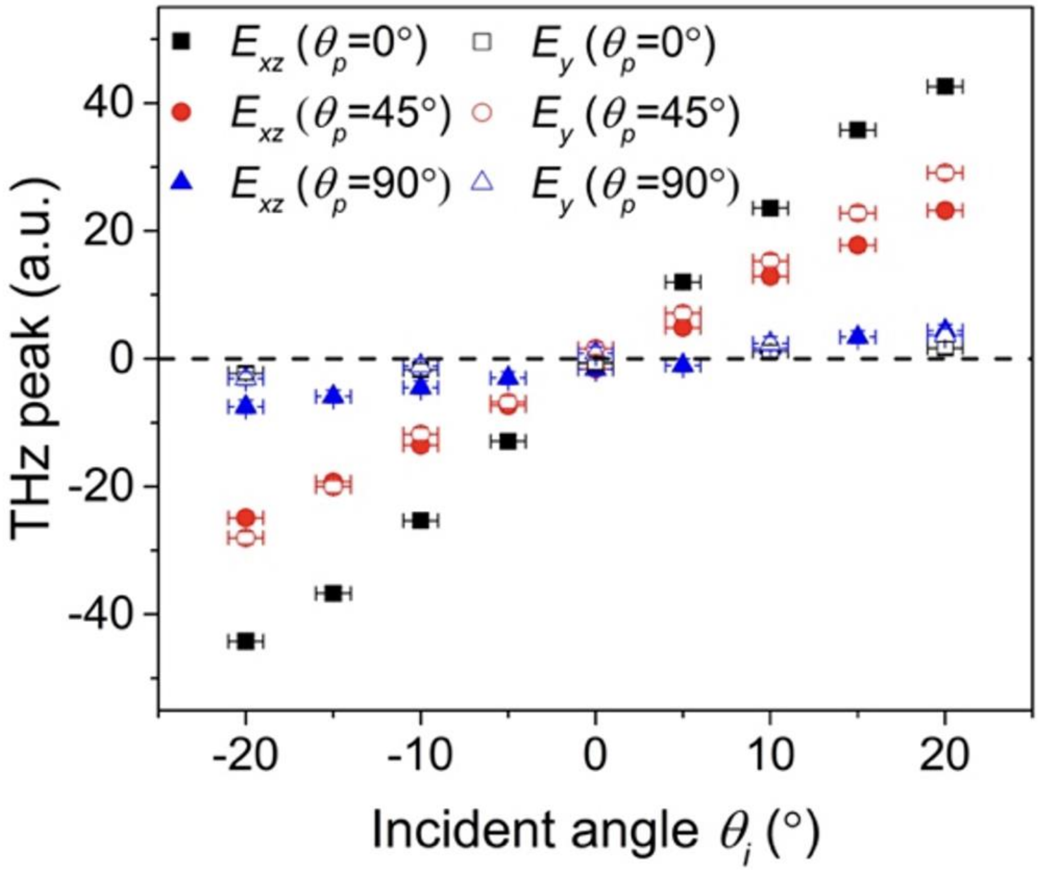
Incident pump beam:
Normal-to-oblique incidence
Linearly-polarized (adjusted by $\lambda/2$-plate) or
Circularly polarized light (adjusted by $\lambda/4$-plate)

THz detection: EO sampling method with 0.5-mm ZnTe crystal. Rotate THz polarizer to measure E_y and E_{xz}.

Room temperature, in air, <5% humidity

Observation 1: THz emission at normal and oblique incidence

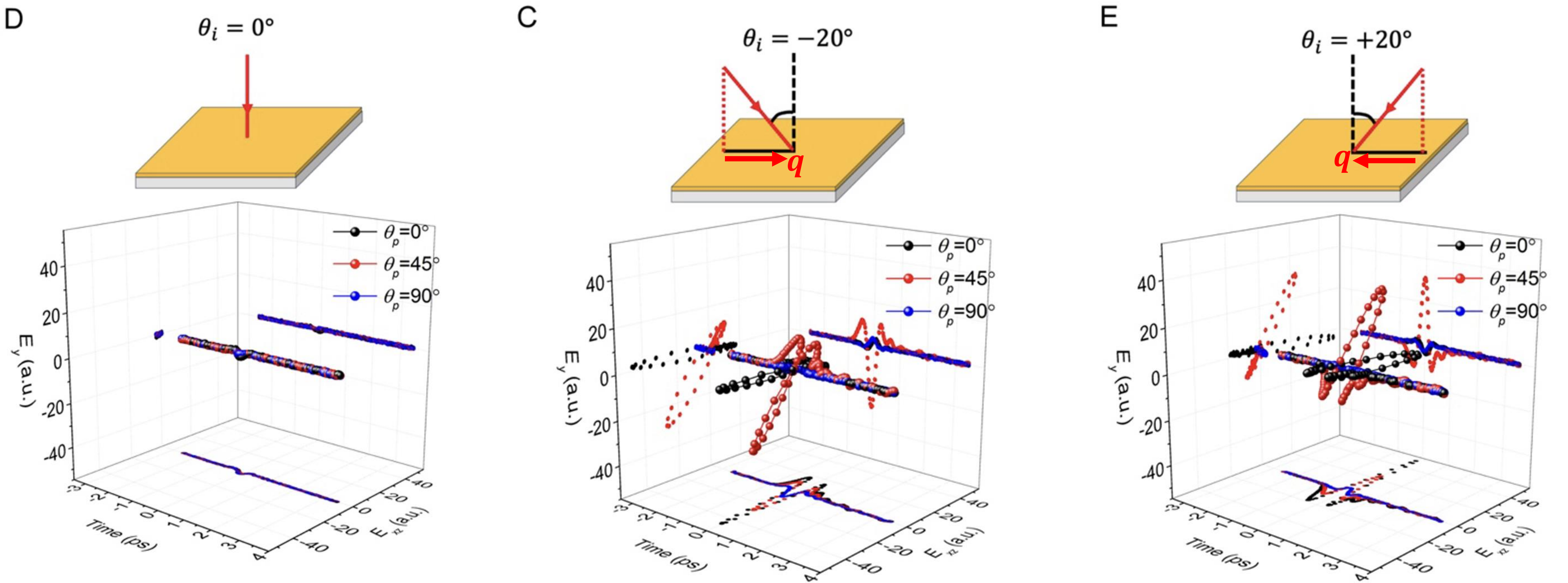




- Zero THz at normal incidence
- THz rapidly turned on at oblique incidence

THz emission enabled by in-plane photon momentum? Photon drag?

Observation 2: THz emission locked to in-plane photon momentum q (at oblique incidence, linearly polarized)



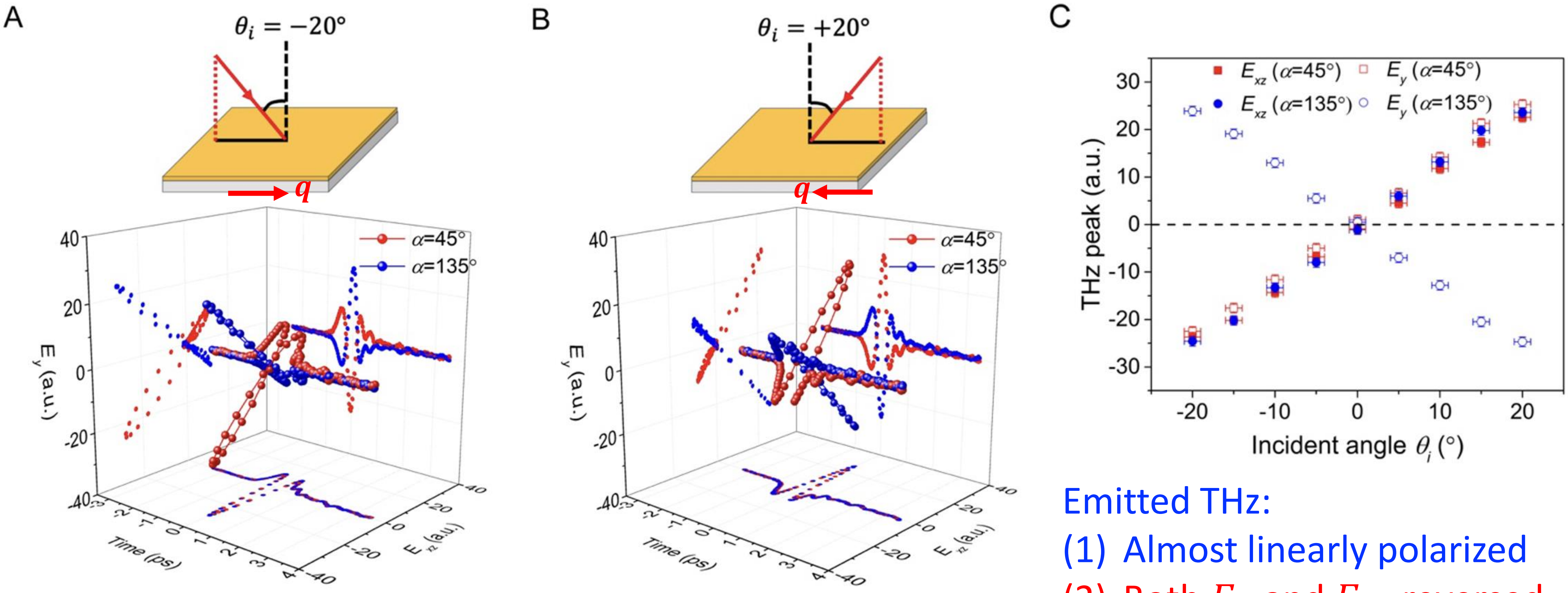
Emitted THz:

(1) Almost linearly polarized (slight ellipticity)

(2) Both E_y and E_{xz} reversed when θ_i is reversed (i.e. when $\vec{q}$ reverses direction)

(3) Relative amounts of E_y and E_{xz} can be tuned by θ_p

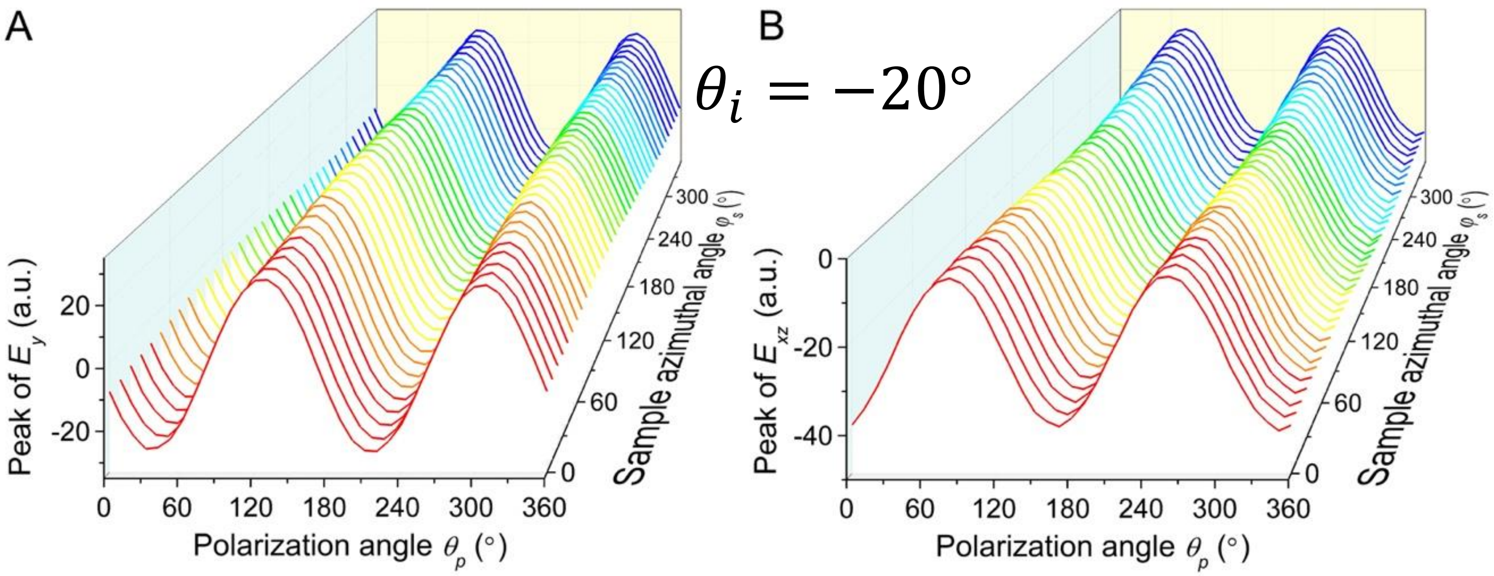
Observation 3: THz emission locked to in-plane photon momentum q (at oblique incidence, circularly polarized)

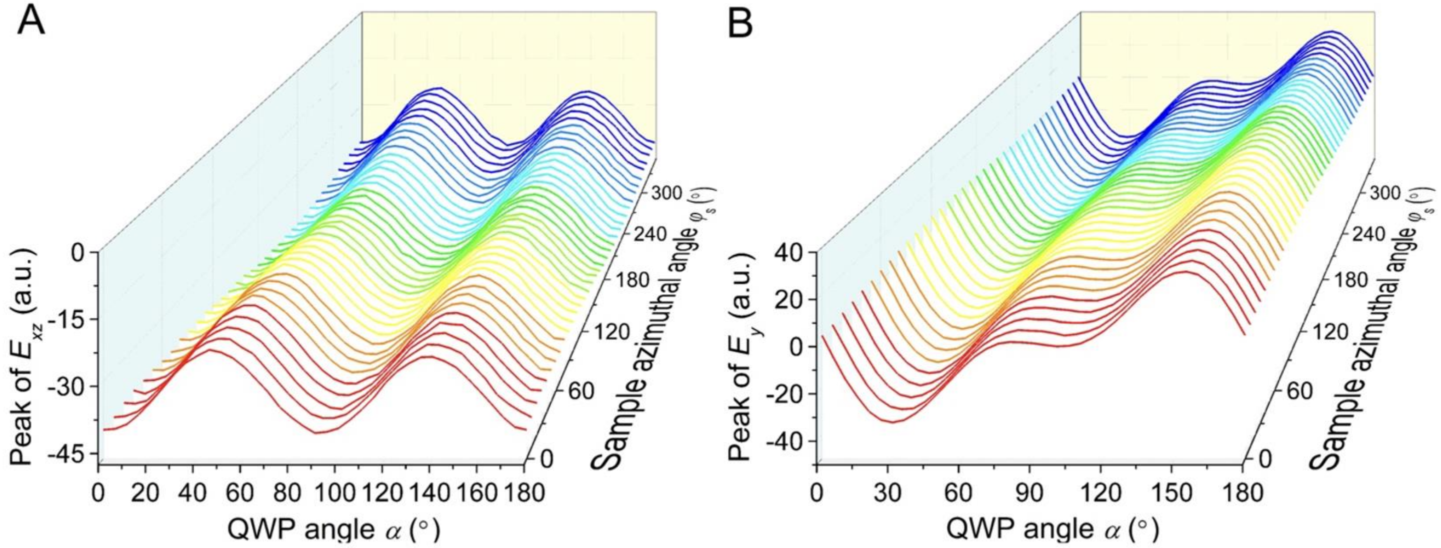


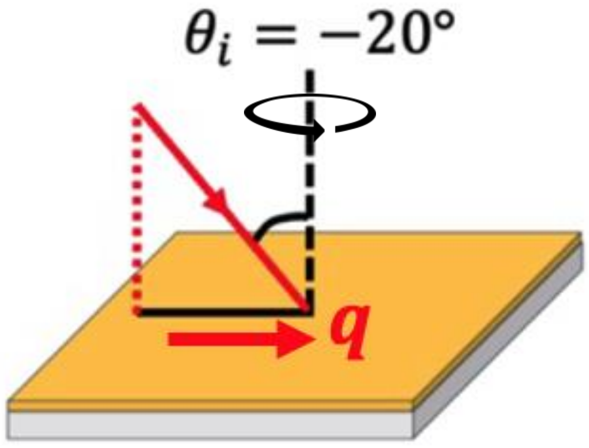
Emitted THz:

1. Almost linearly polarized
2. Both E_y and E_{xz} reversed when θ_i (hence $\vec{q}$) reversed
3. Increases linearly with $|\vec{q}|$

Observation 4: THz emission locked to in-plane photon momentum q (at oblique incidence, vary azimuthal angle)

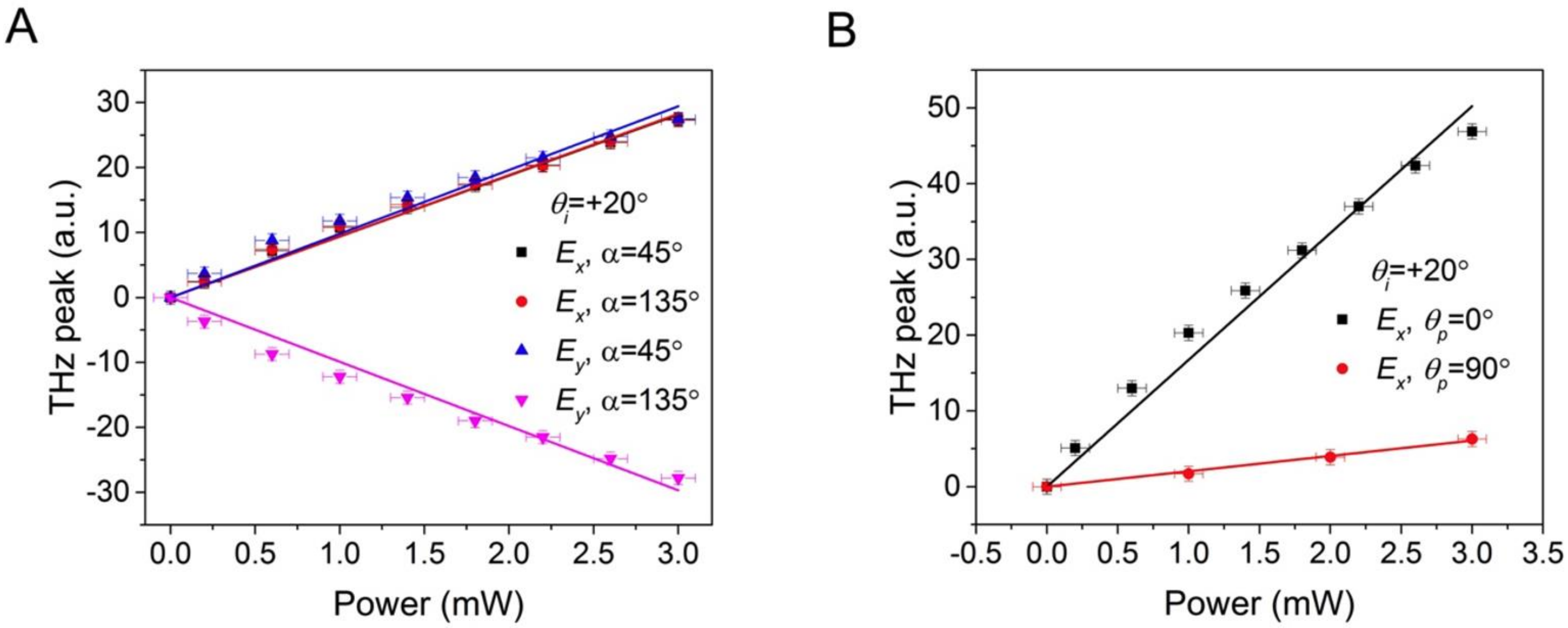






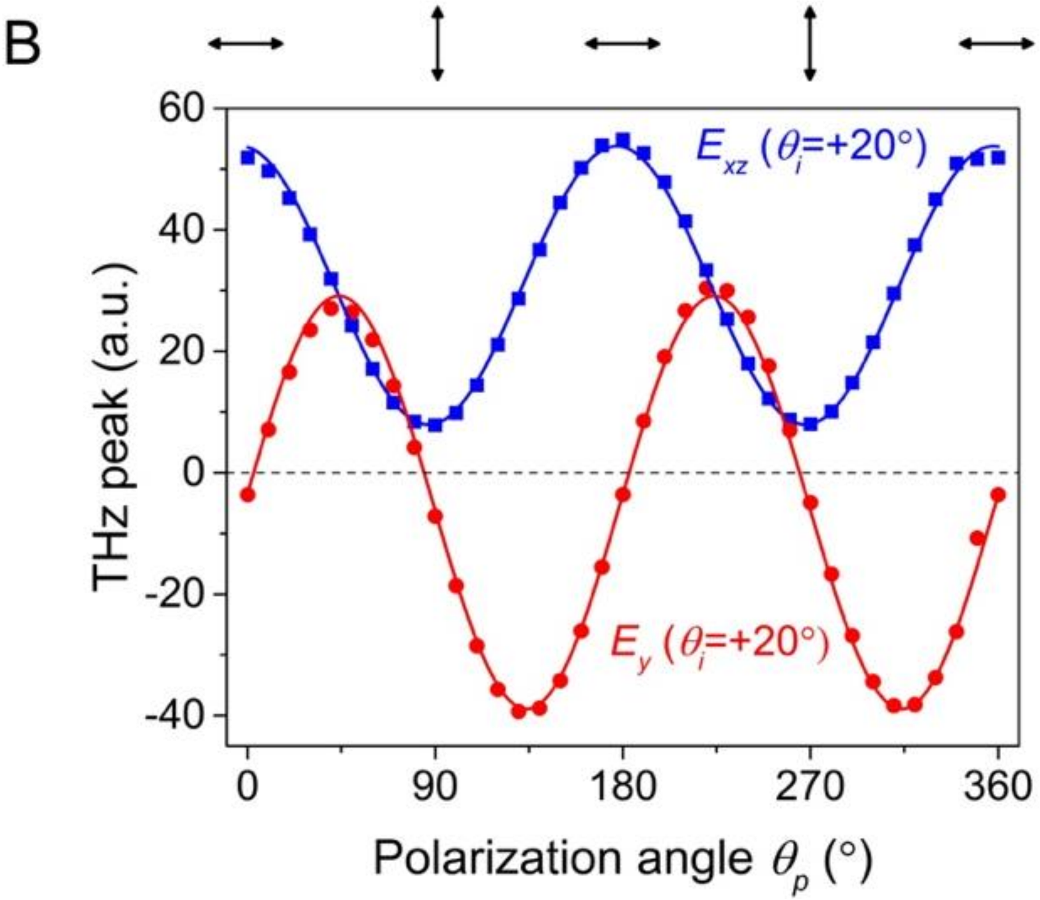
Emitted THz waveform does not change with φ_s
$\Longrightarrow$ PM-locking

Observation 5: THz emission amplitude linear in pump power



THz amplitude linear in laser power (E^*E)
⟹ THz generation arises from 2nd-order nonlinear process

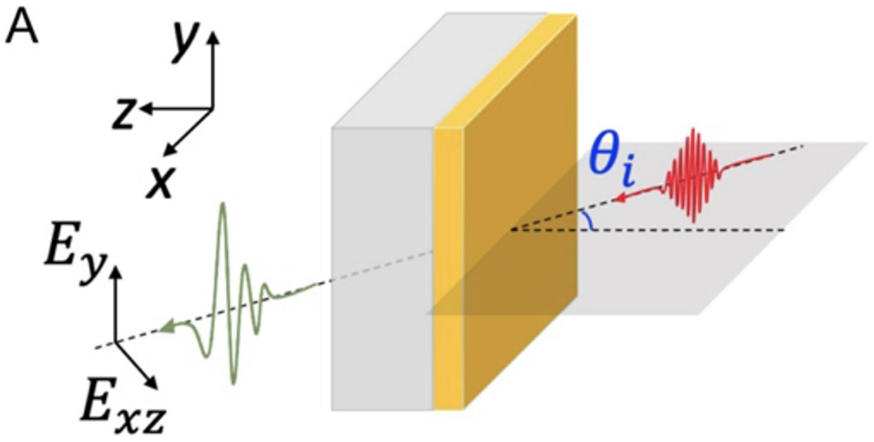
Observation 6: THz emission tuned by (linear) pump polarization



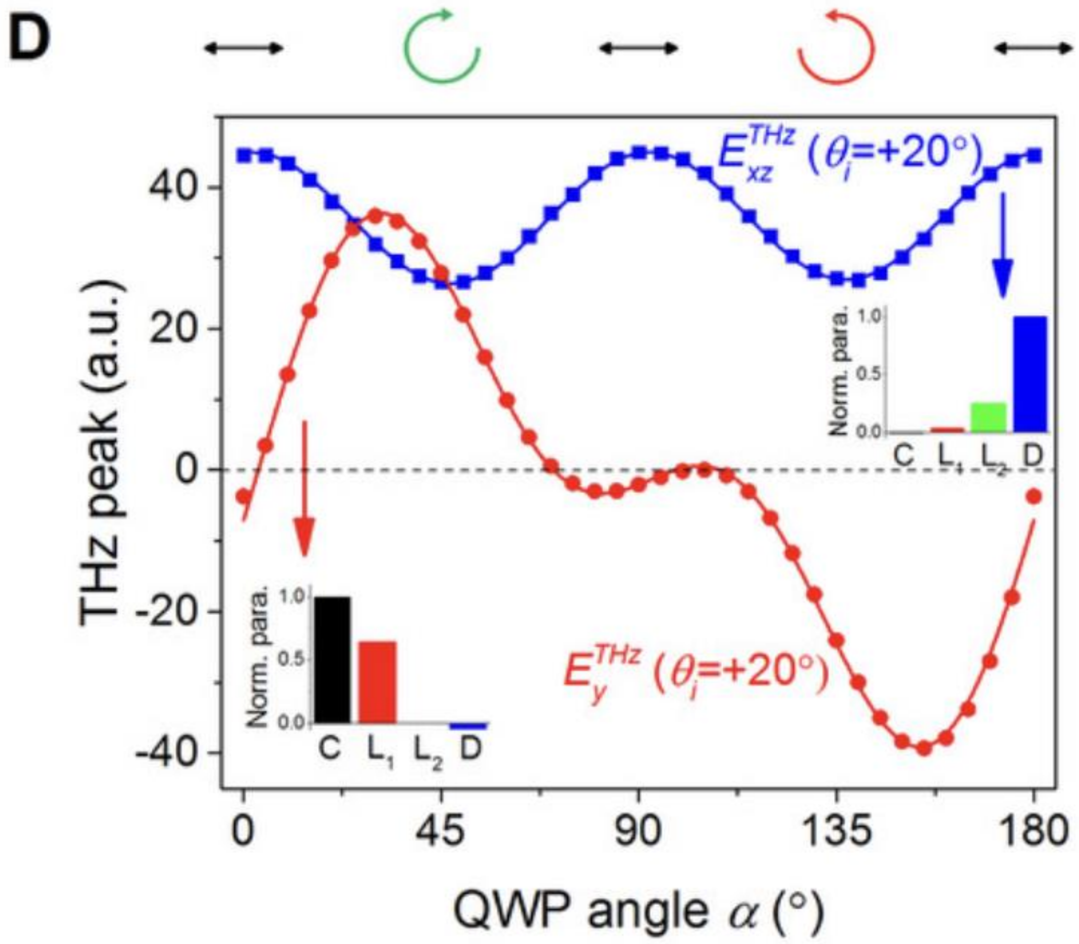
Strong modulation of THz emission on pump polarization θ_p:

- E_{xz} stays positive
- E_y changes sign

$\Rightarrow$ can choose θ_p to maximize/minimize E_{xz} or E_y



Observation 7: THz emission tuned by pump helicity



C = Circular Photon-Drag Effect (CPDE)
L_1 & L_2 = Linear Photon-Drag Effect (LPDE)

Strong modulation of THz emission on pump helicity:

- E_{xz} stays positive
- E_y changes sign

Fit with

$$C \sin 2\alpha + L_1 \sin 4\alpha + L_2 \cos 4\alpha + D$$

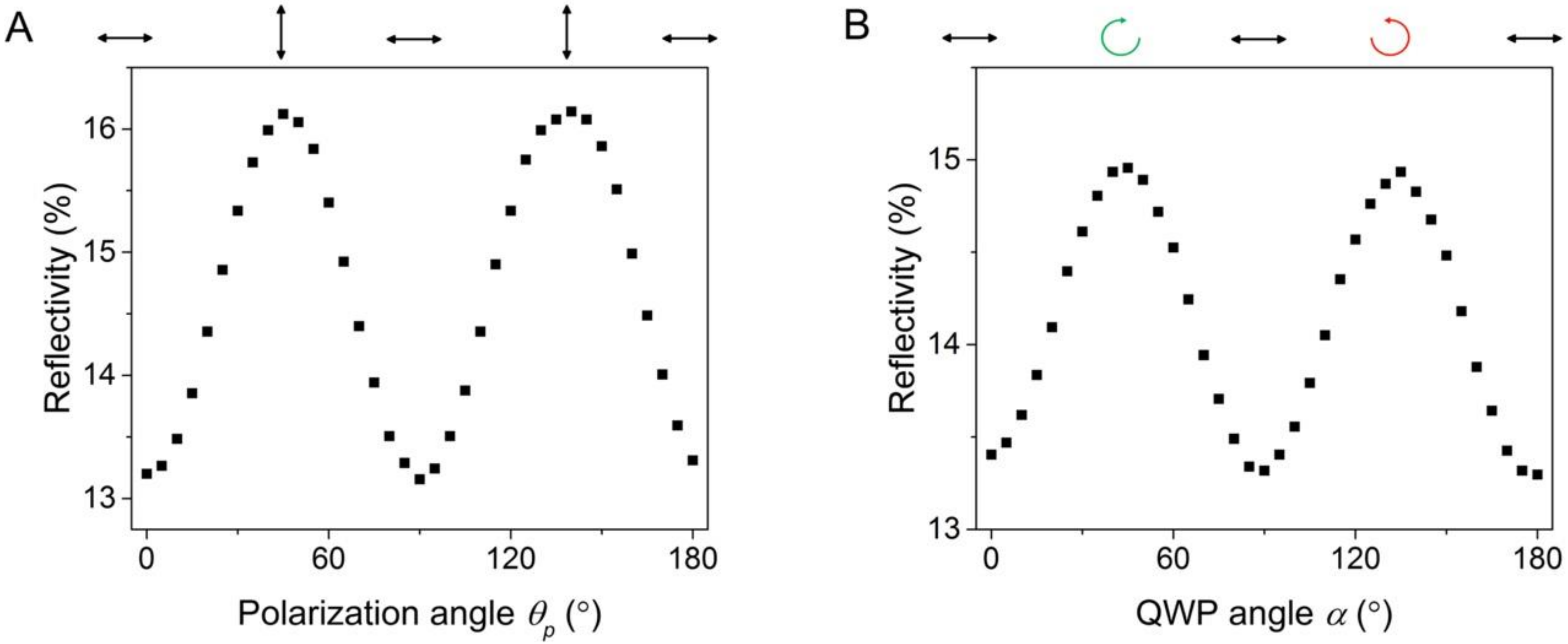
- consistent with symmetry analysis (later)

CPDE affects E_y , but not affect E_{xz}.

Can choose α to maximize/minimize E_y

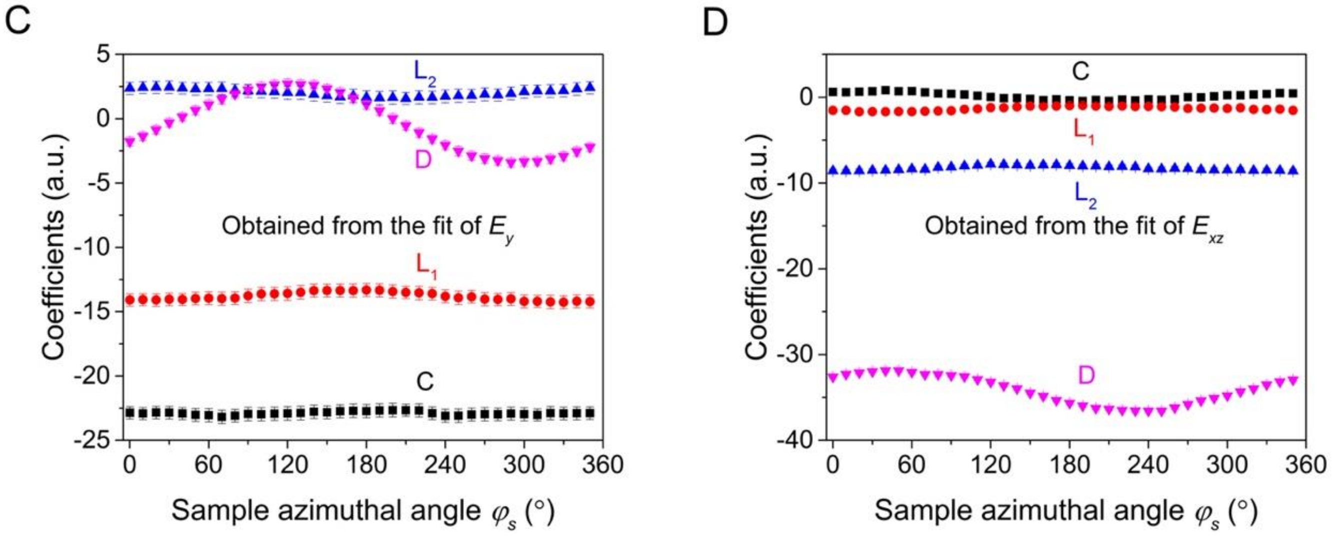
Photo-Dember term may be inside D term in E_{xz}.

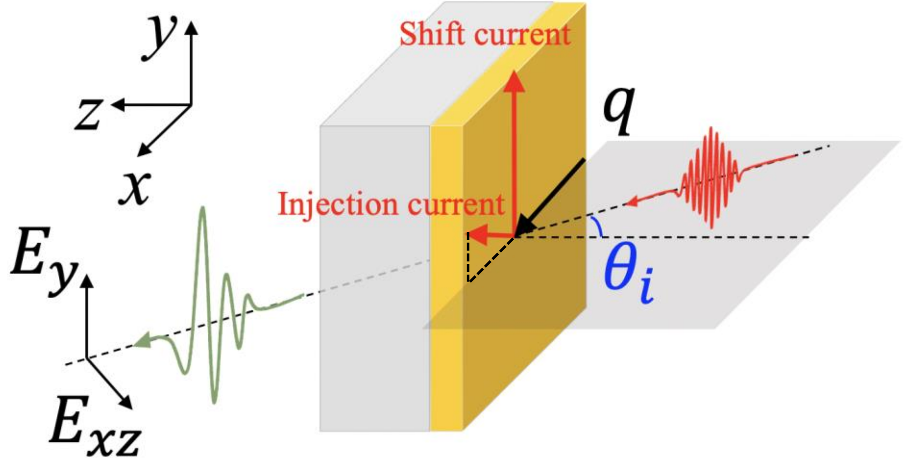
Is the strong THz modulation by polarization & helicity caused by polarization- and helicity-dependent absorption?



Answer: No, absorption changes with θ_p and α are only ~20%.
$\Longrightarrow$ Large THz modulation caused by quantum geometry? Asymmetric sampling of interband transition matrix dipoles in momentum space.

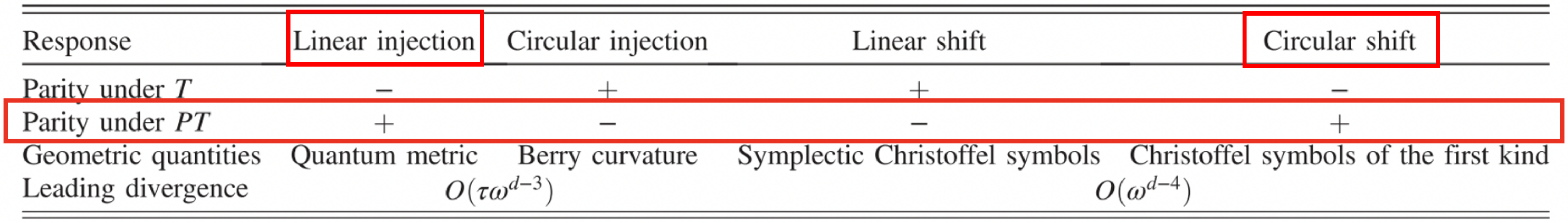
Identification of the photocurrents





(P: inversion symmetry; T: time reversal symmetry)

PRX **10,** 041041 (2020)

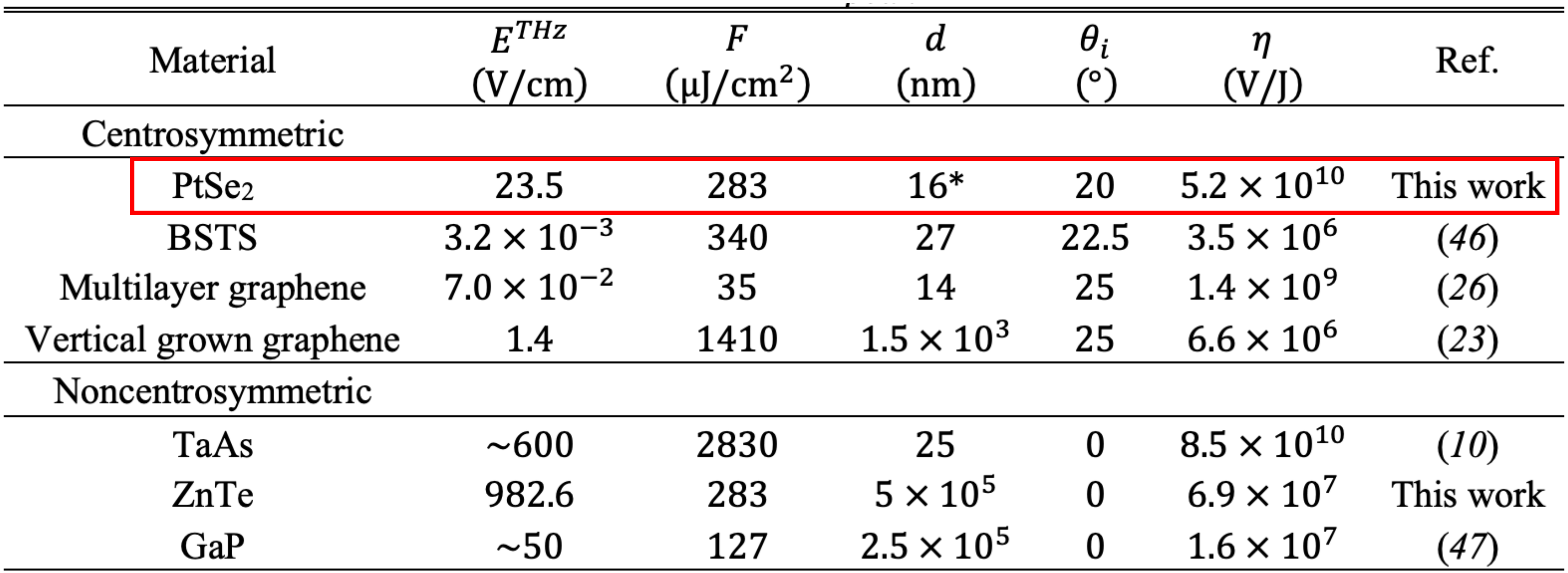


Response	Linear injection	Circular injection	Linear shift	Circular shift
Parity under T	$-$	$+$	$+$	$-$
Parity under PT	$+$	$-$	$-$	$+$
Geometric quantities	Quantum metric	Berry curvature	Symplectic Christoffel symbols	Christoffel symbols of the first kind
Leading divergence	$O(\tau\omega^{d-3})$		$O(\omega^{d-4})$	

Shift current ("C" term): (1) activated by circularly-polarized light, (2) in the y-direction, i.e. perpendicular to in-plane photon momentum q.

Injection current ("L" terms): (1) activated by linearly-polarized light, (2) lies along the x-y plane.

Giant THz emission efficiency in $PtSe_2$



Material	E^{THz} (V/cm)	F ($\mu J/cm^2$)	d (nm)	θ_i (°)	η (V/J)	Ref.
Centrosymmetric						
$PtSe_2$	23.5	283	16*	20	5.2×10^{10}	This work
BSTS	3.2×10^{-3}	340	27	22.5	3.5×10^{6}	(*46*)
Multilayer graphene	7.0×10^{-2}	35	14	25	1.4×10^{9}	(*26*)
Vertical grown graphene	1.4	1410	1.5×10^{3}	25	6.6×10^{6}	(*23*)
Noncentrosymmetric						
TaAs	~600	2830	25	0	8.5×10^{10}	(*10*)
ZnTe	982.6	283	5×10^{5}	0	6.9×10^{7}	This work
GaP	~50	127	2.5×10^{5}	0	1.6×10^{7}	(*47*)

Summary

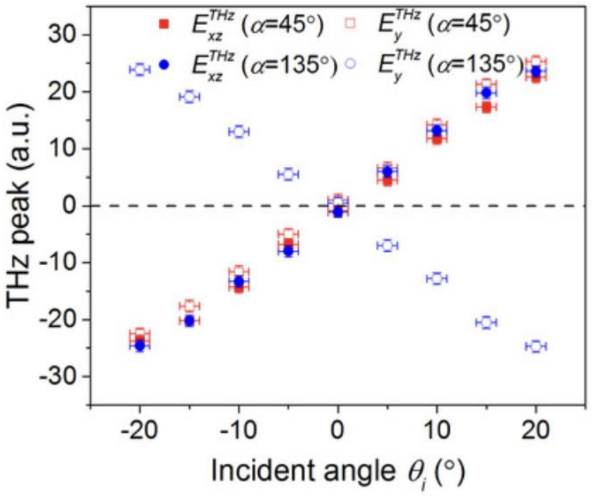
THz emission in centrosymmetric polycrystalline Dirac semimetal $PtSe_2$:

- Zero at normal incidence (expected), but turned on at oblique incidence
- Locked to in-plane photon momentum that breaks the centrosymmetry of the system. Directionality persists even in our polycrystalline samples.
- Strongly modulated by light polarization and helicity
- Large THz efficiency may arise from non-trivial quantum geometry of this material
- Future direction: DFT + finite-q photocurrent calculations to explain large THz efficiency, linking to quantum geometry.

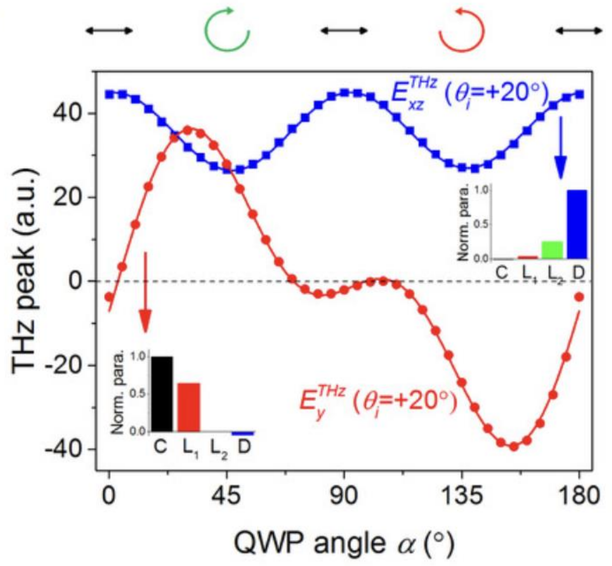
Science Advances **9,** eadd7856 (2023)

Helicity dependence: Expt (top) vs Theory (bottom)

C



D

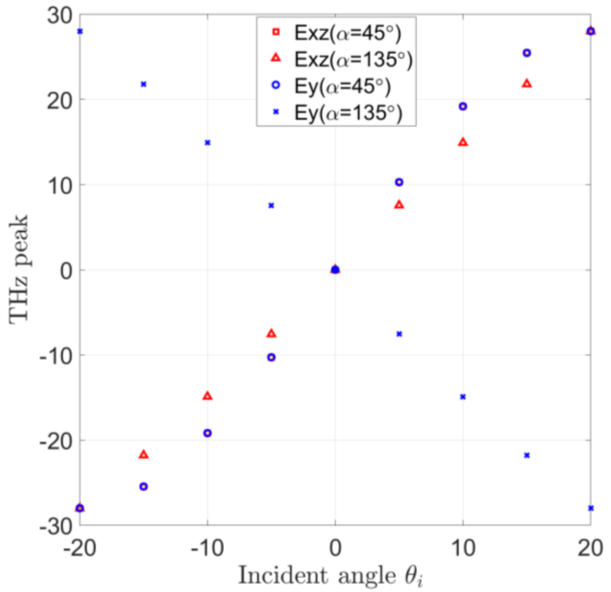


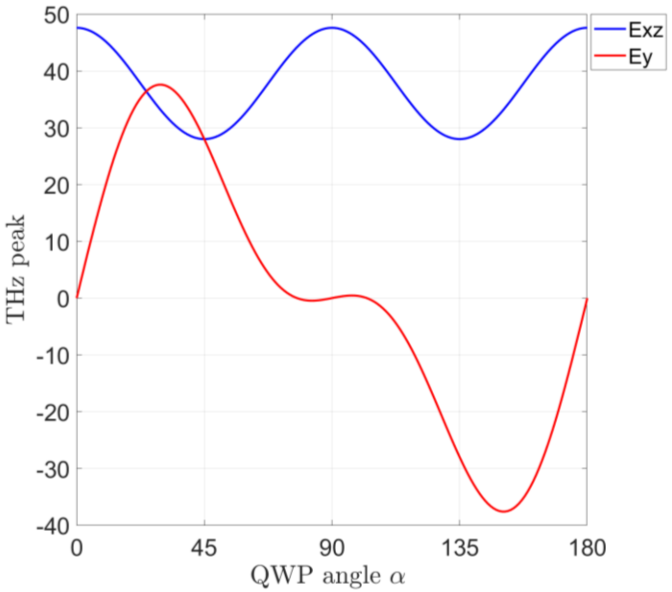
$$E_{xz} \propto$$

$$D_{xz} + 0 sin4\alpha + L_{2xz} cos4\alpha + 0 sin2\alpha$$

$$E_y \propto$$

$$0 + L_{1y} sin4\alpha + 0 cos4\alpha + C_y sin2\alpha$$





- Reproduced same trends as data.
- Allows prediction of parameter set (QWP, HWP, incident angle) for largest THz amplitude, or the largest $E_y - E_{xz}$ anisotropy.
- Surprise: our polycrystal showed single crystal character.

Team Members

THz emission

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Sample growth

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Theory

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Thank You