Surface states mediated spin-to-charge conversion in BiSb-based topological insulators probed by THz emission spectroscopy

H. Jaffrès[1], E. Rongione[1,2,3], S. Massabeau[1], L. Baringthon[1,4], D. She[1], F. Bertran[4], P. Le Fèvre[4], G. Patriarche[5], A. Lemaître[5], R. Lebrun[1,3], N. Reyren[1], S. Dhillon[2], J.-M. George[1]

[1] Unité Mixte de Physique CNRS-Thales *UMφ* (France)

[2] Laboratoire de Physique de l'Ecole Normale Supérieure *LPENS* (France)

[3] Thales Research and Technology *TRT-Fr* (France)

[4] Soleil synchrotron

[5] Centre de Nanosciences et Nanotechnologies *C2N* (France)

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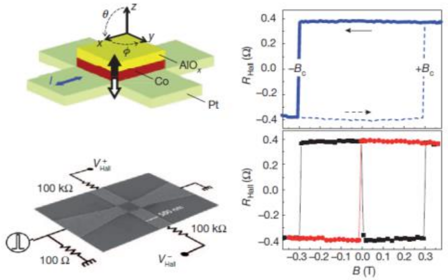
General applications using spin-to-charge conversion

Preamble on spintronics

Pt, Beta-Ta *STT-FMR*

Switching Magnetization

Spin-Transfer et Domain-wall motion

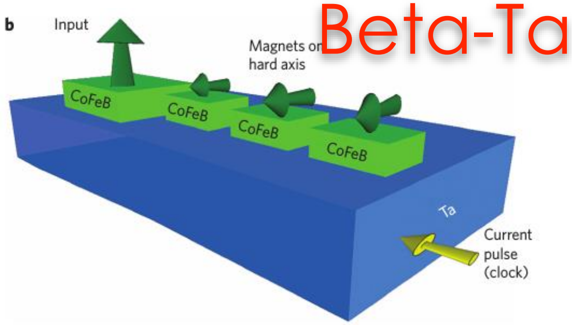


M. Miron *et al.*, Nature 476, 189 (2012),
L. Liu *et al.*, PRL, 109, 096602 (2012),
L. Liu *et al.*, Science 336, 6081 (2012)
M. Cubukcu et al., APL: 104, 042406 (2014)

S. Emori et al., Nat. Mat. 12, 611 (2013)
JC Rojas-sanchez et al, APL (2016)

SHE Spin Torque based Clocking

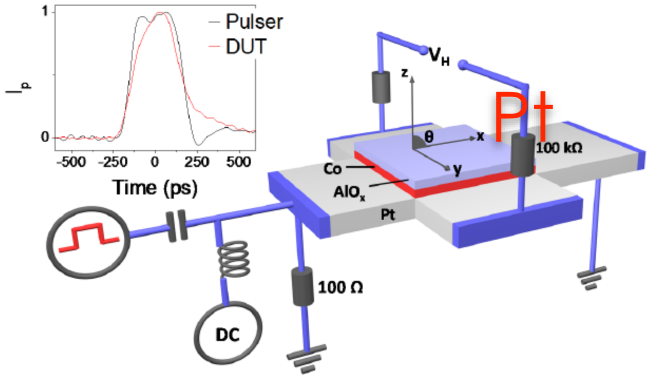
Beta-Ta



D. Bhowmik *et al.*,Nnano, 241 (2013)

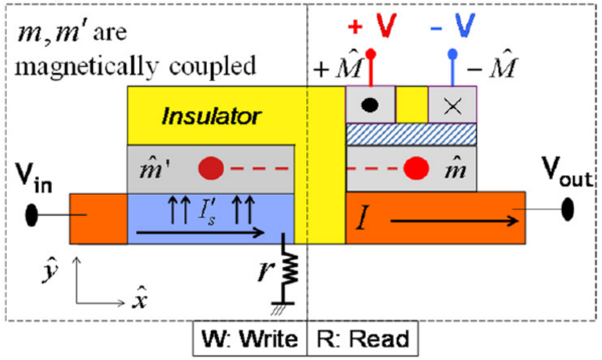
Ultra Fast Switching

Pt



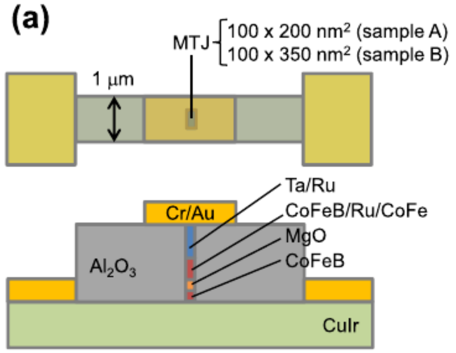
K. Garello *et al.*, arXive: (2014)

Read/Write Heads using GSHE



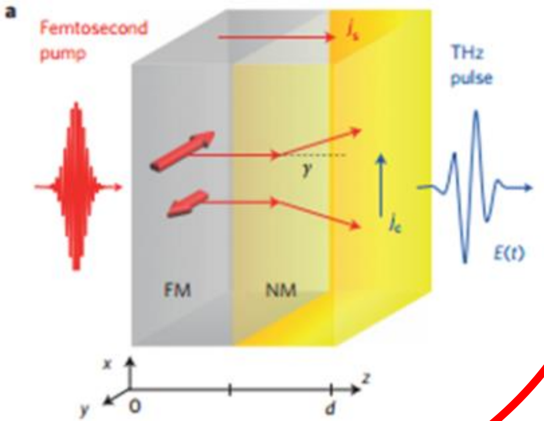
S. Datta *et al.*, APL101, 252411 (2012)

SHE based MTJs



M. Yamanouchi *et al.*, APL102, 212408 (2013)

THz emission by fs laser excitation



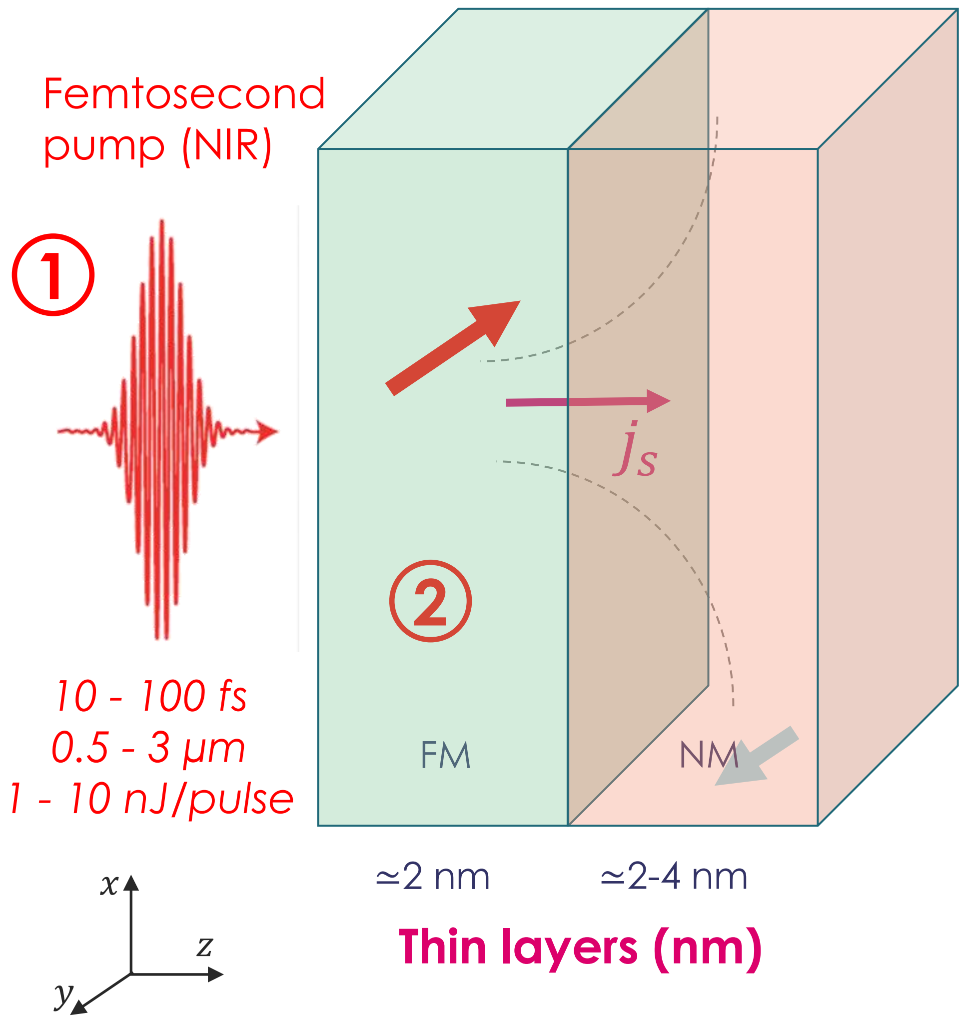
M. Sharad et al., arXiv: 1401.0015 (2013)

TDS : qualifying spin-currents
Complementary FMR spin pumping - ISHE

Spin Hall effect generates THz

Spintronic THz emitters : fundamentals of THz emission process

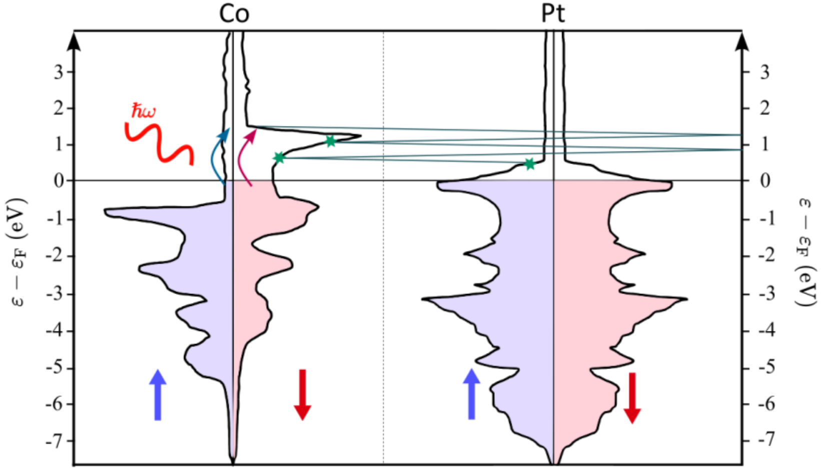
ferromagnet / non-magnetic material



Thin layers (nm)

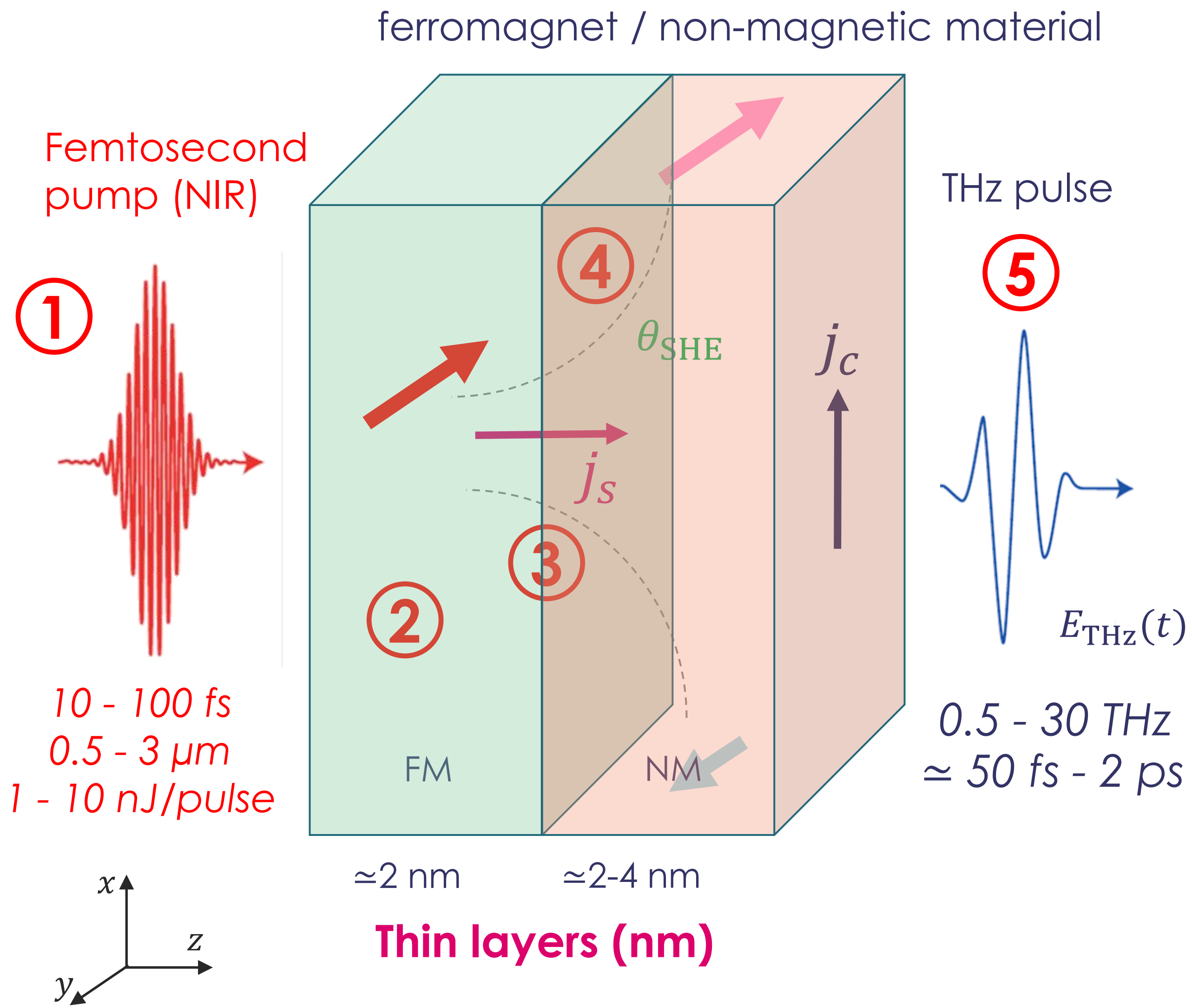
① Laser-induced ultrafast demagnetisation

② Spin current generation



Adapted from **T. Seifert *et al.*, Nat. Phot., 10, 483-488 (2016)**

Spintronic THz emitters : fundamentals of THz emission process



Adapted from **T. Seifert *et al.*, Nat. Phot., 10, 483-488 (2016)**

1. Laser-induced ultrafast demagnetisation
2. Spin current generation
3. Spin-selective transmission at the interface
4. Spin-to-charge conversion
   - Rashba-Edelstein at interfaces or surface states
   - Spin Hall effect in bulk

$$\theta_{\mathrm{SHE}}^{\mathrm{Pt}} \simeq +5\%$$

5. THz emission process

$$\nabla \times \mathbf{B}(t) = \mu_0 \boldsymbol{j_c}(t) + \mu_0 \epsilon_0 \frac{\partial \mathbf{E}}{\partial t}$$

$$\mathbf{E}(t) \propto \frac{\partial \boldsymbol{j_c}(t)}{\partial t}$$

T. J. Huisman *et al.*, Nat. Nanotech., 11, 455–458 (2016)
W. T. Lu *et al.*, Phys. Rev. B, 101, 014435 (2020)
E. Papaioannou, R. Beigang, Nanophotonics, 10(4), 1243-1257 (2020)

Material engineering of spintronic THz emitters

Magnetic reservoir

3d ferromagnets

T. Seifert *et al.*, Nat. Phot., 10, 483-488 (2016)
T.-H. Dang et al., *Appl. Phys. Rev.* 7, 041409 (2020)

Conventional magnetic reservoir

M. Fix et al. *Appl. Phys. Lett.* 117, 132407 (2020)
Spin valves as magnetically switchable spintronic THz emitters

Use of exchange bias for pinning

R. Schneider et al. *Appl. Phys. Lett.* 115, 152401 (2019)
Spintronic GdFe/Pt THz emitters

Use of FeGd alloy for magnetic compensation

Heusler | Weyl semi-metals

R. Gupta *et al.*, Adv. Optical Mater., 9, 2001987 (2021)
J. Hawecker, ER, *et al.*, Appl. Phys. Lett. 120, 122406 (2022)

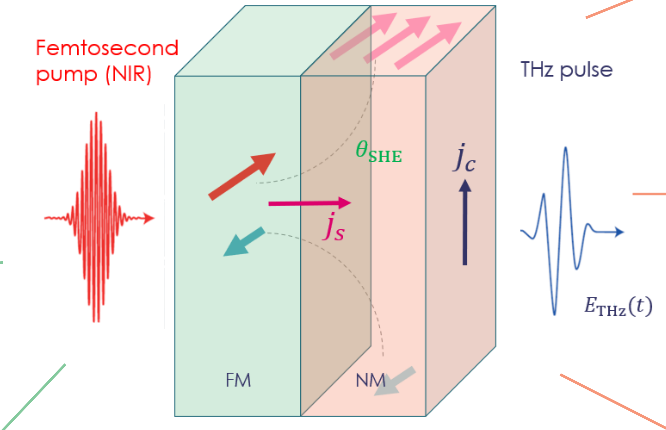
Enhanced spin-polarization near the Fermi level

Antiferromagnets

H. Qiu *et al.*, Nat. Phys. 17, 388–394 (2021)
E. Rongione *et al.*, Nat. Comm. 14, 11818 (2023)

THz magnon modes set by the high exchange field
→ Narrowband emitters

Enhancement paths explored in this work



Wave engineering

Antenna-coupling

M. Talara *et al.*, Appl. Phys. Express, 14, 4, 042008 (2021)

Optical cavities

R. I. Herapath *et al.*, Applied Physics Letters, 6, (2019)
ER *et al.*, in preparation (2023)

Spin-orbit converter

5d heavy metals

T. Seifert *et al.*, Nat. Photon., 10, 483-488 (2016)
T.H. Dang, J. Hawecker, ER, *et al.*, APR 7, 041409 (2020)

Strong inverse spin Hall effect
THz spin-sink

2D materials (TMDC)

D. Khusyainov et al., Materials, 14, 21, 6479 (2021)
L. Nádvorník *et al.*, arXiv:2208.00846 (2022)

Avoiding THz absorption and NIR-active excitons

Topological insulators

X. Wang *et al.*, Adv. Mater., 30, 1802356 (2018)
M. Tong *et al.*, Nano Letters, 21 (1), 60-67 (2021)
E. Rongione *et al.*, Adv. Optical Mater., 10, 2102061 (2022)
H. Park *et al.*, Adv. Sci. 9, 2200948 (2022)
E. Rongione *et al.*, adv. Sci. 2023, 2301124

Novel platform for interfacial SCC

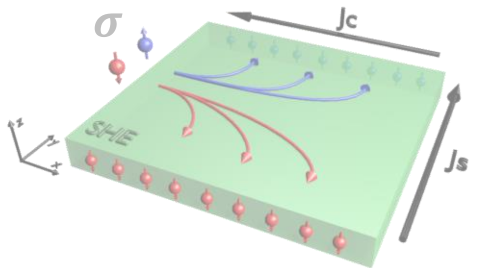
Two different interconversion mechanisms for THz generation

Inverse spin Hall effect

ISHE – BULK MECHANISM

Conversion on the bulk length-scale
→ possible radiation and spin-current absorption

$$E_{\mathrm{THz}} \propto \frac{1}{2}(g_{\uparrow}+g_{\downarrow})\sigma_{\mathrm{SHE}}\, l_{sf}\, (j_s \times \sigma)$$



Key parameter: spin Hall conductivity $\theta_{\mathrm{SHE}}^{\mathrm{eff}}$

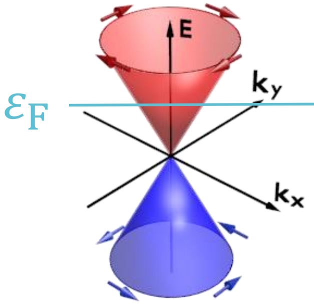
Material	Pt	Au:W	Au:Ta
σ_{SHE} $(\mathrm{k}\Omega^{-1}.\mathrm{cm}^{-1})$	3.5	1.6	5
l_{sf} (nm)	3	3	1.5

T. H. Dang, ER *et al.*, Appl. Phys. Rev., 7, 041409 (2020)
J. Hawecker, ER *et al.*, Adv. Optical Mater., 9, 2100412 (2021)
C. Rojas-Sanchez *et al.*, Phys. Rev. Lett., 112, 106602 (2014)
T. Wang *et al.*, Sci. Rep., 7, 1306 (2017)

Inverse Rashba-Edelstein effect

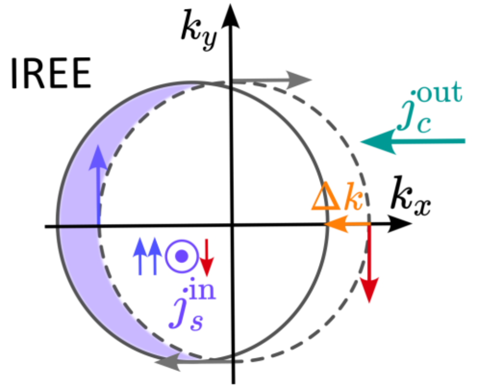
IREE – INTERFACIAL MECHANISM

Topological insulators (TI)
Strong interfacial spin-orbit coupling



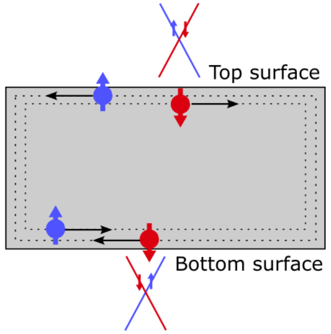
Fermi surface

$$j_c^{2D} = \lambda_{\mathrm{IEE}} \cdot j_s^{3D}$$



Topological surface states

Insulating bulk ($\rho \simeq$500 μΩ.cm)



Key parameter: Fermi velocity v_F

Material	Bi_2Te_2Se	a-Sn	Bi_2Se_3
v_F (x10^5 m.s^{-1})	11.2	7.3	2.9
$\hbar v_F$ (eV.Å)	7.4	4.8	1.9

Ultrafast SCC with TI/FM
V. Sharma *et al.*, Phys. Rev. Materials 5, 124410 (2021)
H. Park *et al.*, Adv. Sci. 2022, 9, 2200948 (2022)

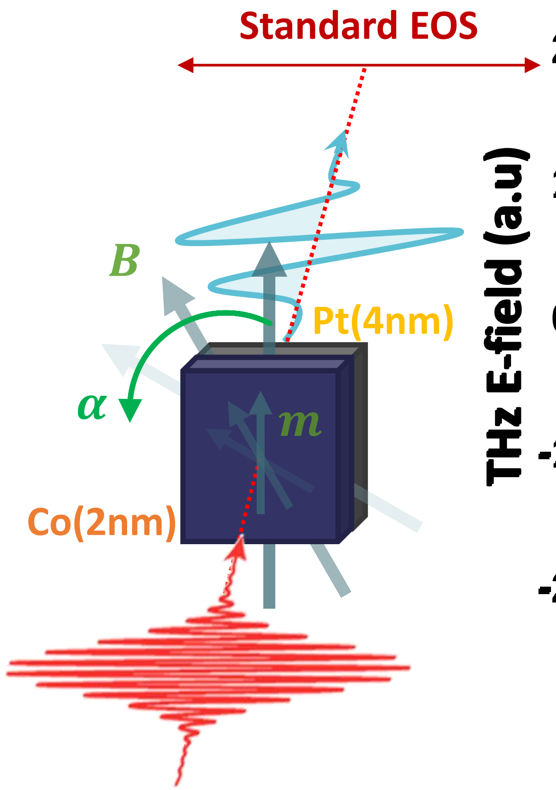
C.-F. Pai, Nat. Mat., 17, 755–757 (2018)
M. Tong *et al.*, Nano Lett., 21, 1, 60-67 (2021)

THz emission from ISHE based metallic spintronic emitter Co/Pt

Characterization and properties of spintronic emitters

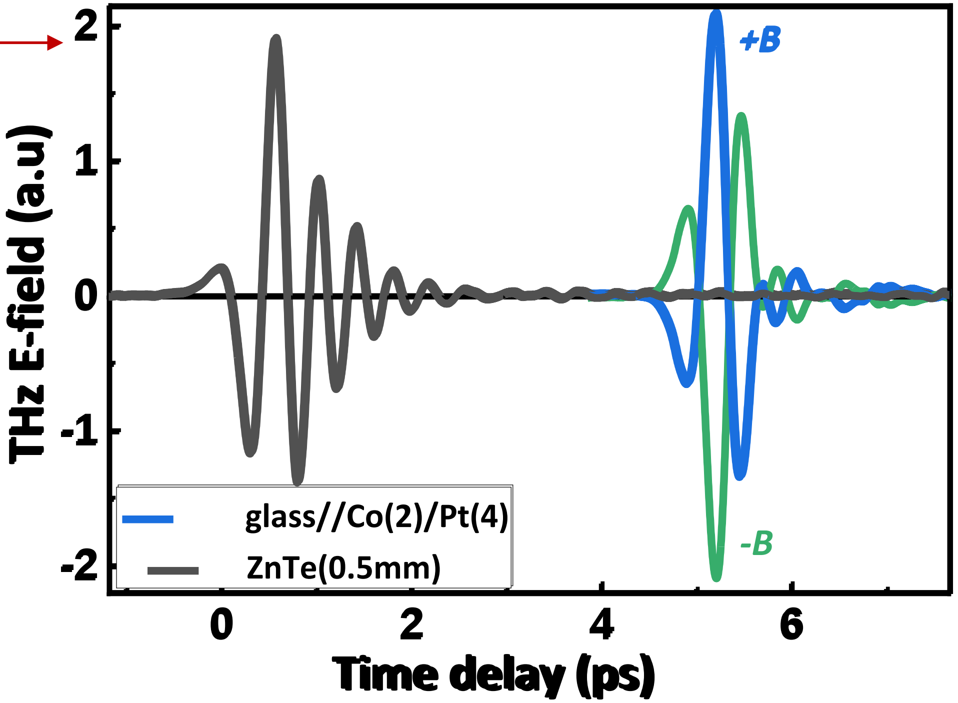
T. H. Dang, ER et al., Appl. Phys. Rev., 7, 041409 (2020)

Nanometer thin emitter

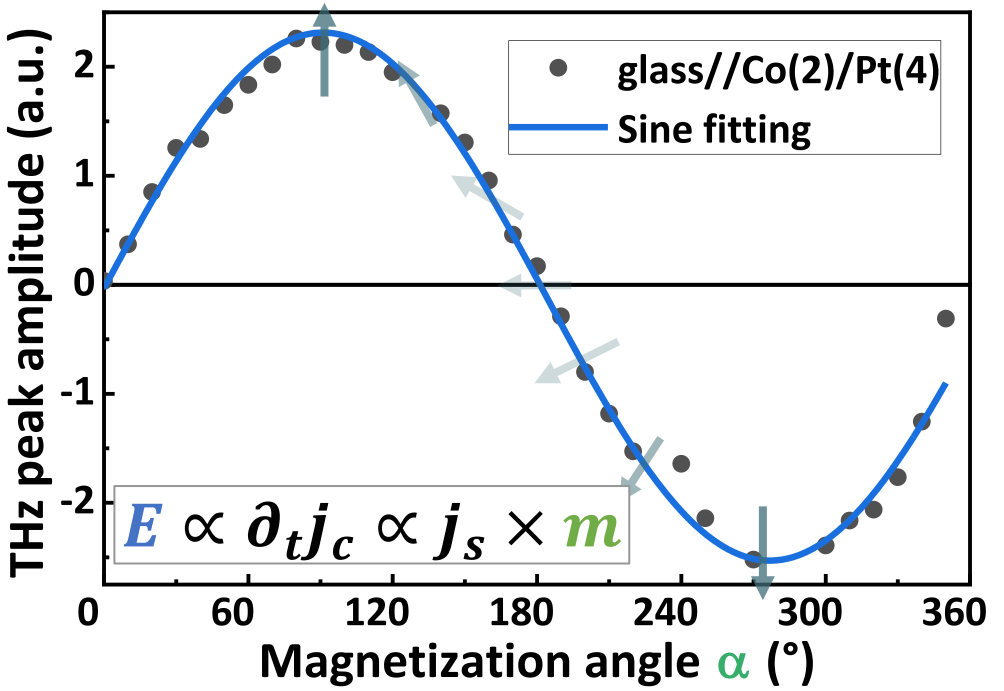


Femtosecond infrared pump

Large output field



In-plane magnetic field rotation



Sine curve: signature of spin-to-charge conversion
Maps dipolar emission with magnetic control

Similar rules applies for the IREE emission type → interfacial magnetization m_{IF}

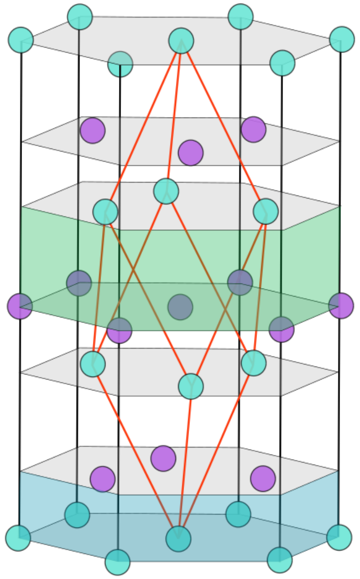
Investigation of $Bi_{1-x}Sb_x$ topological insulator

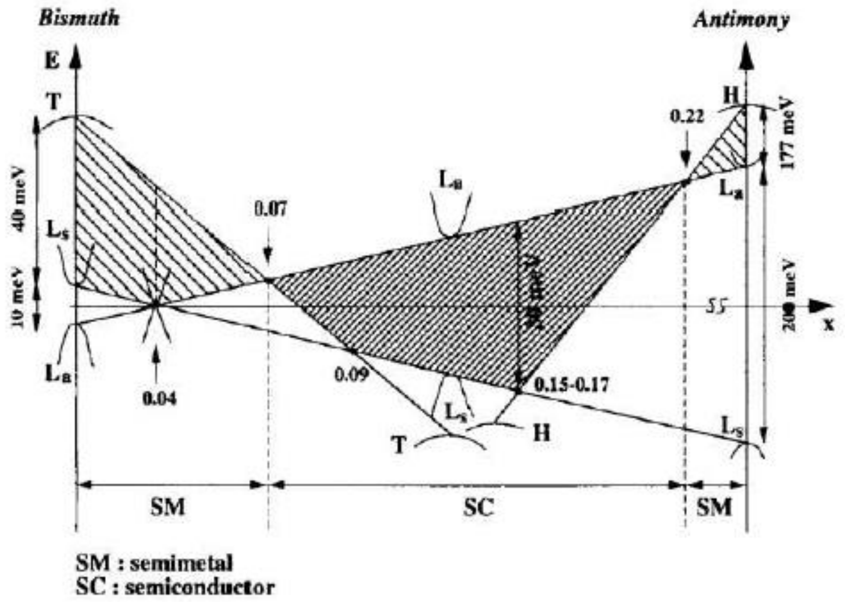
B. Lenoir et al., Semiconductors and Semimetals, Elsevier, 69:101–37 (2001)
H. Benia *et al.*, Phys. Rev. B, 91, 161406(R) (2015)

A topological insulator with stoichiometric control

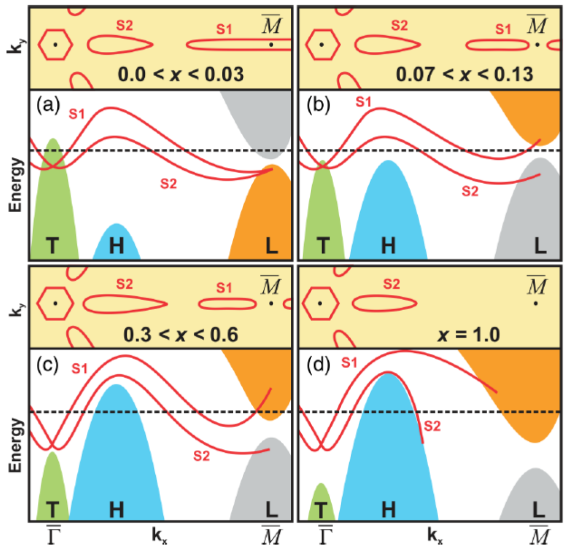
Topological window:

Strong expected SCC efficiency





B Lenoir *et al.*, ICT'96, pages 1–13. IEEE, 1996.



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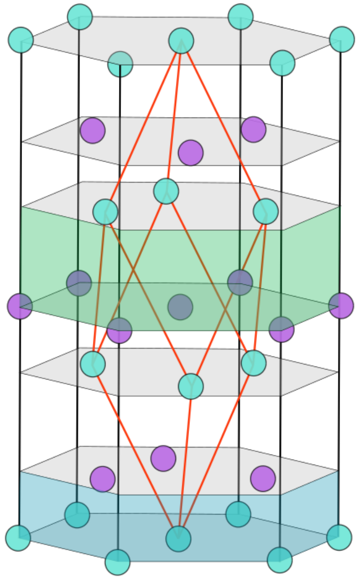
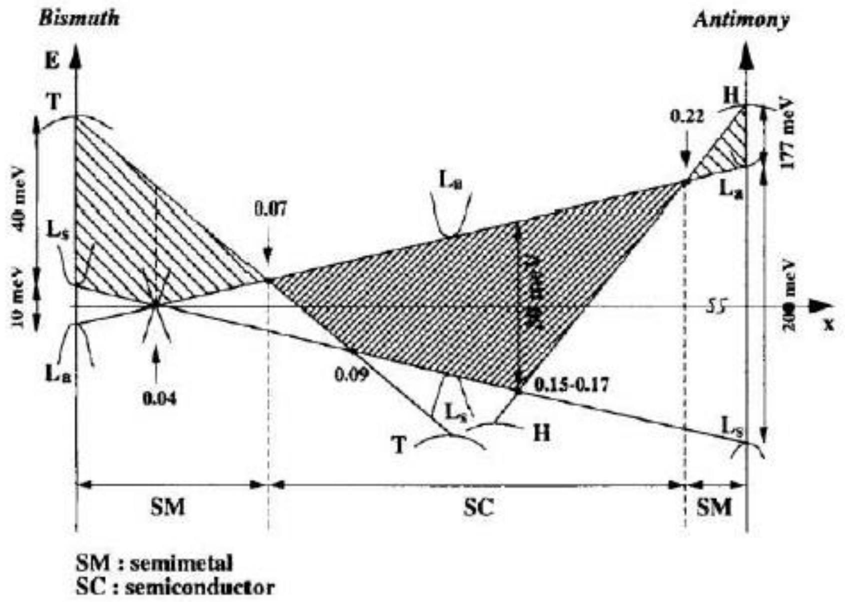
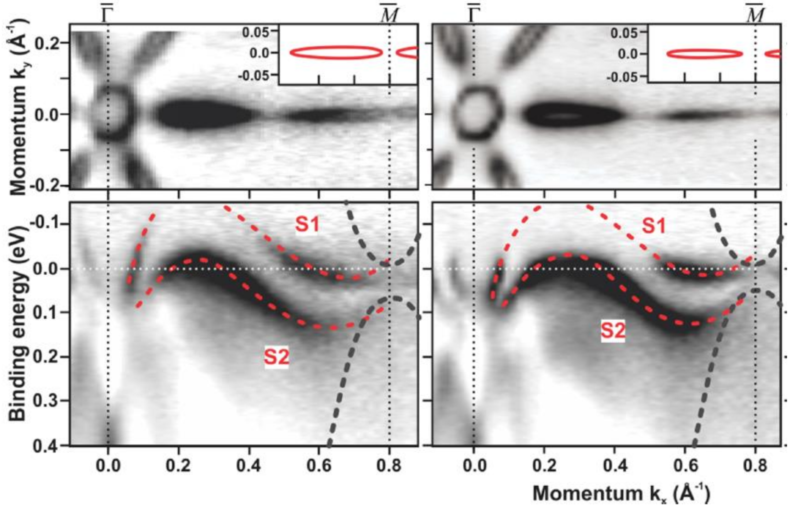




B Lenoir *et al.*, ICT'96, pages 1–13. IEEE, 1996.



Angular Resolved Photo-Emission Spectroscopy (ARPES)

$Bi_{1-x}Sb_x$ TI alloys as a template for spintronics devices

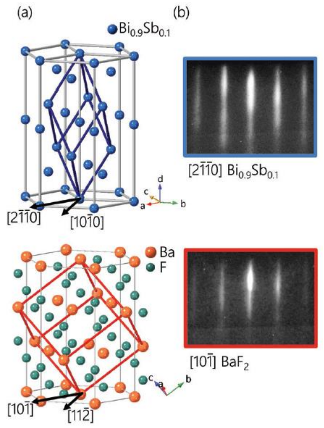
RESEARCH ARTICLE

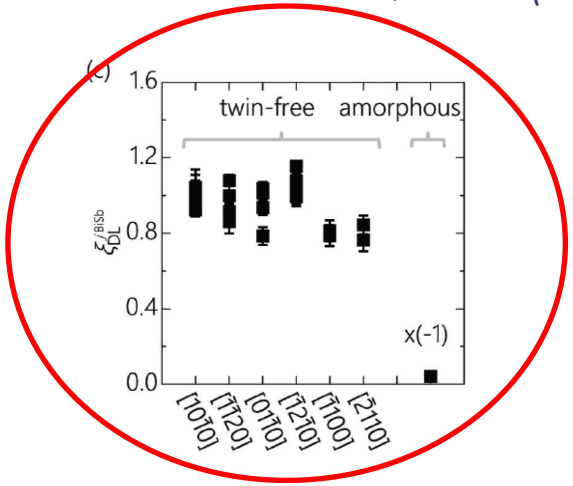
ADVANCED MATERIALS
www.advmat.de

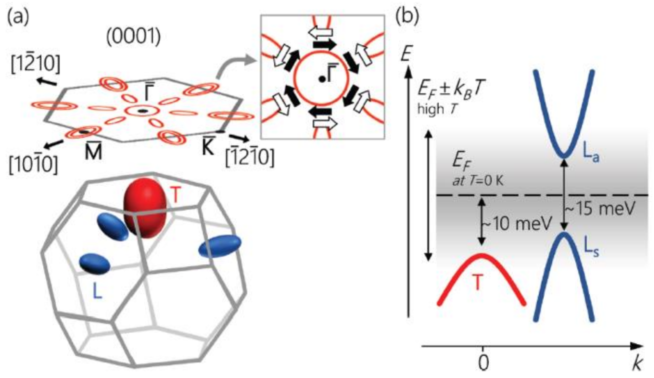
Spin–Orbit Torques and Spin Hall Magnetoresistance Generated by Twin-Free and Amorphous $Bi_{0.9}Sb_{0.1}$ Topological Insulator Films

Federico Binda, Stefano Fedel, Santos Francisco Alvarado, Paul Noël, and Pietro Gambardella**

P. Gambardella et al.,
ETH, Zurich (2023)







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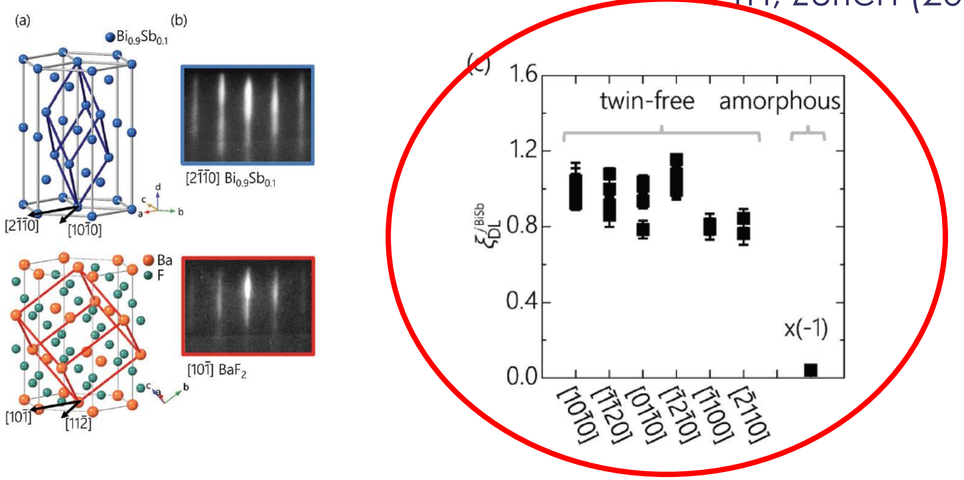
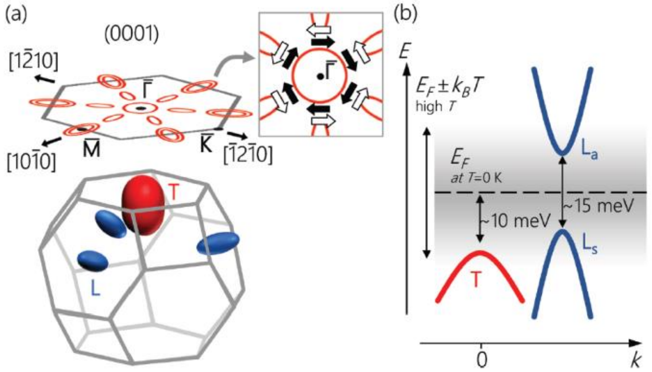
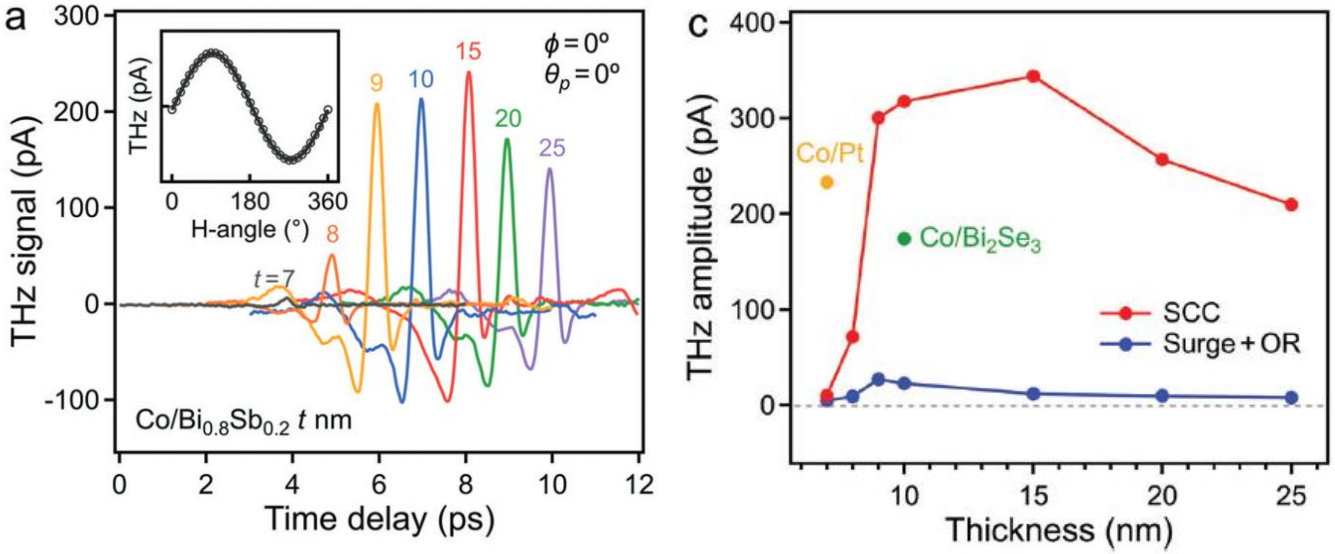
P. Gambardella, ETH, Zurich (2023)





Recent studies for ultrafast spin-injection (THz emission)



S. Rho et al., *Adv. Funct. Mater.* 2023, 2300175 (2023)
H. Park *et al.*, Adv. Sci. 2022, 9, 2200948 (2022)
V. Sharma *et al.*, Phys. Rev. Materials 5, 124410 (2021)

$Bi_{1-x}Sb_x$ topological insulator : complex Fermi surface

E. Rongione *et al.*, adv. Sci. 2023, 2301124

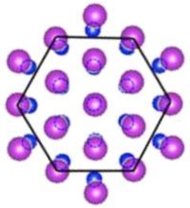
Gap increase by strain engineering and alloying composition | thickness

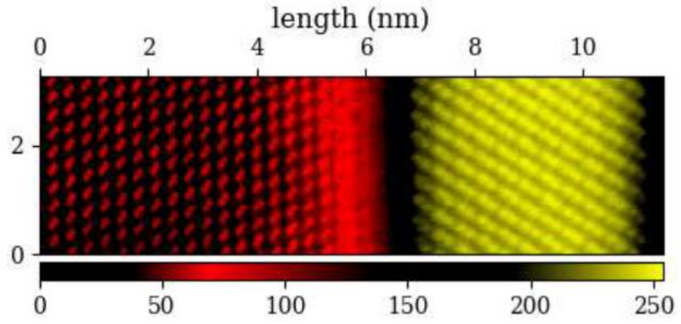
PHYSICAL REVIEW MATERIALS **6**, 074204 (2022)

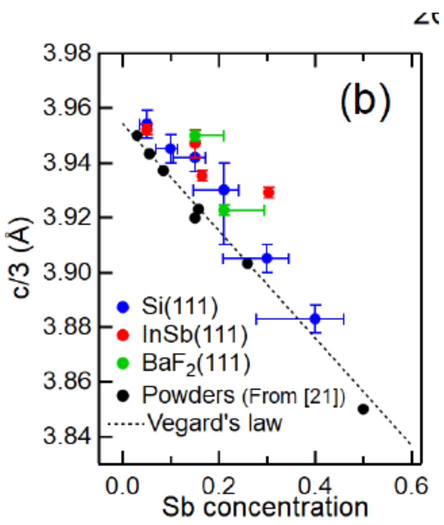
Topological surface states in ultrathin $Bi_{1-x}Sb_x$ layers

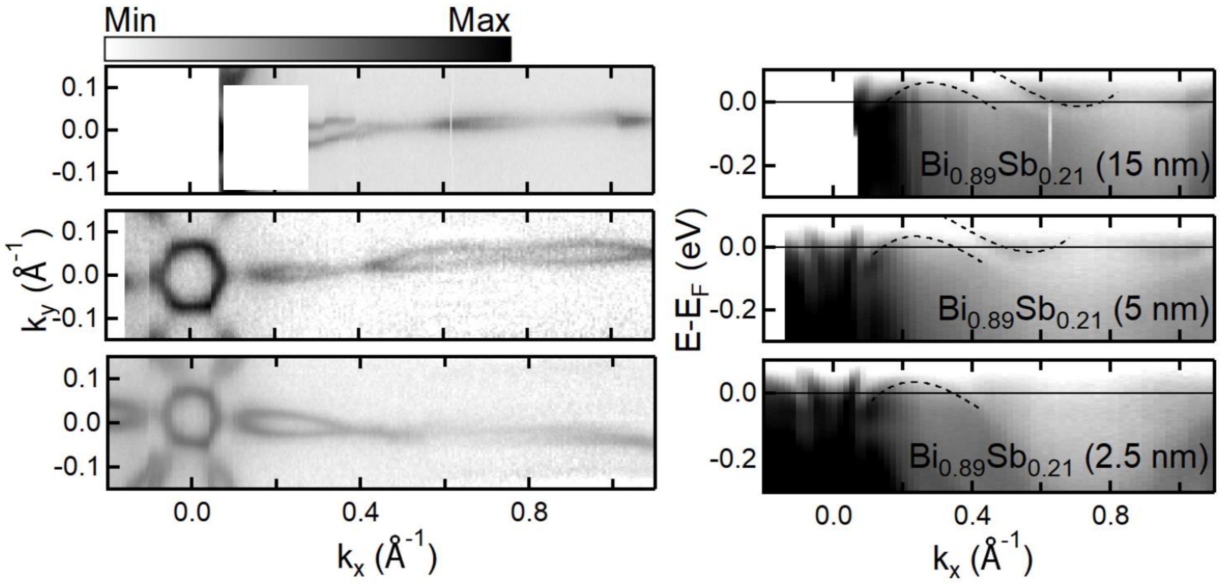
Laëtitia Baringthon,[1,2,3] Thi Huong Dang,[1] Henri Jaffrès,[1] Nicolas Reyren,[1] Jean-Marie George,[1] Martina Morassi,[2] Gilles Patriarche,[2] Aristide Lemaitre,[2] François Bertran,[3] and Patrick Le Fèvre[3,*]
[1]*Unité Mixte de Physique, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France*
[2]*Université Paris-Saclay, CNRS, Centre de Nanosciences et de Nanotechnologies, 91120 Palaiseau, France*
[3]*Synchrotron SOLEIL, Boîte Postale 48, L'Orme des Merisiers, Saint-Aubin, 91192 Gif-sur-Yvette, France*

Recent report of the TSS down to 2.5 nm









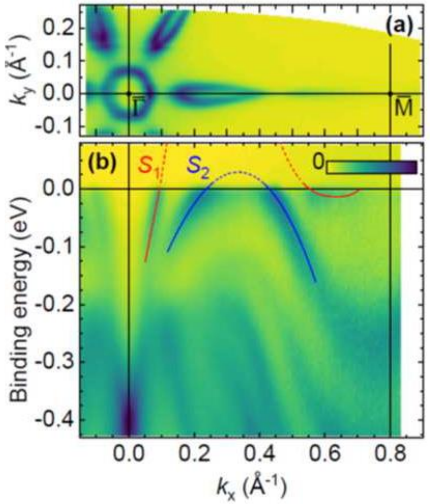
Epitaxial $Bi_{1-x}Sb_x$ (ARPES)

L. Baringthon et al., Phys. Rev. Materials 6, 074204 (2022)

$Bi_{1-x}Sb_x$ topological insulator : Electronics Band structure

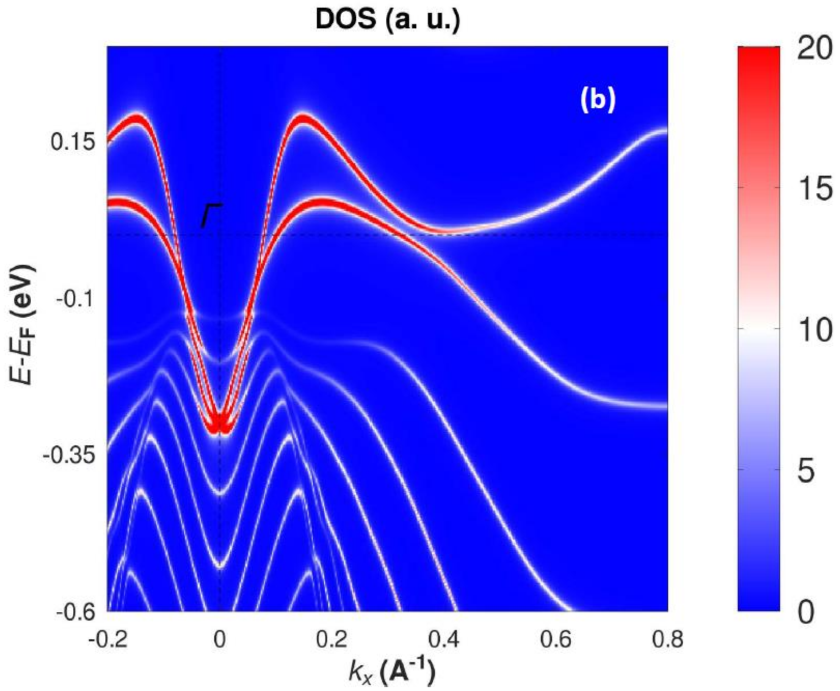
L. Baringthon *et al.*, Phys. Rev. Materials 6, 074204 (2022)

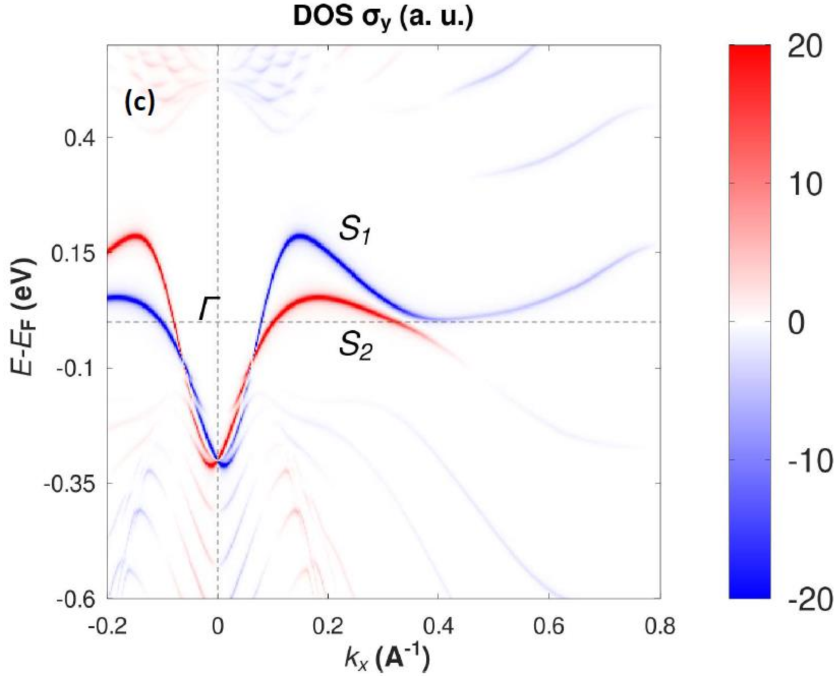
$Bi_{0.85}Sb_{0.15}$(5nm)



ARPES ELECTRONIC BAND STRUCTURE

sp^3 Tight-Binding modeling of BiSb





Angular Resolved Photo-Emission Spectroscopy (ARPES)
Sweep concentration across the topological window
Thickness variations: ultrathin $Bi_{1-x}Sb_x$ (2.5 nm) mastered at SOLEIL

Investigation of dynamical TI interconversion properties → THz emission spectroscopy

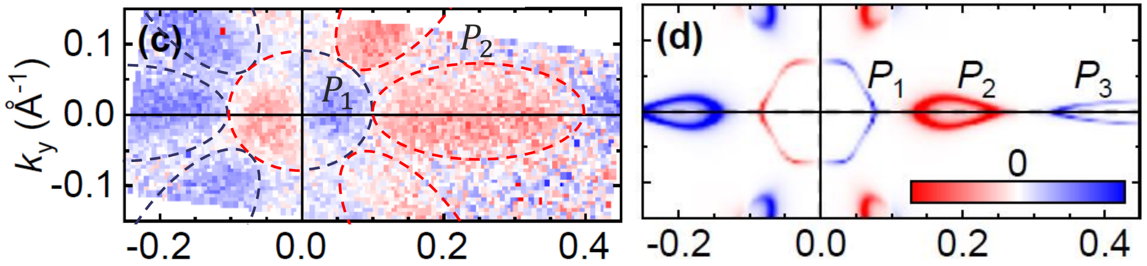
$Bi_{1-x}Sb_x$ topological surface states and spin-texture

Spin-resolved ARPES

E. Rongione *et al.*, adv. Sci. 2023, 2301124

Measured by L. Baringthon, D. She, N. Reyren, J.-M. George and P. Le Fèvre (at SOLEIL synchrotron)

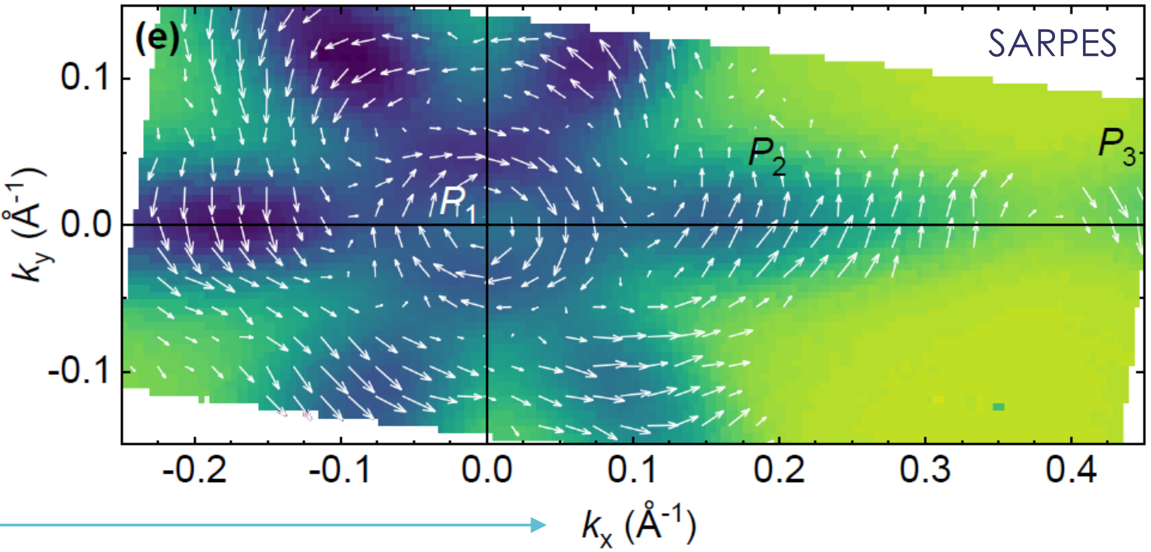
SARPES experiments
Projection of the spin σ_y
Hexagonal spin-texture



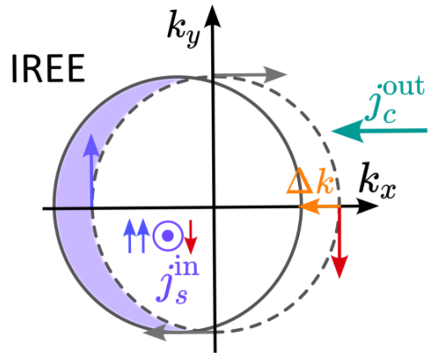
Tight-binding calculations
Projection of the spin σ_y
Rashba-contour
Agreement with the experimental measurements

Spin-projected density of states
Opposed contours near $\bar{\Gamma}$ and in the outward region

Spin-momentum locking



$k_x \equiv \overline{\Gamma M}$ direction



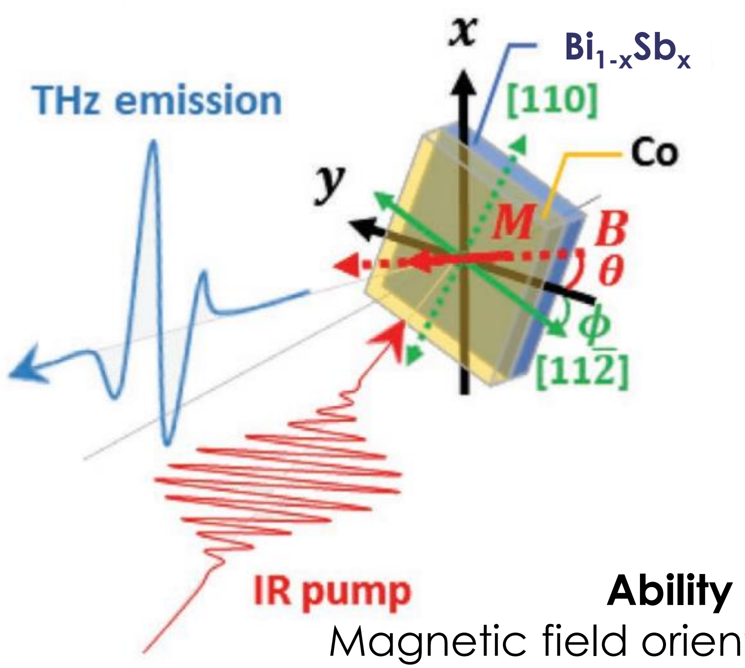
Rashba-like TSS Fermi contour
→ Co deposition for SCC

$Bi_{1-x}Sb_x$ is a favorable playground for highly-efficient IREE
Investigation of dynamical interconversion by THz emission spectroscopy

THz emission spectroscopy of Co|$Bi_{1-x}Sb_x$ bilayers

Our study: studying ultrafast spin-charge mechanisms of TI/FM

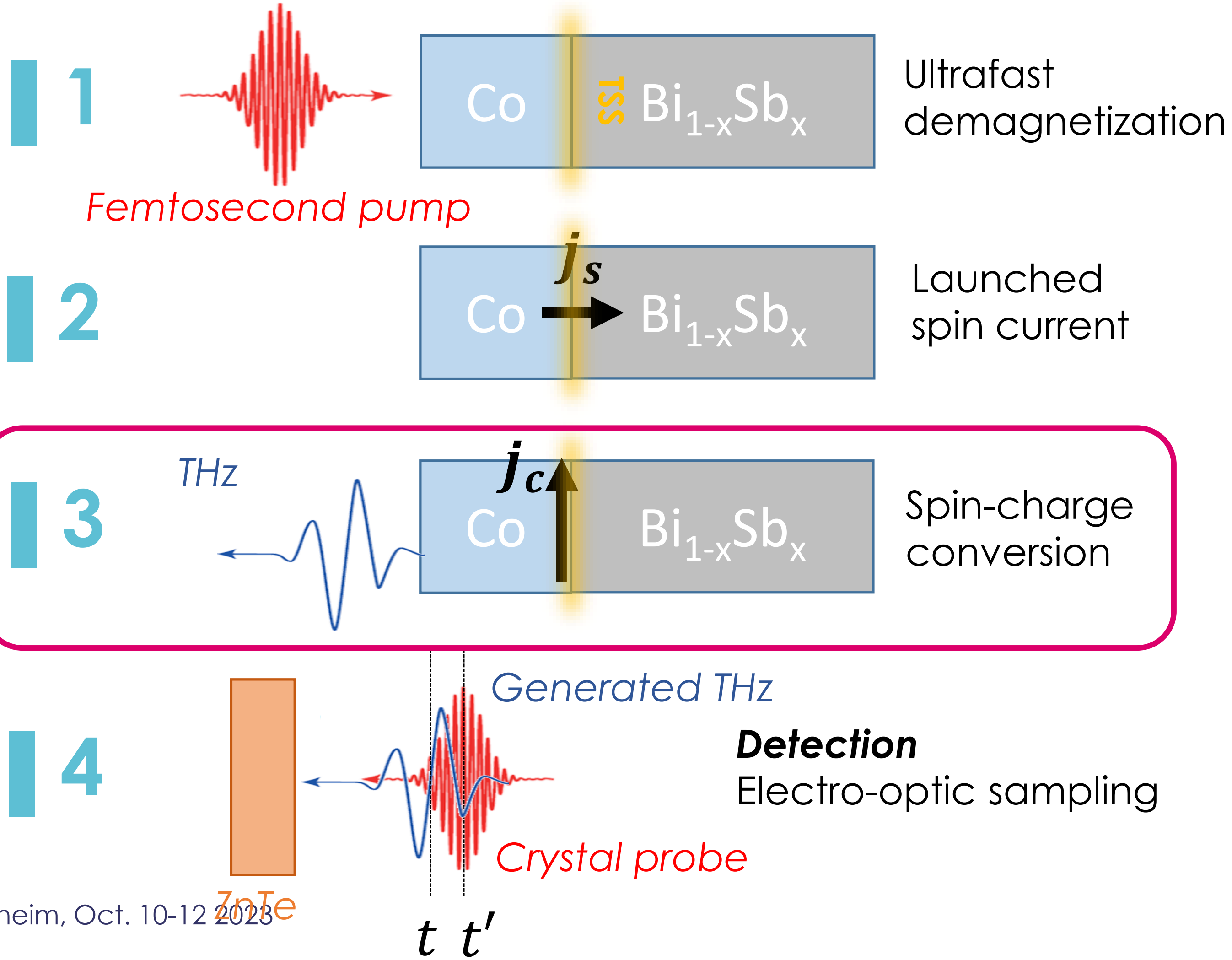
Experimental setup



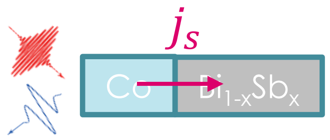
Ability to tune:
Magnetic field orientation θ
Crystalline orientation ϕ

THz spin-charge conversion mapped by THz emission spectroscopy

THz emission and detection process

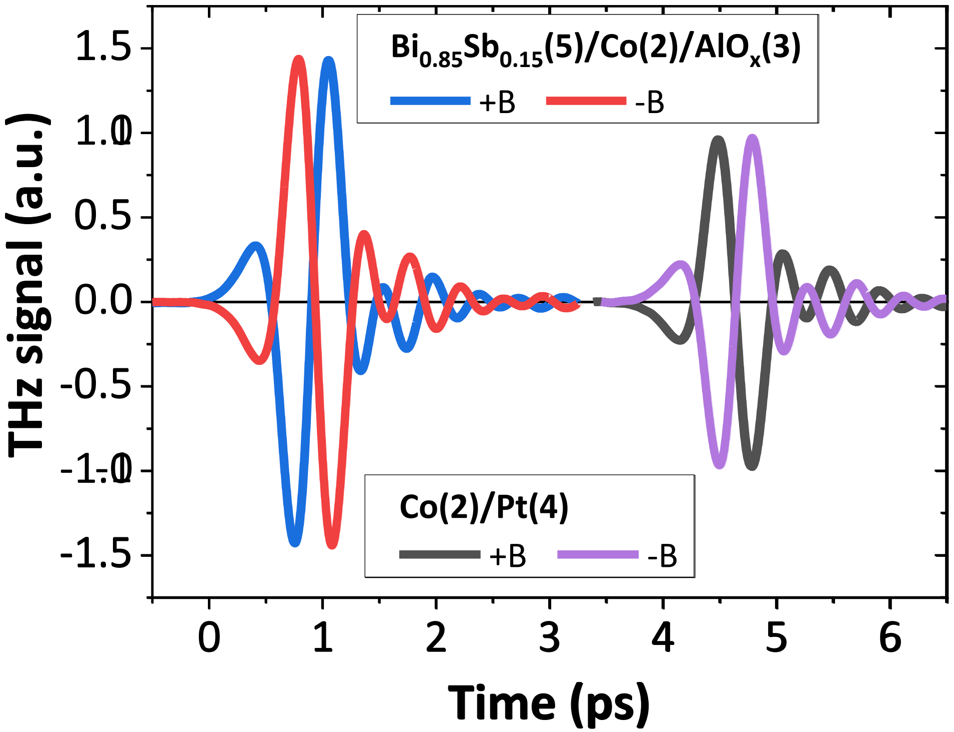


THz emission features from $Bi_{1-x}Sb_x$/Co

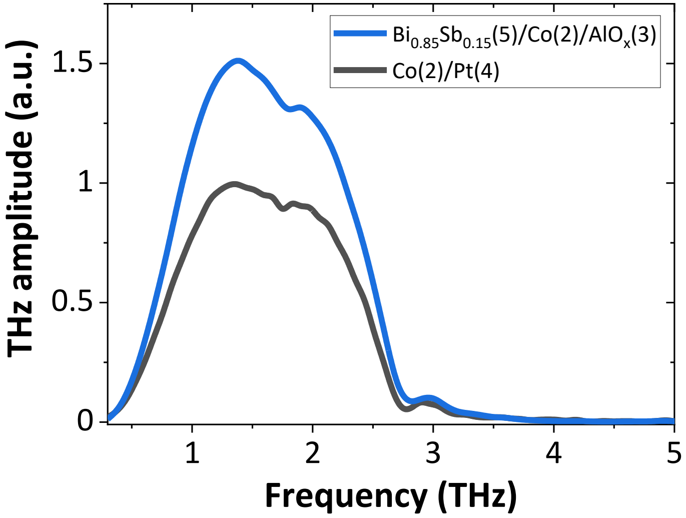


E. Rongione *et al.*, adv. Sci. 2023, 2301124

How does the THz emission from TI/FM scale with Co/Pt reference?



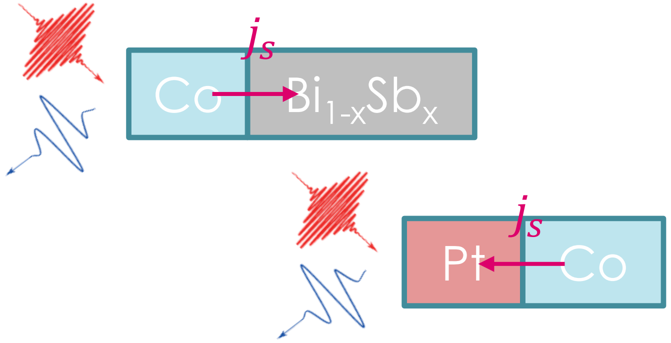
THz output



THz amplitude enhancement x1.5
THz power enhancement x2.25

SCC sign

Opposite THz phase



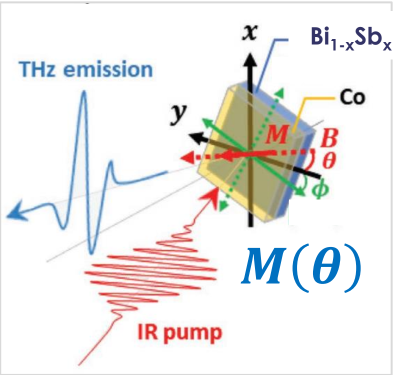
$$E_{THz} \propto \alpha_{SCC}(j_s \times m_{int})$$

Sign of the interconversion in $Bi_{1-x}Sb_x$/Co = sign of ISHE in Pt

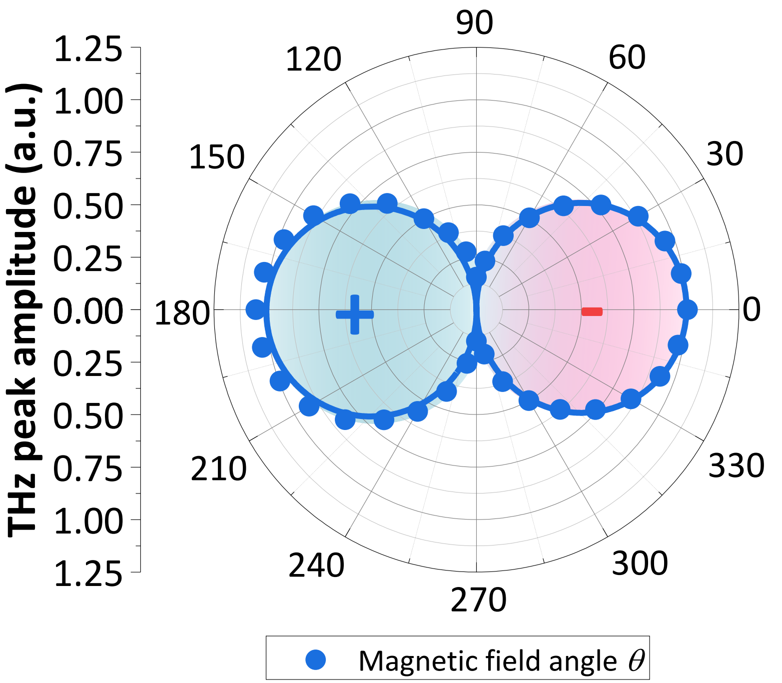
Where does this strong THz emission come from?

Emission angular symmetries for Co | $Bi_{1-x}Sb_x$

E. Rongione *et al.*, adv. Sci. 2023, 2301124

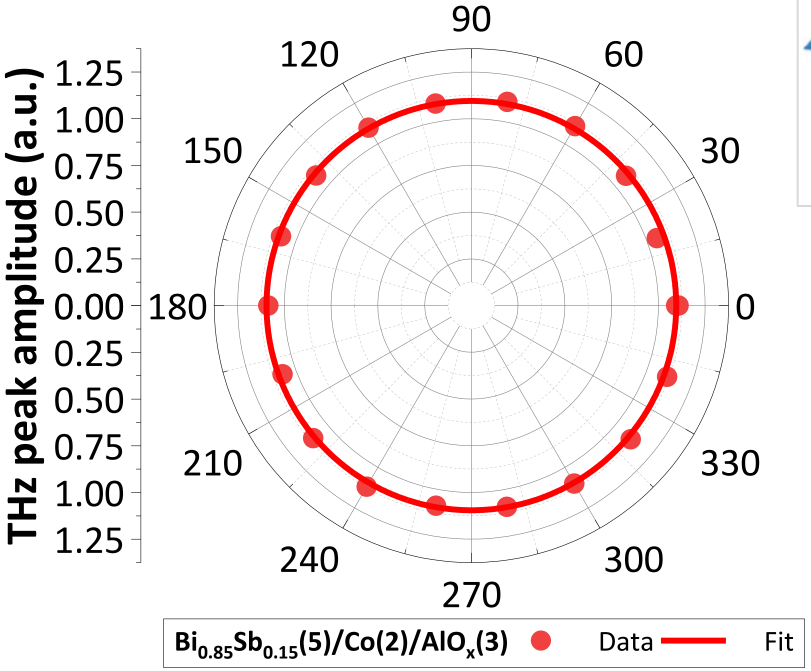


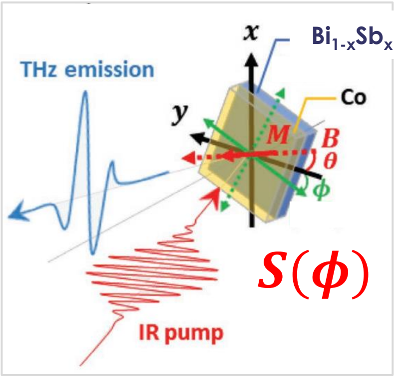
Magnetic field dependence



Uniaxial dependence → SCC-based emission

Crystallographic orientation dependence



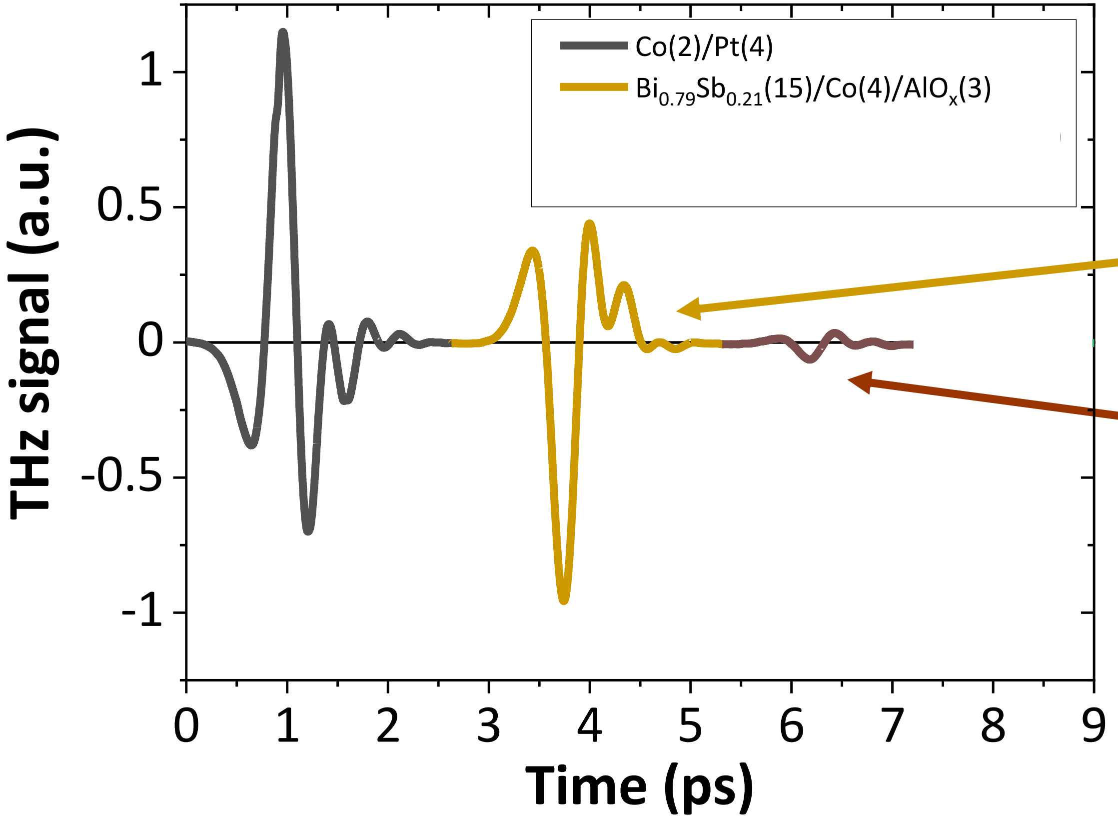


Isotropic emission → Proof of SCC

THz emission arises from spin-charge conversion

Role of the $Bi_{1-x}Sb_x$/Co interface

E. Rongione *et al.*, adv. Sci. 2023, 2301124



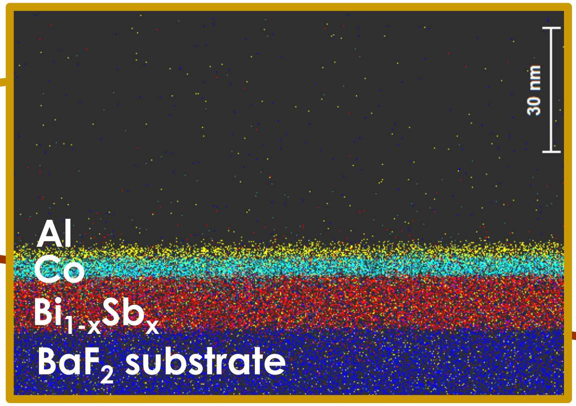
1) Interface is crucial

→ Efficient spin-injection

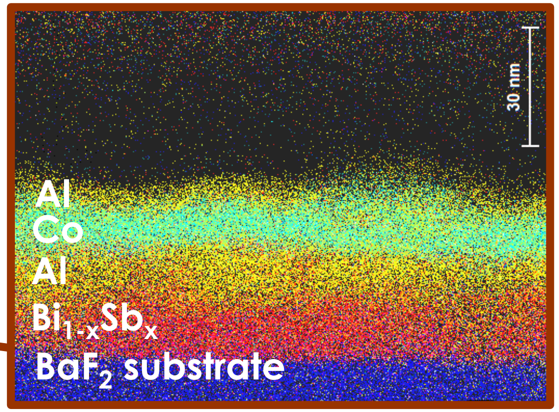
$$E_{THz} \propto \alpha_{SCC} (j_s \times m_{int})$$

→ Inefficient spin-injection

→ Destruction of the TSS



High interface quality



Al intermixing spacer

Transmission electron microscopy

2) Spin-injector is essential

→ Negligeable level of signal = non-magnetic contributions

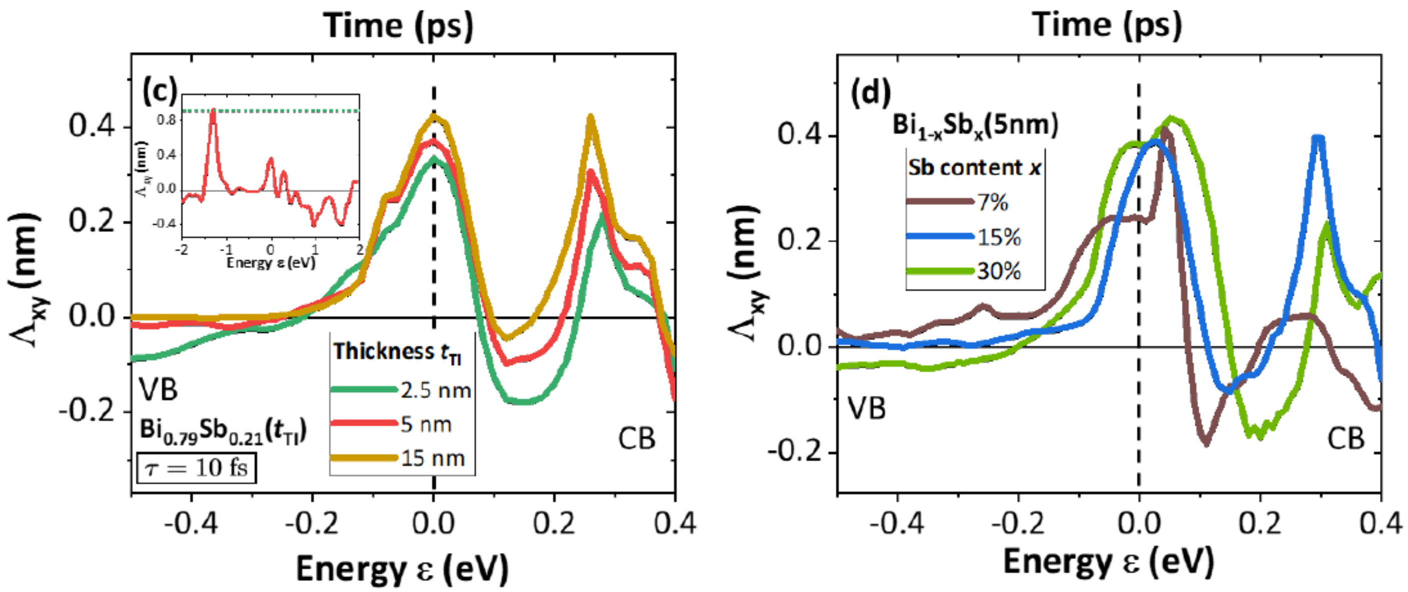
$Bi_{1-x}Sb_x$/Co THz emission → interface-sensitive

THz emission spectroscopy: efficient tool to investigate spin-injection at interfaces

Linear response theory for spin-charge interconversion in Co|$Bi_{1-x}Sb_x$

E. Rongione *et al.*, adv. Sci. 2023, 2301124

Slab-TB framework



Inverse Rashba-Edelstein length

$$\Lambda_{xy}^{\mathrm{IREE}} = \frac{\sum_{n,k}\left(\sigma_{nk}^{y} v_{nk}^{x} \tau_s \frac{\partial f_{nk}}{\partial \varepsilon}\right)}{\sum_{n,k}\left(\frac{\partial f_{nk}}{\partial \varepsilon}\right)}$$

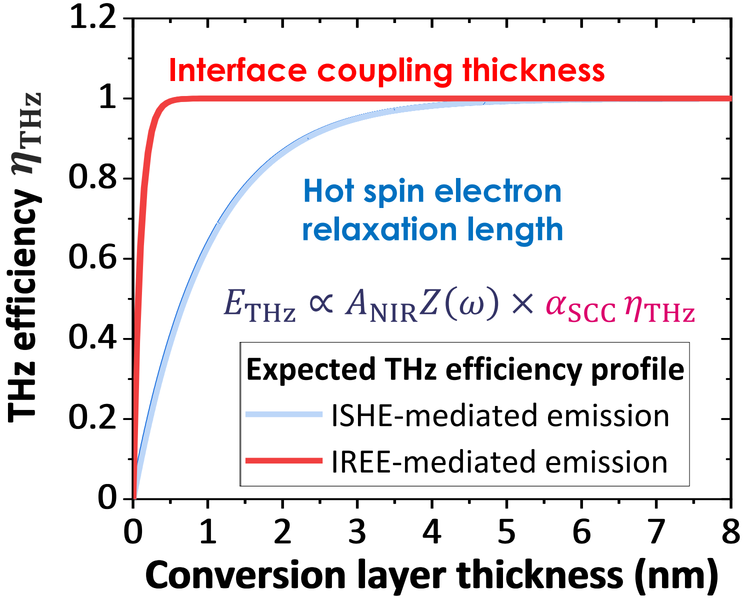
Typical spin relaxation time on TSS = 10 fs (from TB calculation)

$Bi_{1-x}Sb_x$ thickness dependence on the THz emission

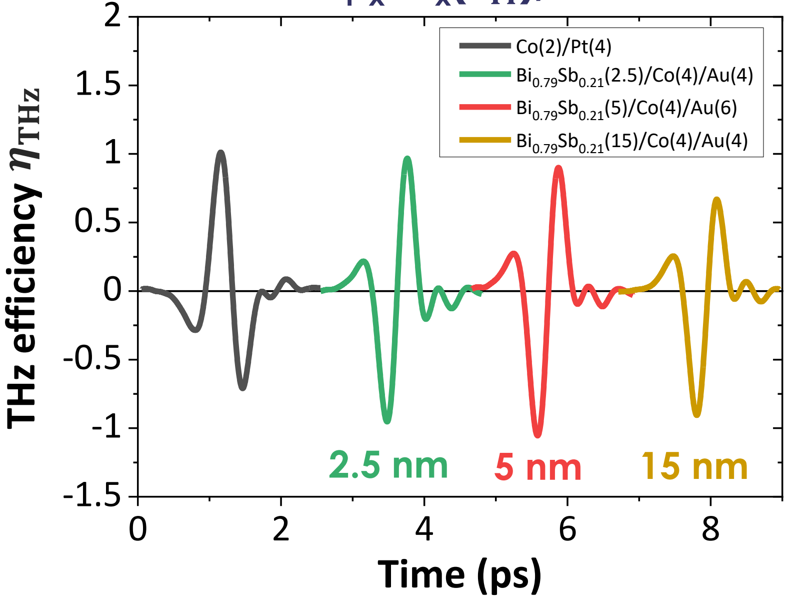
E. Rongione *et al.*, adv. Sci. 2023, 2301124

How to discriminate conversion mechanisms in TI/Co from THz emission?

THz efficiency thickness dependence



$Bi_{1-x}Sb_x(t_{TI})$/Co



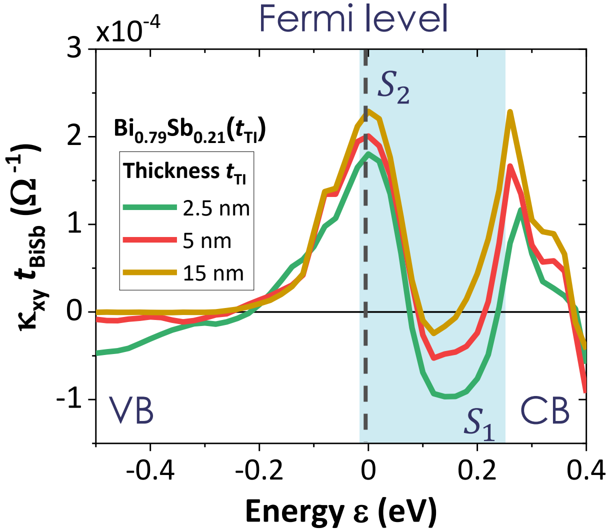
THz efficiency larger than metallic STE Co/Pt
High thin film quality

IREE efficiency

Sizeable THz signal at 2.5 nm

→ Interfacial scenario for SCC

→ Possible enhancement by TSS

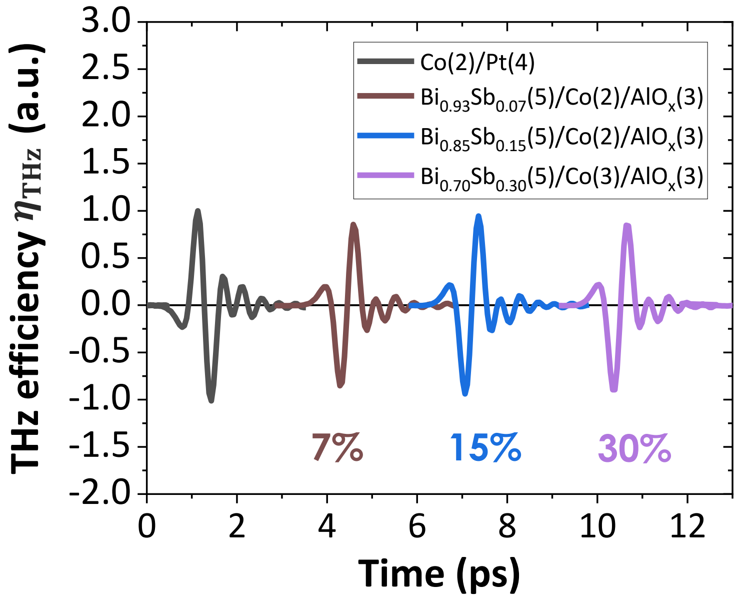


Thickness-independent THz signal → Evidence for interfacial-mediated SCC scenario by TSS

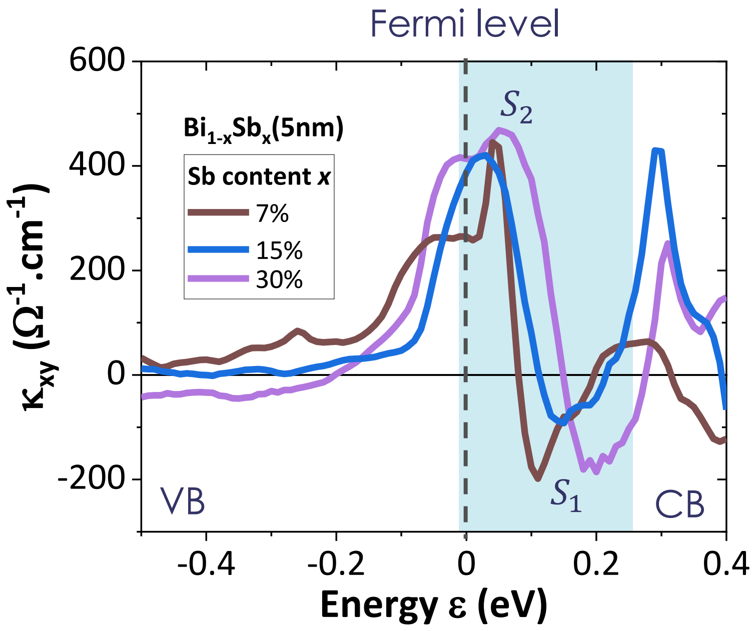
$Bi_{1-x}Sb_x$ alloy concentration dependence on the THz emission

E. Rongione *et al.*, adv. Sci. 2023, 2301124

Experimental THz SCC



IREE efficiency



Role of the surface states

Topological character might not be necessary for IREE conversion

Only hybridized Rashba-like surface states

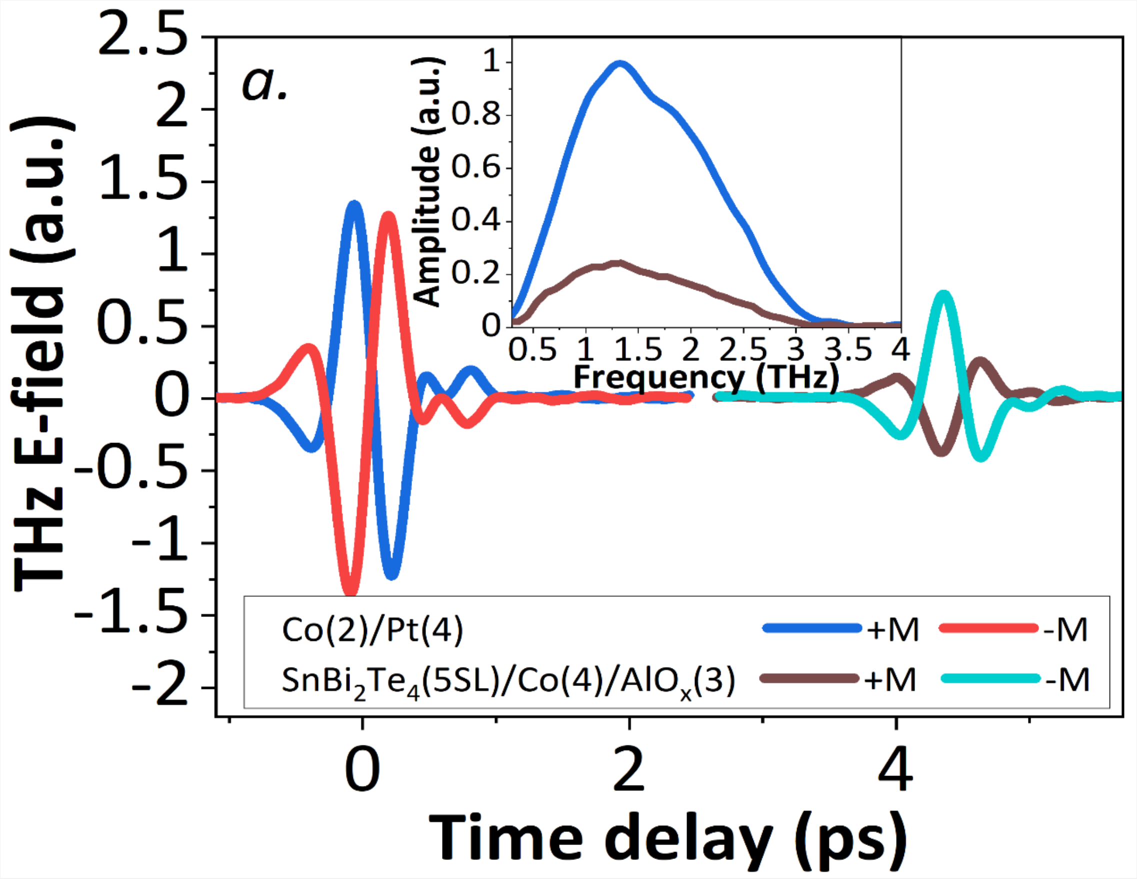
Perspectives for integrated TI-based spintronic devices

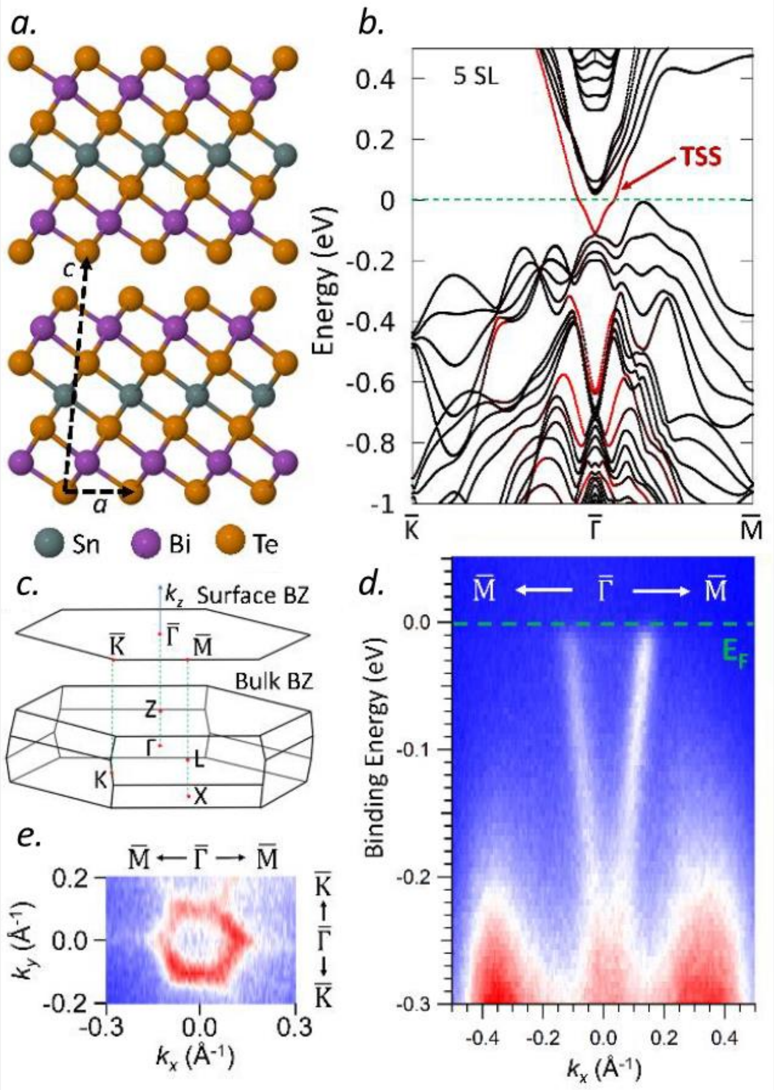
THz SCC via hybridized Rashba-like surface states

Bi_2SnTe_4 topological insulators probed by THz

ER *et al.*, Adv. Optical Mater., 10, 2102061 (2022)

Opposite phase on THz => **BST and Pt has the same SHE sign**





Si(HR)//Co(2)/Pt(4)

MBE2382: Si(111)/InAs(111)//BST(5SL)/Co(4)/Al(3)

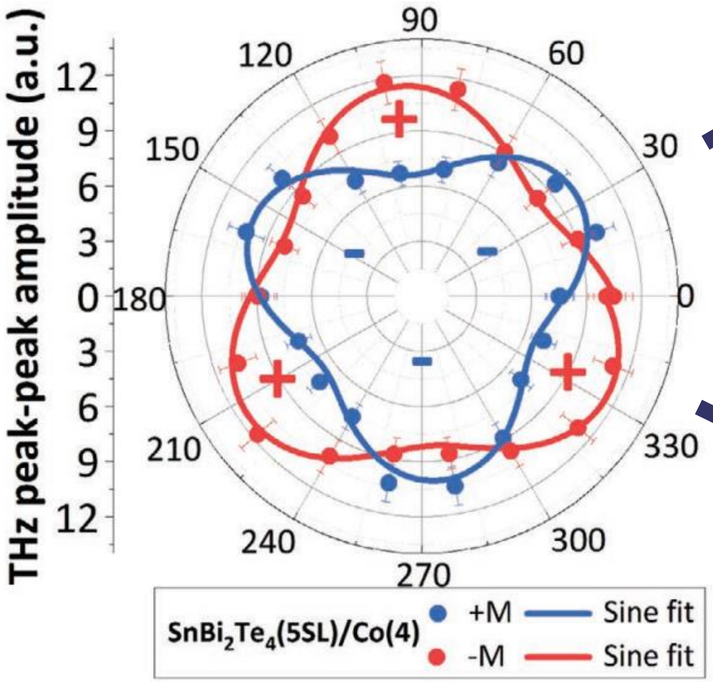
Collaboration with A. Dimoulas, INN, Athens (Greece) – European FET PROACTIVE SKYTOP (2019-2023)

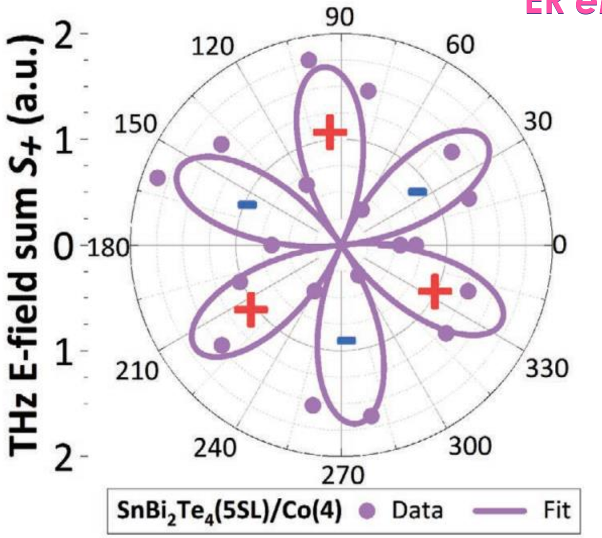
Extraction of the emission symmetries for Co|$SnBi_2Te_4$

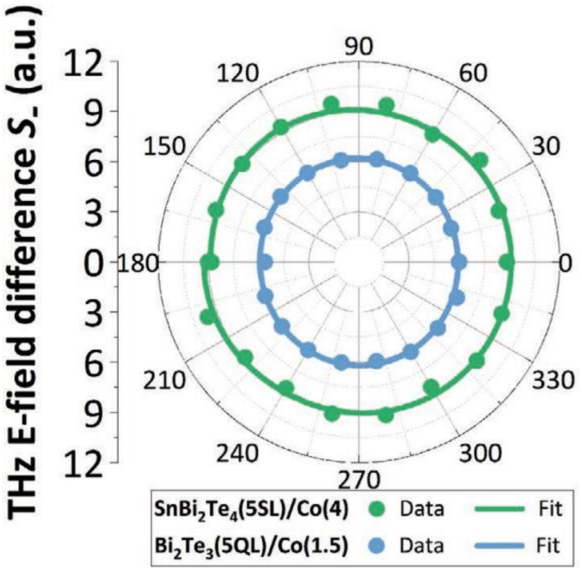
ER *et al.*, Adv. Optical Mater., 10, 2102061 (2022)

Necessity to separate THz contributions

Measured azimuthal dependence







Non-magnetic contributions

$$S_+ = \frac{S(+M) + S(-M)}{2}$$

Typically 15-20% of the THz signal

Magnetic contributions

$$S_- = \frac{S(+M) - S(-M)}{2}$$

Stronger THz signal for $SnBi_2Te_4$ compared to its parent material Bi_2Te_3

Spin-charge related contribution: main component isotropic *vs.* cyrstalline orientation

Conclusion and perspectives

THz spintronic emitters based on heavy-metal based heterostructures

Gapless broadband emission ; **polarization tuned by magnetic field** ; ***nm*-thin passive emitters**

Impact of material properties and interfaces on THz emission

- THz emission spectroscopy: powerful tool to **investigate spin-to-charge interconversion efficiency**
- Key emission parameters → **interface quality with** $g_{\uparrow} + g_{\downarrow}$ and **spin Hall conductivity** σ_{SHE}

For more information about this work: T. H. Dang, ER et al., Appl. Phys. Rev., 7, 041409 (2020)
J. Hawecker, ER et al., Adv. Optical Mater., 2100412 (2021)

New type of emitters based on TI surface-states - Spectroscopy

- TI are interesting candidates for high power emitters due to strong interfacial conversion

→ Isotropic conversion mapped by **isotropic crystalline emission** and **thickness-independent emission**

→ **Demonstration of strong THz emission** : $Bi_{1-x}Sb_x$|Co

→ Reliable technique to measure spin-charge interconversion

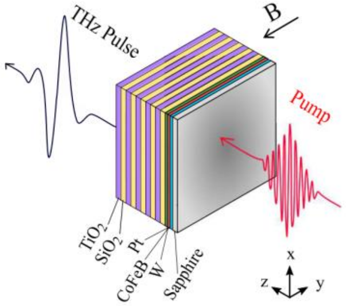
For more information about this work: L. Baringthon et al., Phys Rev Materials (2022)
E. Rongione et al., Advanced Science 2023

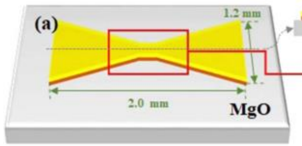
Perspectives on THz spintronics

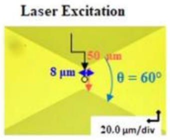
THz engineering

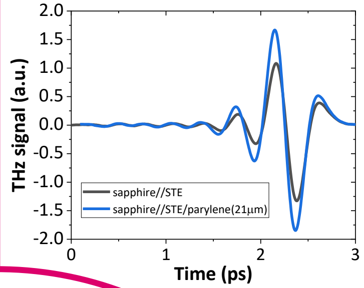
R. I. Herapath *et al.*, Applied Physics Letters, 6, (2019)
M. Talara *et al.*, Appl. Phys. Express, 14, 4, 042008 (2021)
ER *et al.*, in preparation (2023)

cavities, antenna, anti-reflective coatings, *etc.*









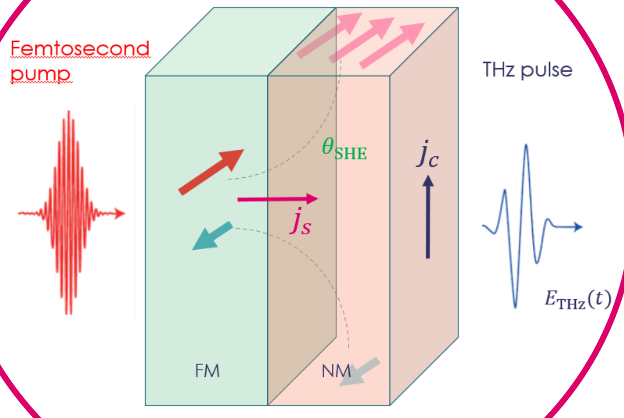
Heterostructure engineering

combining all enhancement tracks together

- spin-sink: x2.5 in power
- THz cavity (ARC): x2.6 in power
- NIR Bragg mirror: x4 in power
- THz antenna : x2 in amplitude

Total : <u>x25-50</u> enhancement in THz power

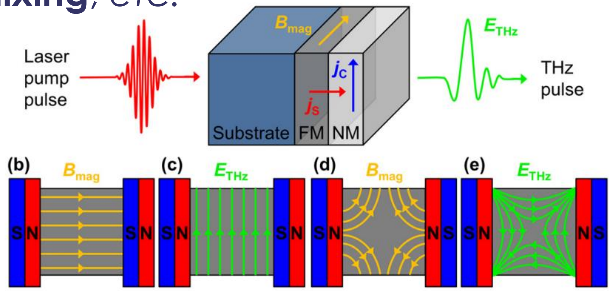
Medium term



Enhancing THz spintronics functionalities

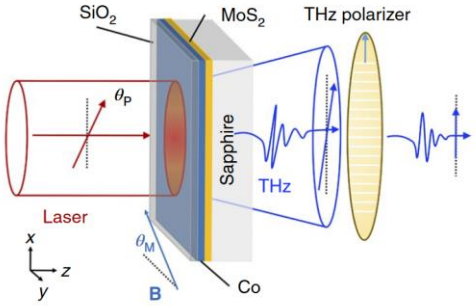
M. T. Hibberd *et al.*, Appl. Phys. Lett., 114, 3, 031101 (2019)
O. Gueckstock *et al.*, Optica, 8, 7, 1013 (2021)
G. Lezier *et al.*, Appl. Phys. Lett., 120, 15, 152404 (2022)

Polarization control, THz modulation, narrowband emission, **spintronic photomixing**, *etc.*



Ultrafast SCC

Two-fold objectives



- Study the spin diffusion properties at the **very short timescales**
- Access to spin **interfaces**

- Find stronger SCC: Pure interfacial systems: 2D materials

L. Cheng *et al.*, Nat. Phys., 15, 4, 347–351 (2019)
L. Nádvorník *et al.*, arXiv:2208.00846 (2022)
E. Rongione *et al.*, in prepration (2023)

Spectroscopy tool in ultrafast magnetism

Recent spectroscopy tool

Ultrafast demagnetization

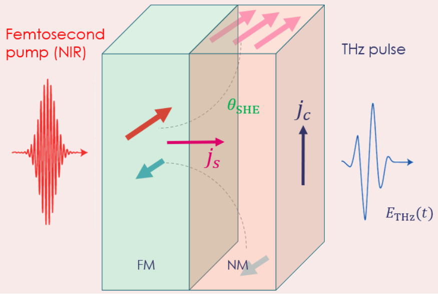
E. Beaurepaire *et al.*, Phys. Rev. Lett., 76, 22, 4250–4253 (1996)

Probing ultrafast spin transport

A. Melnikov *et al.*, Phys. Rev. Lett., 107, 7, 076601, (2011)
M. Battiato *et al.*, Phys. Rev. B, 86, 2, 024404, (2012)
T. Kampfrath *et al.*, Nature Nanotech., 8, 4, 256–260 (2013)

ISHE + spin-transport → towards THz emission

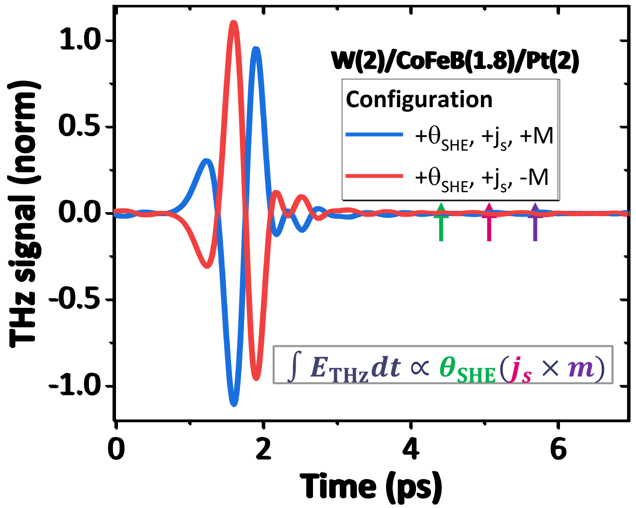
T. Seifert *et al.*, Nat. Phot., 10, 483-488 (2016)
T. J. Huisman *et al.*, Nat. Nanotech., 11, 455–458 (2016)

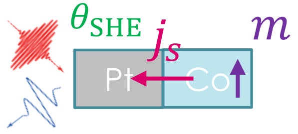


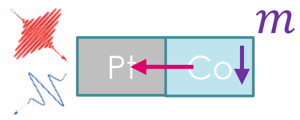
Recent reviews (2022)

C. Bull *et al.*, APL Materials 9, 090701 (2021)
T. Seifert *et al.*, Appl. Phys. Lett. 120, 180401 (2022)

Link with spintronics







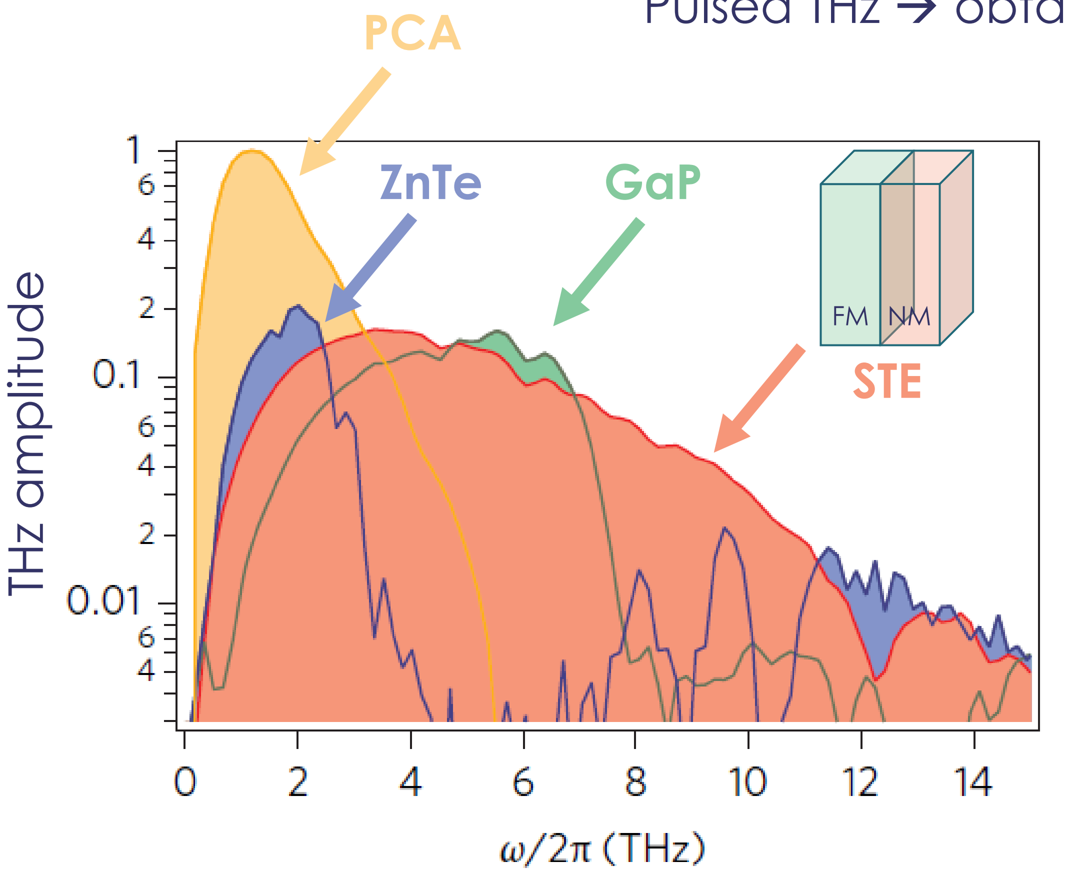
- Determination of the spin Hall angle θ_{SHE}
- Determination of the spin current sign j_s
- Determination of the magnetization sign m

Probe new interconversion mechanisms and THz functionalities

State-of-the-art of pulsed THz sources

S. S. Dhillon *et al.*, J. Phys. D: Appl. Phys., 50, 043001 (2017)
M. Tonouchi, Nature Photonics, 1, 97–105, (2007)

Pulsed THz → obtained with ultrashort (fs) laser pump



Photoconductive antennas
Optical-gap semiconductors
Electron-hole recombination time

Non-linear crystals
ZnTe, GaP, GaSe
Limited by phonon absorptions

Spintronic THz emitters

T. Seifert *et al.*, Nat. Phot., 10, 483-488 (2016)
T. J. Huisman *et al.*, Nat. Nanotech., 11, 455–458 (2016)

Gapless broadband THz generation (0.3-30 THz)
Polarization control by B-field

Pulsed emission → demonstrated
CW emission → preliminary results (IEMN)

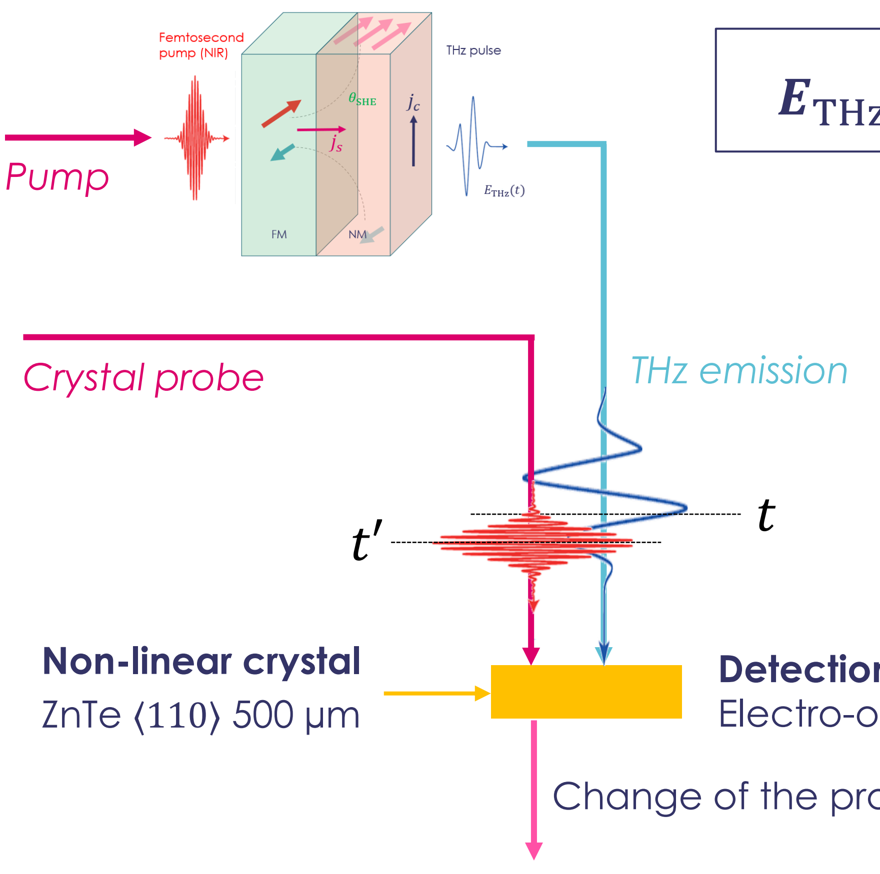
Spintronic THz emitters are technological-friendly THz sources: cheap, modulable and patternable sources

THz spectroscopy of spin-injection and charge conversion

Collab. nano-THz team, S. Dhillon (LPENS)

THz emission spectroscopy

THz generation



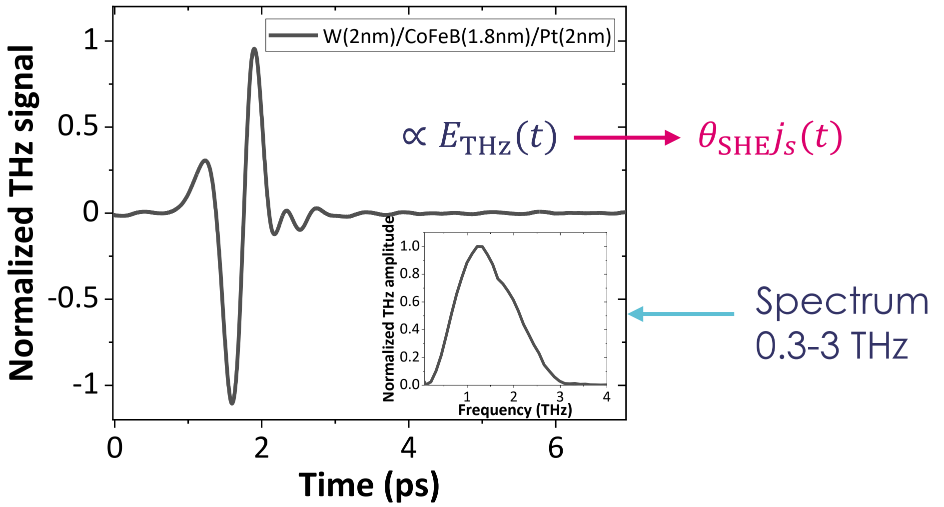
$$\boldsymbol{E}_{\mathrm{THz}} \propto \frac{\partial \boldsymbol{j}_c}{\partial t} \propto \theta_{\mathrm{SHE}}(\boldsymbol{j}_s \times \boldsymbol{\sigma})$$

Detection
Electro-optic sampling

Change of the probe polarization

Balanced photodetection

Time-resolved THz pulse



- Spectroscopy of sub-picosecond currents
- Access to the amplitude and phase
  → Determination of the charge current sign and dynamics

Access to fundamental relaxation times in spintronic THz emitters

Spectroscopy tool in ultrafast magnetism

Recent spectroscopy tool

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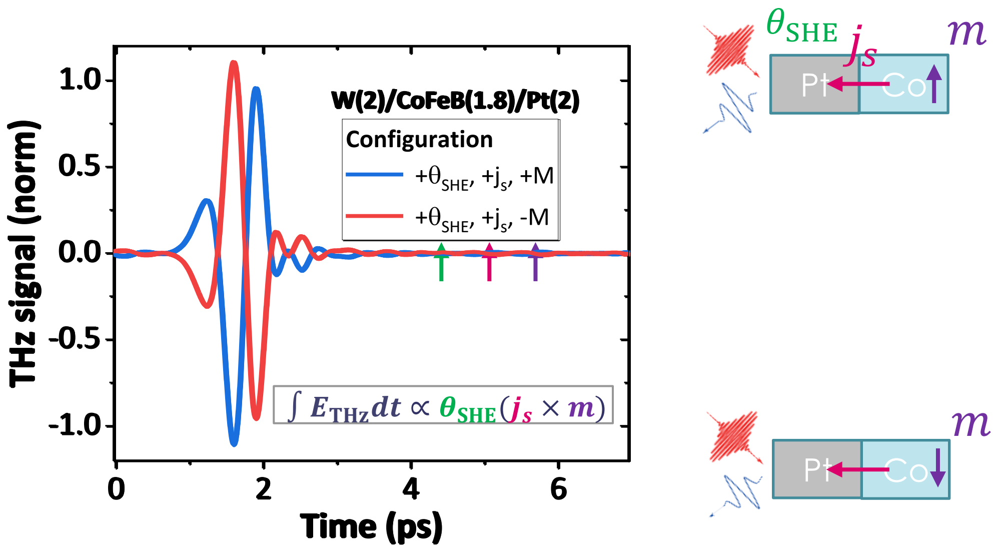
T. J. Huisman *et al.*, Nat. Nanotech., 11, 455–458 (2016)

- Determination of the spin Hall angle θ_{SHE}

Recent reviews (2022)

C. Bull *et al.*, APL Materials 9, 090701 (2021)
T. Seifert *et al.*, Appl. Phys. Lett. 120, 180401 (2022)

Link with spintronics



- Determination of the spin Hall angle θ_{SHE}
- Determination of the spin current sign j_s
- Determination of the magnetization sign m

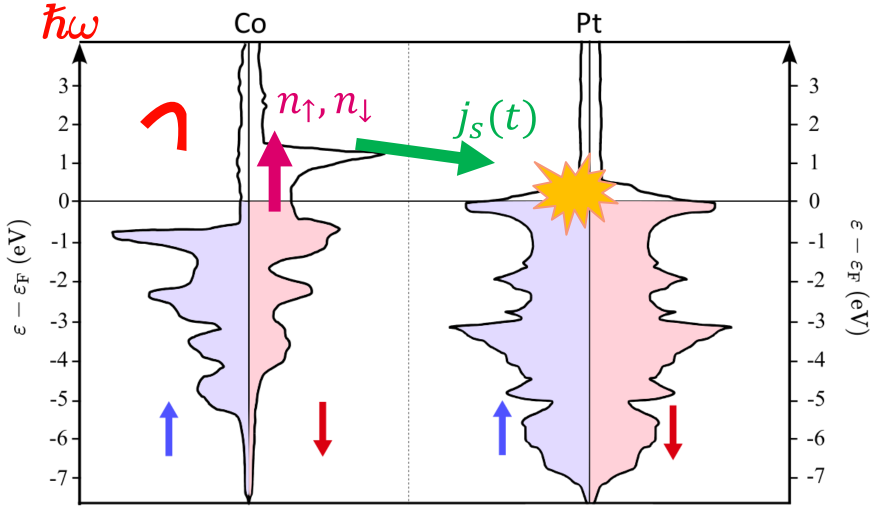
Probe new interconversion mechanisms and THz functionalities

Outlook of the 3*d*/5*d* THz emitter optimization

T.H. Dang, J. Hawecker, ER *et al.*, Appl. Phys. Rev. 7, 041409 (2020)

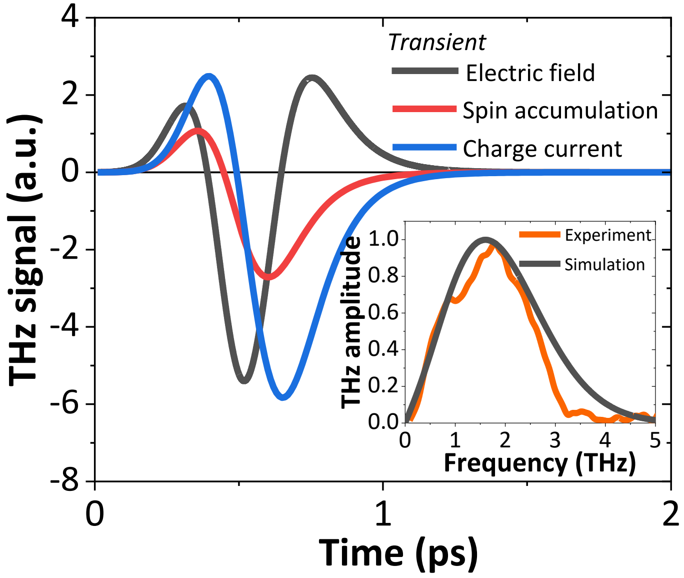
Wave-diffusion model: an alternative to the superdiffusive model

Finite-difference time-domain method



- Spin-dependent pumping $n_\uparrow \neq n_\downarrow$
- Hot electron spin diffusion $l_{sf} \simeq 2-3$ nm
- Spin relaxation $\theta_{SHE} \simeq 5\%$

Modelling of the dynamical SCC-based THz pulse



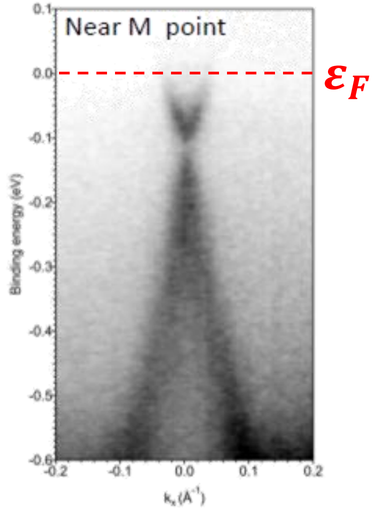
Dynamical spin-relaxation processes can be studied
Engineering of multilayers and interfaces

S. Kaltenborn *et al.*, Phys. Rev. B 85, 235101 (2012)
M. Battiato *et al.*, Phys. Rev. B 86, 024404 (2012)
R. Rouzegar *et al.*, Phys. Rev. B 106, 144427 (2022)
Density of states extracted from D.A. Papaconstantopoulos, “Handbook of the Band Structure of Elemental Solids”, Springer (2015)

THz azimuthal conversion profile

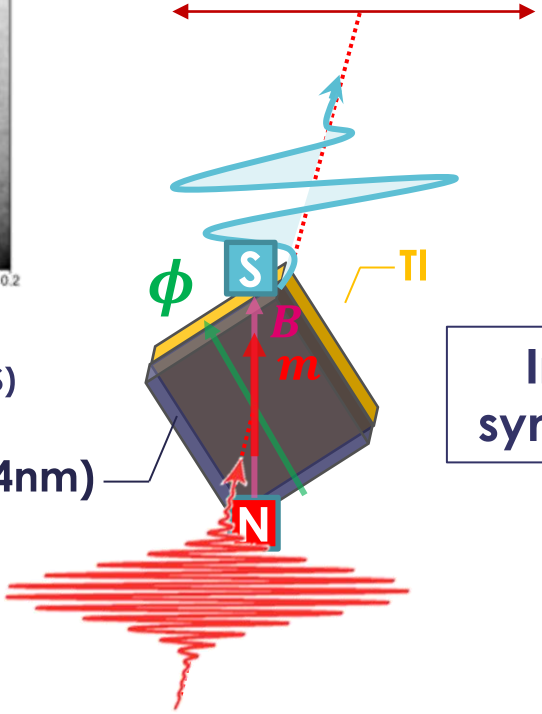
Angular resolved photoemission spectroscopy (ARPES)

ER *et al.*, Adv. Optical Mater., 10, 2102061 (2022)



Topological surface states (TSS)

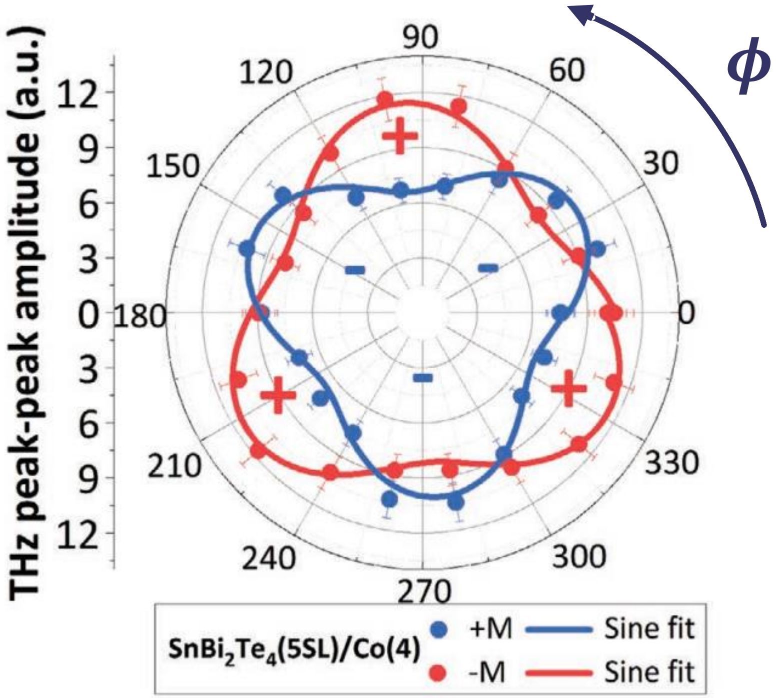
How to discriminate conversion mechanisms in TI/Co from THz emission?



Femtosecond NIR pump

Interconversion symmetry mapping

Azimuthal crystalline dependence



$SnBi_2Te_4/Co$

Complex THz emission azimuthal dependence → where is the spin-charge process?

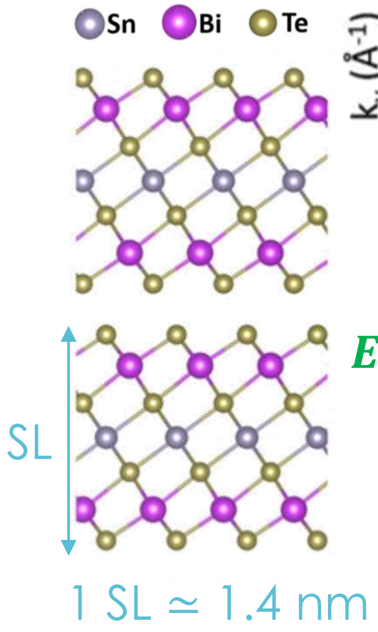
Investigation of $SnBi_2Te_4$ and $Bi_{1-x}Sb_x$ topological insulators

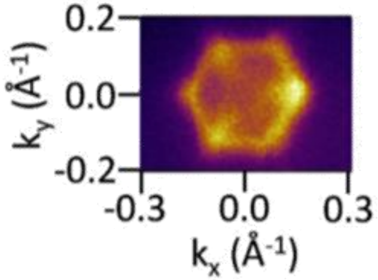
S. Fragkos *et al.*, Phys. Rev. Materials, 5, 014203 (2021)

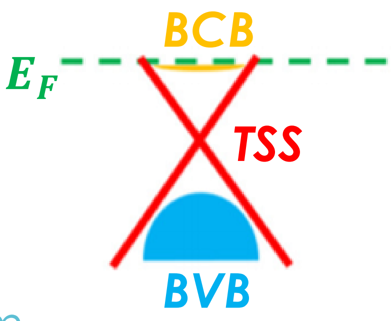
H. Benia *et al.*, Phys. Rev. B, 91, 161406(R) (2015)

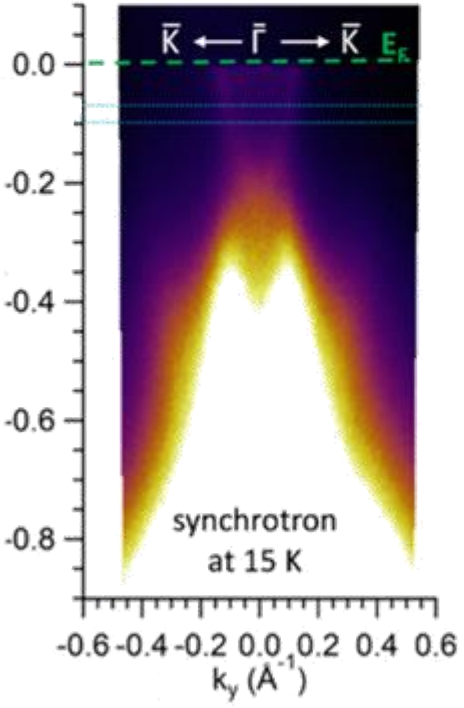
$SnBi_2Te_4$

Band structure pinning at Fermi level ∝ Bi/Sn ratio





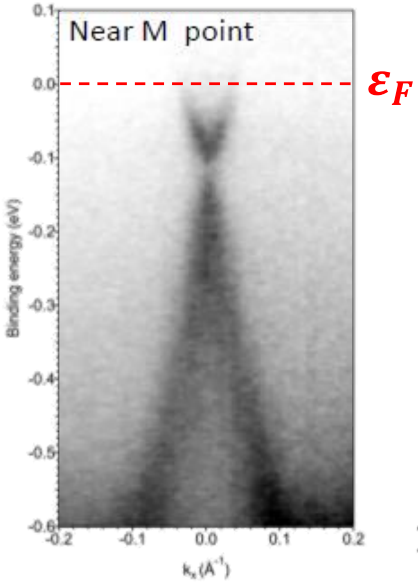




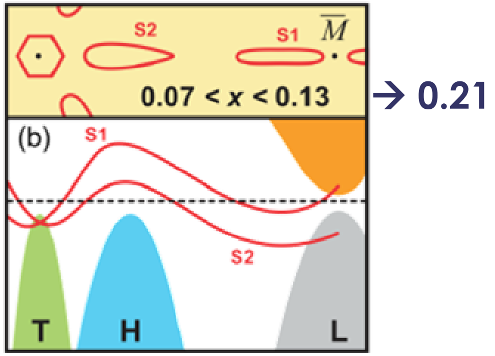
MBE growth of $SnBi_2Te_4$ made by S. Fragkos, A. Dimoulas team - INN (Greece)

$Bi_{1-x}Sb_x$

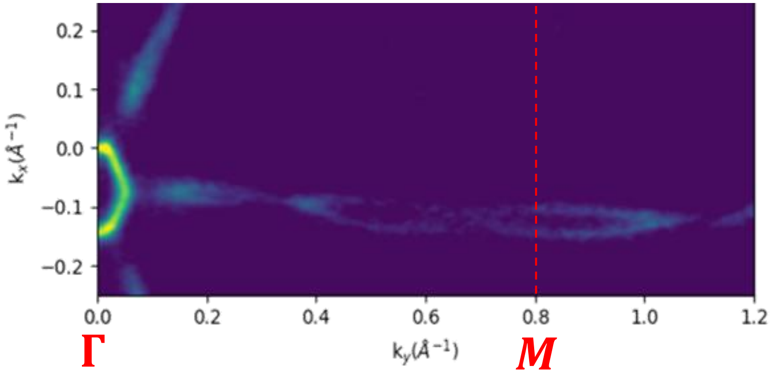
Angular Resolved Photo-Emission Spectroscopy (ARPES)



$Bi_{0.79}Sb_{0.21}$(15nm)



Experimental characterization of the TSS



MBE growth of BiSb made by L. Baringthon, P. Lefèvre team - SOLEIL (France)

Investigation of dynamical TI interconversion properties → THz emission spectroscopy

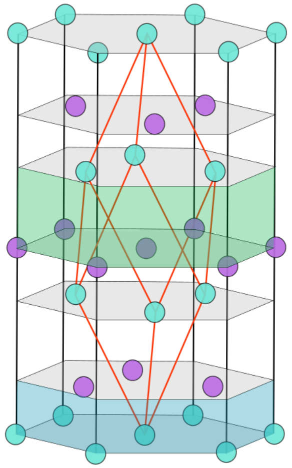
Investigation of $Bi_{1-x}Sb_x$ topological insulator

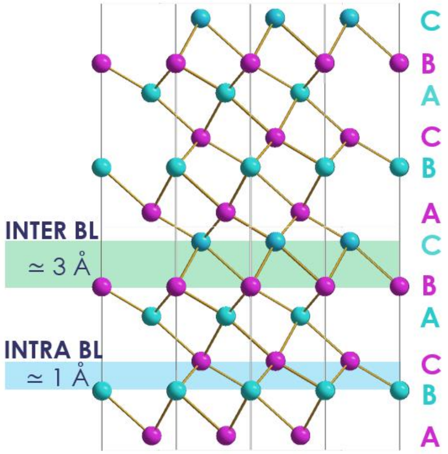
L. Baringthon *et al.*, Phys. Rev. Materials 6, 074204 (2022)

MBE growth of $Bi_{1-x}Sb_x$: L. Baringthon, P. Le Fèvre's team - SOLEIL (France)

B. Lenoir et al., Semiconductors and Semimetals, Elsevier, 69:101–37 (2001)
H. Benia *et al.*, Phys. Rev. B, 91, 161406(R) (2015)
N. H. D. Khang et al., Nature Materials 17, 9, 808–13 (2018)

A topological insulator with stoichiometric control



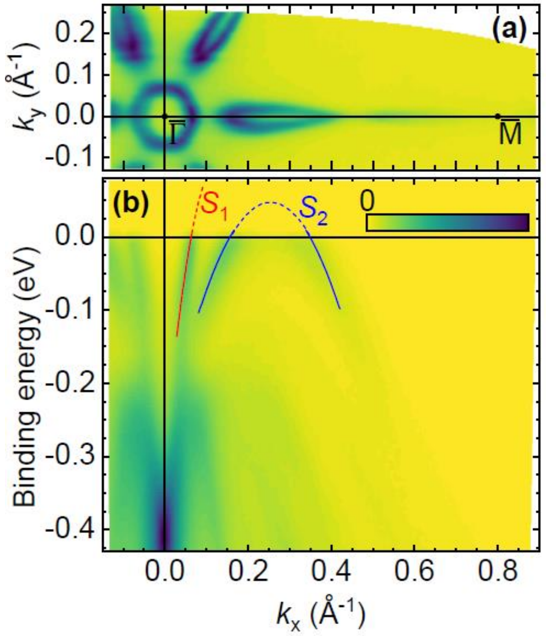


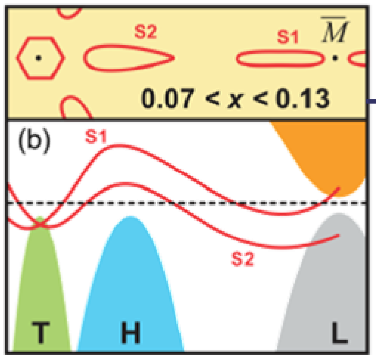
Hexagonal unit cell, with a bilayer structure (1 BL = 0.4 nm)

Stoichiometric changes

→ Tuning the inter (and intra) layers distances (strain)
→ Modification of the hopping terms between neighbours
→ Accounting in TB model (empirical Rashba surface terms)

TSS mapping and strong SCC efficiency





→ **0.21**

Experimental characterization of the TSS

$Bi_{0.85}Sb_{0.15}$(5nm)

$\rho_{\mathrm{BiSb}} \simeq 400$ µΩ.cm

$\theta_{\mathrm{SHE}}^{\mathrm{BiSb}} \simeq$ x52 (≡+5200%)

Angular Resolved Photo-Emission Spectroscopy (ARPES)

Sweep concentration across the topological window

Thickness variations: ultrathin $Bi_{1-x}Sb_x$ (2.5 nm) mastered at SOLEIL