Geometric aspects of quadratic optical responses

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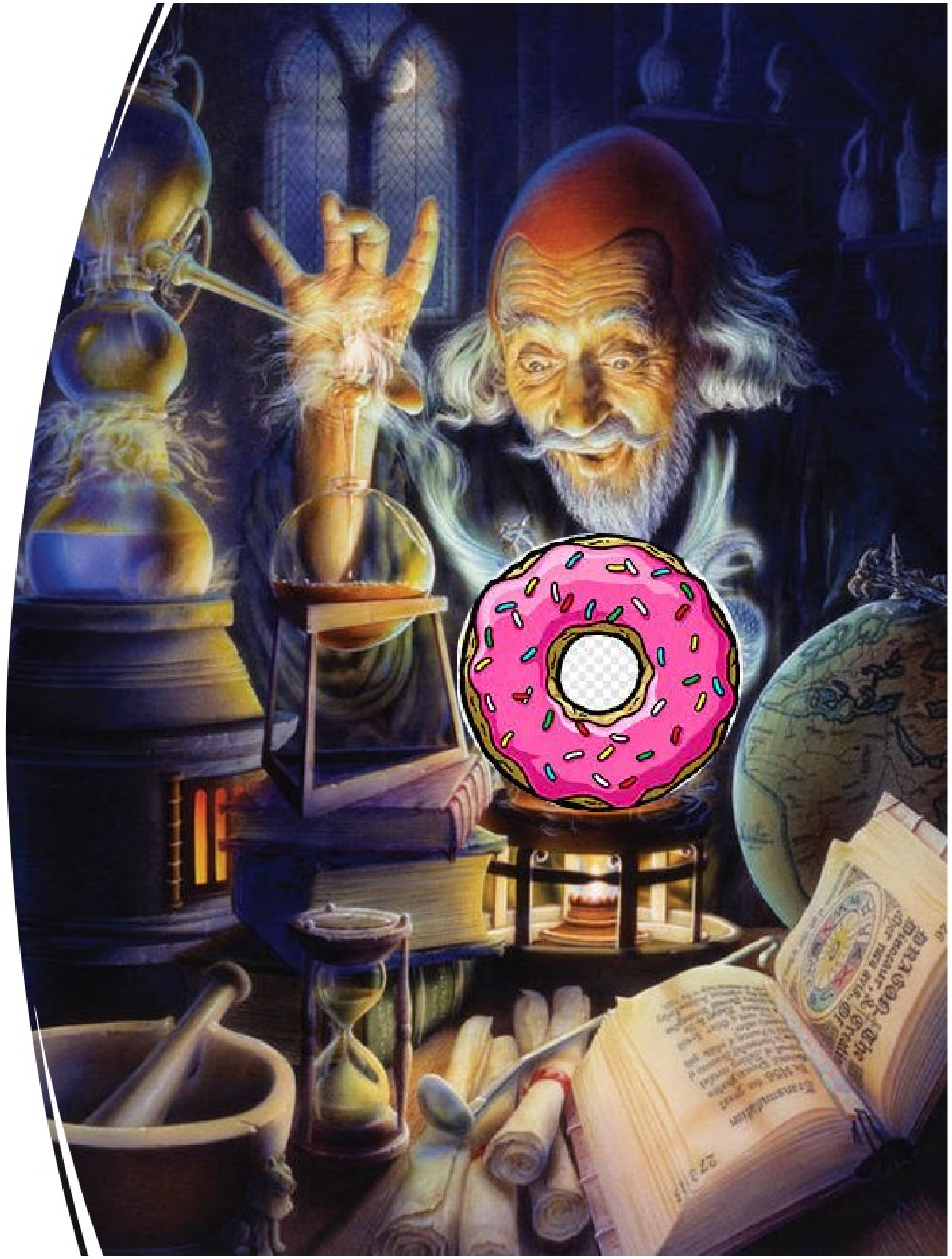
"Optical alchemy"

goes topological



Quadratic response to electric field

$$J_a = \sigma^{(2)}_{abc} \; \cdot \; E_b \cdot E_c$$

Quadratic response to electric field

$$\begin{array}{ccccc} - & 0 & & - & - \\ J_a = & \sigma^{(2)}_{abc} & \cdot & E_b & \cdot\, E_c \end{array}$$

- **Inversion** symmetry: $J_a \to -J_a$ $\qquad$ $E_a \to -E_a$

<u>Material needs to be acentric for finite quadratic absorption</u>

Quadratic response to electric field

$$J_a(\omega_\Sigma;\omega_1,\omega_2) = \sigma^{(2)}_{abc}(\omega_1,\omega_2) \ \cdot\ E_b(\omega_1)\cdot E_c(\omega_2)$$

$$\omega_\Sigma = \omega_1 + \omega_2$$

- **Inversion** symmetry: $J_a \rightarrow -J_a$ $\qquad E_a \rightarrow -E_a$

<u>Material needs to be acentric for finite quadratic absorption</u>

- Monochromatic wave $E(t) = E(\omega)e^{-i\omega t} + E(-\omega)e^{i\omega t}$

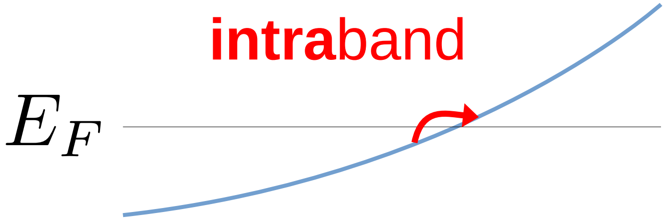
$$E(\omega_1)\cdot E(\omega_2) \rightarrow J(\underline{0};\omega,-\omega) \quad \text{and} \quad J(\underline{2\omega};\omega,+\omega)$$

<u>d.c. and second-harmonic responses</u>

Outline: three effects

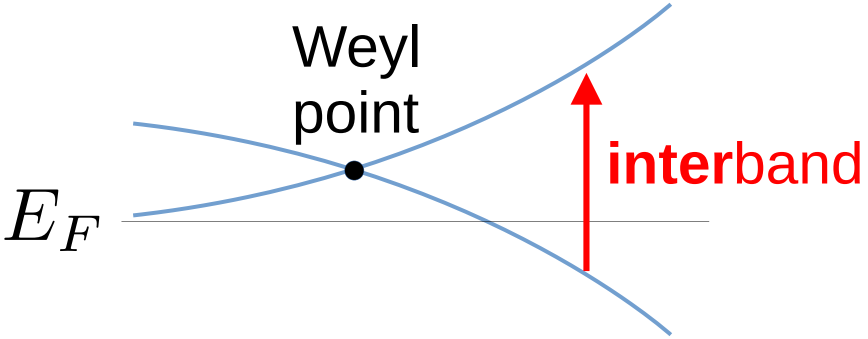
- Nonlinear Hall effect

$$J(0)\ \&\ J(2\omega)$$



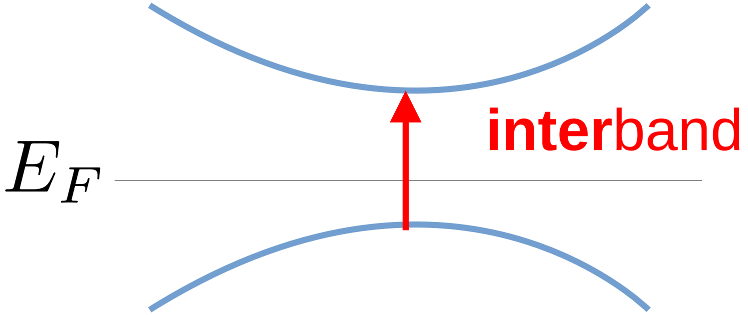
- Quantized circular photogalvanic effect

$$J(0)$$



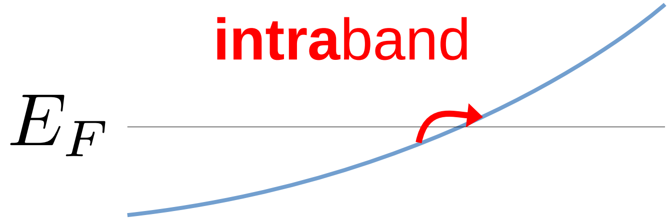
- Shift current across topological phase transition (TPT)

$$J(0)$$



Outline: three effects

- Nonlinear Hall effect



$$J(0)\ \&\ J(2\omega)$$

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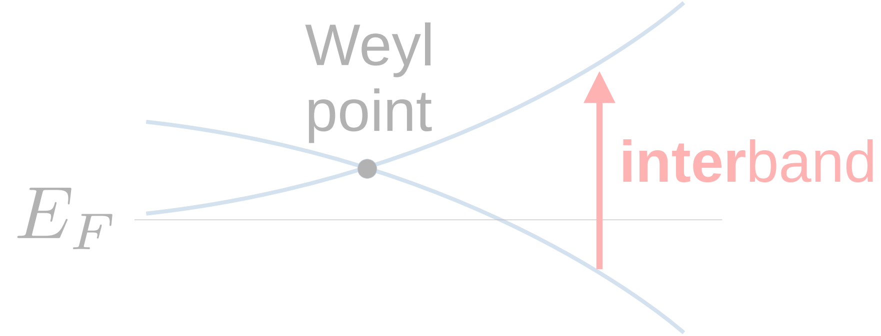
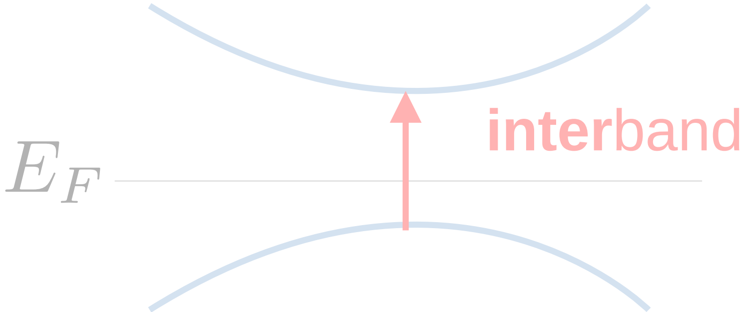


$$J(0)$$

- Shift current across topological phase transition (TPT)

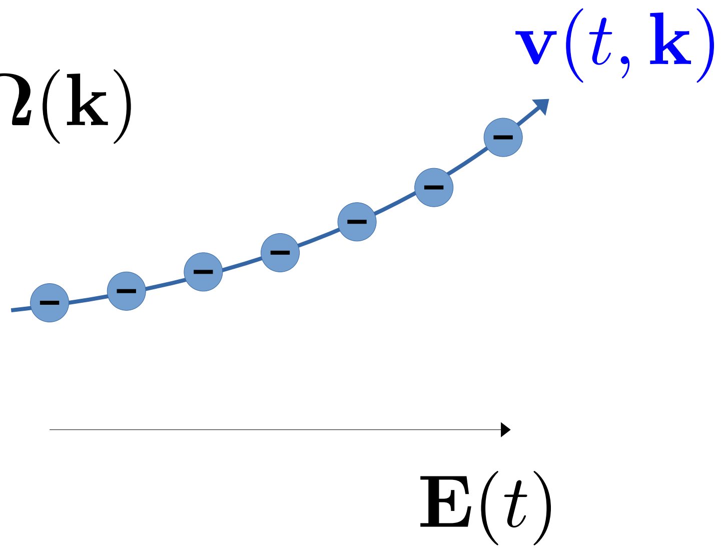
$$J(0)$$



1. Nonlinear Hall Effect

- Anomalous velocity in presence of an electric field

$$\mathbf{v}(t,\mathbf{k}) = \frac{1}{\hbar}\boldsymbol{\nabla}\epsilon(\mathbf{k}) - \frac{e}{\hbar}\mathbf{E}(t) \times \boldsymbol{\Omega}(\mathbf{k})$$



- Directly influences current

$$\mathbf{J}(t) = -e\int_k f(t,\mathbf{k})\mathbf{v}(t,\mathbf{k})$$

Occupation factor

1. Nonlinear Hall Effect

Boltzmann equation in the relaxation time approximation

$$-e\tau E_a \partial_{k_a} f + \tau \partial_t f = f_0 - f$$

Solve at different powers in the electric field

Sodemann and Fu, PRL **115**, 216806 (2015)

$$-ez\,E_a(t)\cdot\partial_a f + \tau\,\partial_t f = f_0 - f$$

$$E_a(t) = \mathrm{Re}\{E_a\, e^{i\omega t}\}$$

$$f = \mathrm{Re}\{f_0 + f_1 + f_2\}$$ Seek expansion in powers of E

f_0 : equilibrium; $\quad f_1 = f_1^{\omega} e^{i\omega t}$; $\quad f_2 = f_2^{0} + f_2^{2\omega} e^{2i\omega t}$

At linear order:

$$-ez\cdot\mathrm{Re}\{E_a e^{i\omega t}\}\,\partial_a f_0 + z\,\mathrm{Re}\{i\omega\tau\cdot f_1^{\omega} e^{i\omega t}\} = -\mathrm{Re}\left\{f_1^{\omega} e^{i\omega t}\right\}$$

$$-ezE_a\cdot\partial_a f_0 - i\tau\omega\, f_1^{\omega} = -f_1^{\omega} \Rightarrow \boxed{f_1^{\omega} = \frac{e\tau E_a \partial_a f_0}{1 + i\omega\tau}}$$

$$J_a = -e\int f\cdot v_a\, dk = -e\int f\cdot\Big(\underbrace{\hbar^{-1}\partial_a \mathcal{E}(k)}_{\text{no E-field}} - \frac{e}{\hbar}\cdot\underbrace{\varepsilon_{abc}\, E_c(t)\cdot\Omega_b}_{\text{linear in } E}\Big)\, dk$$

Transverse piece at linear order:

$$J^{\omega}_{1,a} = \frac{e^2}{\hbar} E_c\, \varepsilon_{abc}\int f_0\cdot\Omega_b\, dk \qquad \text{AHC}$$

Transverse piece at second order: NHE

$$J^{0,2\omega}_{2,a} = E_b\cdot E_c^{(*)}\cdot \frac{e^3\tau}{\hbar^2(1+i\omega\tau)}\,\varepsilon_{adc}\int \partial_b f_0\cdot\Omega_d\, dk.$$

Berry curvature dipole $\quad -\int f_0\cdot\partial_b\Omega_d\, dk$

1. Nonlinear Hall Effect

Quantum Anomalous Hall Effect	Nonlinear Hall Effect
$\sigma_{ab}^{(1)} = -\frac{e^2}{\hbar}\varepsilon_{abc}\int_k f_0(k)\Omega_c(k)$	$\overline{\sigma}_{abc}^{(2)} = -\varepsilon_{adc}\frac{e^3\tau}{2\hbar^2(1+i\omega\tau)}\int_k f_0(k)\partial_{k_b}\Omega_d(k)$
Finite in insulators and metals	Finite only in metals
Quantized in insulators	Not quantized and depends on scattering time (becomes independent for $\omega\tau \gg 1$)
Vanishes if Time-Reversal symmetry present	Vanishes if Inversion symmetry present
Cr and V doped $(Bi,Sb)_2Te_3$ thin films. Metallic ferromagnets like Fe.	TaAs (type I) and WTe_2 (type II) Weyl semimetal families

1. Nonlinear Hall Effect

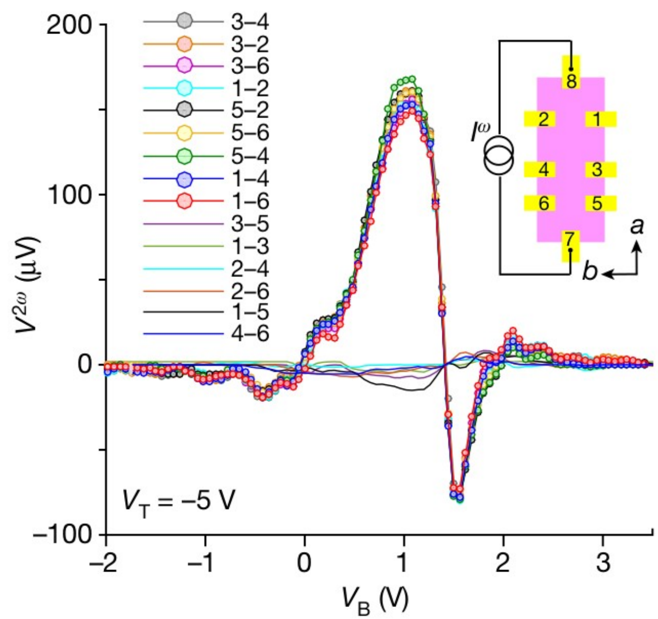
Experimentally measurable effect. More tomorrow in Q. Ma's lecture.

LETTER

https://doi.org/10.1038/s41586-018-0807-6

Observation of the nonlinear Hall effect under time-reversal-symmetric conditions

Qiong Ma[1,13], Su-Yang Xu[1,13], Huitao Shen[1,13], David MacNeill[1], Valla Fatemi[1], Tay-Rong Chang[2], Andrés M. Mier Valdivia[1], Sanfeng Wu[1], Zongzheng Du[3,4,5], Chuang-Han Hsu[6,7], Shiang Fang[8], Quinn D. Gibson[9], Kenji Watanabe[10], Takashi Taniguchi[10], Robert J. Cava[9], Efthimios Kaxiras[8,11], Hai-Zhou Lu[3,4], Hsin Lin[12], Liang Fu[1], Nuh Gedik[1]* & Pablo Jarillo-Herrero[1]*



Type II Weyl semimetal WTe_2

Literature

- Theory

Sodemann and Fu, PRL **115**, 216806 (2015)
Moore and Orenstein, PRL 105, 026805 (2010)
Deyo, Golub, Ivchenko and Spivak, arxiv (2009), 0904.1917
de Juan, Zhang, Morimoto, Sun, Moore and Grushin, PRR 2, 012017(R) (2020)
Gao, Addison, Mele and Rappe, PRR 3, L042032 (2021)

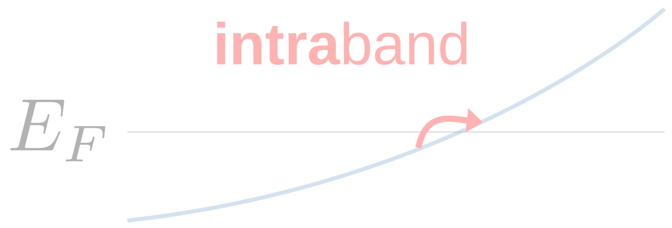
- Ab-initio calculations and experiments

Zhang, Sun and Yan, PRB 97, 041101 (2018)
Ma et. al., Nature 565, 337 (2019)
Min et. al., Nat. Comm. 14, 364 (2023)

2. Quantized CPGE

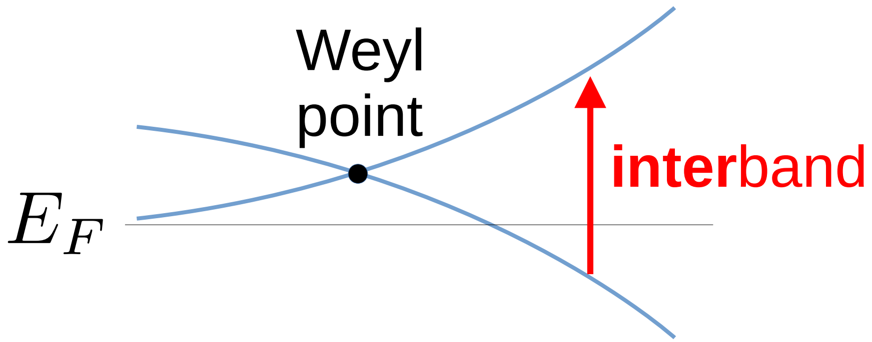
- Nonlinear Hall effect

$$J(0) \;\&\; J(2\omega)$$



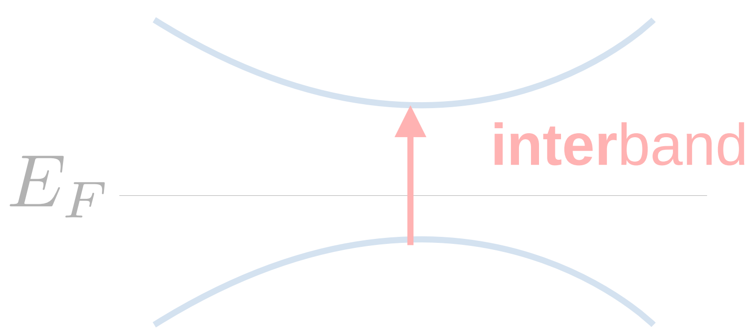
- Quantized circular photogalvanic effect

$$J(0)$$



- Shift current across topological phase transition (TPT)

$$J(0)$$



2. Quantized CPGE

- E-field couples to **position** operator (length gauge)

$$\hat{H} = \hat{H}_0 + e\mathbf{E}(t) \cdot \hat{\mathbf{r}}$$

- Linear order: usual Fermi's Golden rule

$$J_a(\omega) = \sigma^{(1)}_{ab}(\omega) \cdot E_b(\omega)$$

$$\sigma^{(1)}_{ab}(\omega) = \frac{e^2}{\hbar} \int_k \underbrace{\sum_{n,m}}_{\text{Bands}} (f_n - f_m) \overbrace{r^a_{nm} r^b_{mn}}^{\text{Transition matrix elements}} \underbrace{\delta(\omega_{nm} - \omega)}_{\text{Energy conservation}}$$

2. Quantized CPGE

- E-field couples to **position** operator (length gauge)

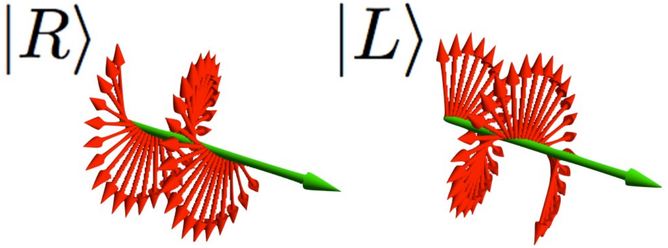
$$\hat{H} = \hat{H}_0 + e\mathbf{E}(t) \cdot \hat{\mathbf{r}}$$

- Second order d.c. “injection-current” Sipe and Shkrebtii, PRB (2000)

$$J_a(0;\omega,-\omega) = \tau \cdot \eta^{(2)}_{abc}(\omega) \cdot E_b(\omega)E_c(-\omega)$$

$$\eta^{(2)}_{abc}(\omega) = \frac{\pi|e^3|}{\hbar^2}\Big(\int_k \sum_{n,m} f_{nm} \frac{\partial \omega_{nm}}{\partial k_a} r^b_{nm} r^c_{mn} \delta(\omega_{nm} - \omega) - b \leftrightarrow c\Big)$$

The Circular PhotoGalvanic Effect is the part of the DC photocurrent that **switches** with the sense of **circular polarization**



$$\eta^{abc} = \rho^{abc} - \rho^{acb} \begin{cases} b=c \rightarrow \eta^{abb} = 0 \\ b \neq c \rightarrow \eta^{abc} = -\eta^{acb} \end{cases}$$

Circular pol.: $\vec{E}^{\pm}(\omega) = E_0 (\hat{x} \pm i \hat{y})$; $E_x = E_0$ $E_y = \pm i E_0$

$$J_a^{\pm} = \sum_{bc}^{xy} \eta_{abc} E_b(\omega) \overbrace{E_c(-\omega)}^{E_c^*(\omega)} =$$

$$= \overbrace{\eta_{axx}}^{0} |E_x|^2 + \overbrace{\eta_{ayy}}^{0} |E_y|^2 + \eta_{axy} E_0 \cdot (\mp i E_0^*) + \eta_{ayx} (\pm i E_0) \cdot E_0^* =$$

$$= \mp 2i \, \eta_{axy} |E_0|^2 \Rightarrow$$

$$\eta_{axy} = -\eta_{ayx}$$

$$\boxed{\begin{gathered} J_a^{+} = -J_a^{-} \Downarrow \\ \text{CPGE: } J_a^{+} - J_a^{-} \neq 0 \end{gathered}}$$

2. Quantized CPGE

Searching for quantization

Recall

$$r^a_{nm} \equiv i\langle u_{n\mathbf{k}}|\partial_{k_a} u_{m\mathbf{k}}\rangle$$

$$\Omega^z_n = i\sum_{n\neq m}\left(r^x_{nm}r^y_{mn} - r^y_{nm}r^x_{mn}\right)$$

$$\eta^{(2)}_{zxy}(\omega) \propto \int_k \sum_{n,m} f_{nm}\frac{\partial\omega_{nm}}{\partial k_z}\left(r^x_{nm}r^y_{mn} - r^y_{nm}r^x_{mn}\right)\delta(\omega_{nm}-\omega)$$

2. Quantized CPGE

Searching for quantization

- General multiband case; cannot recast transition matrix element using expression for Berry curvature ☹

$$\Omega_n^z = i \sum_{n \neq m} \left(r_{nm}^x r_{mn}^y - r_{nm}^y r_{mn}^x \right)$$

$$\eta_{zxy}^{(2)}(\omega) \propto \int_k \sum_{n,m} f_{nm} \frac{\partial \omega_{nm}}{\partial k_z} \left(r_{nm}^x r_{mn}^y - r_{nm}^y r_{mn}^x \right) \delta(\omega_{nm} - \omega)$$

2. Quantized CPGE

Searching for quantization

- General multiband case; cannot recast transition matrix element using expression for Berry curvature

- **Two-band** approximation: $n = 1, m = 2$

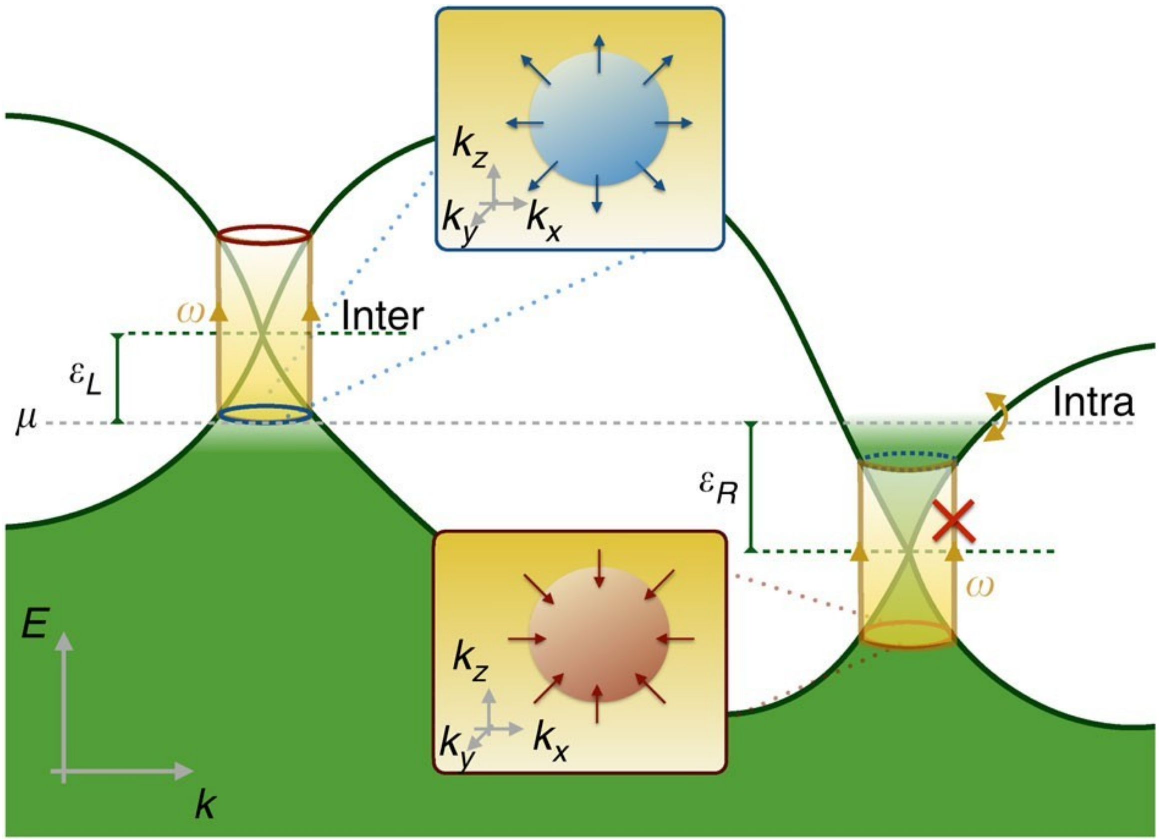
$$\Omega_1^z = i \sum_{n \neq m} \left(r_{12}^x r_{21}^y - r_{12}^y r_{21}^x \right) = -\Omega_2^z$$

$$\eta^{(2)}_{zxy}(\omega) \propto \int_k \sum_{n,m} f_{12} \frac{\partial \omega_{12}}{\partial k_z} \underbrace{\left(r_{12}^x r_{21}^y - r_{12}^y r_{21}^x \right)}_{\Omega_1^z} \delta(\omega_{12} - \omega)$$

Berry curvature in transition matrix element

Ideal case: Weyl semimetal with nodes at different energy

$$i\frac{e^3}{\hbar^2}\int_k f_{12}\frac{\partial\omega_{12}}{\partial k_a}\cdot\Omega^a\cdot\delta(\omega_{12}-\omega)$$



de Juan, Grushin, Morimoto and Moore, Nat. Comm. 8, 15995 (2017)

2. Quantized CPGE

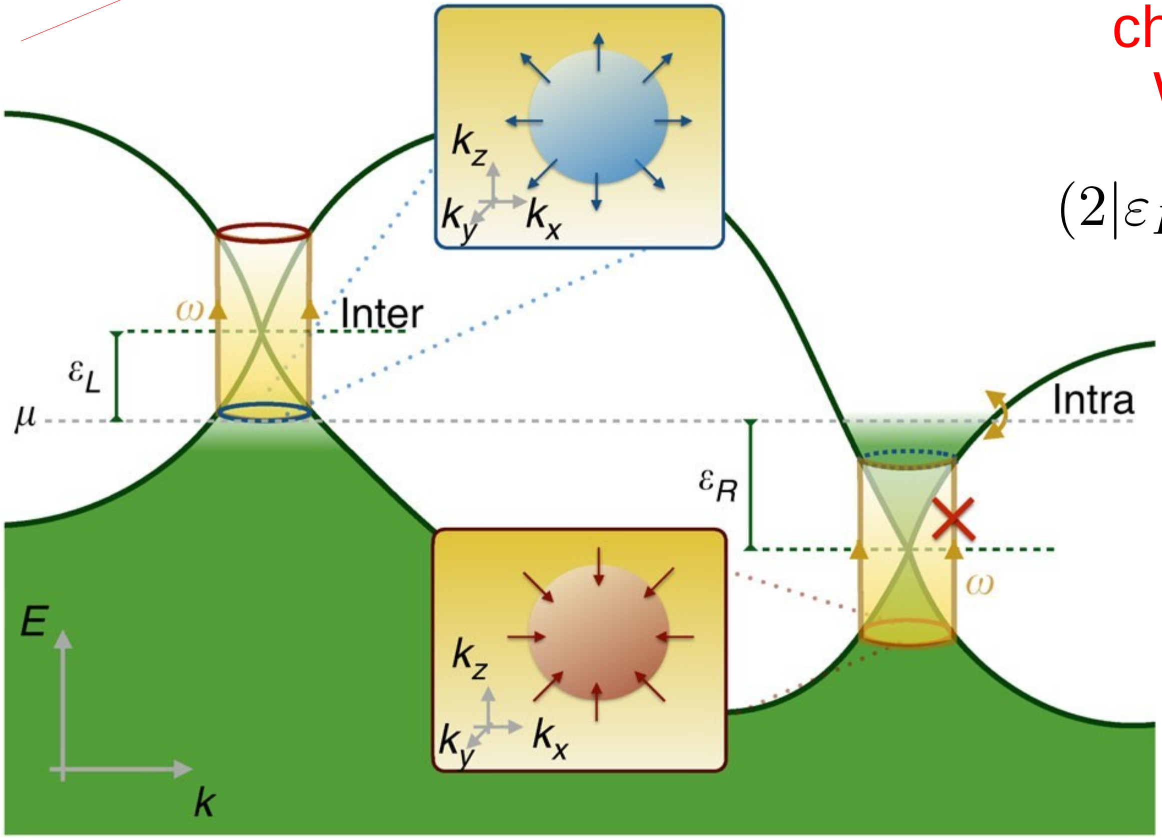
Ideal case: Weyl semimetal with nodes at different energy

$$i\frac{e^3}{\hbar^2}\oint_S dS\cdot\Omega^a = i\pi\frac{e^3}{\hbar^2}C_L$$

Surface enclosing Weyl node

Topological charge of Left Weyl node

$$(2|\varepsilon_L| < \hbar\omega < 2|\varepsilon_R|)$$



de Juan, Grushin, Morimoto and Moore, Nat. Comm. 8, 15995 (2017)

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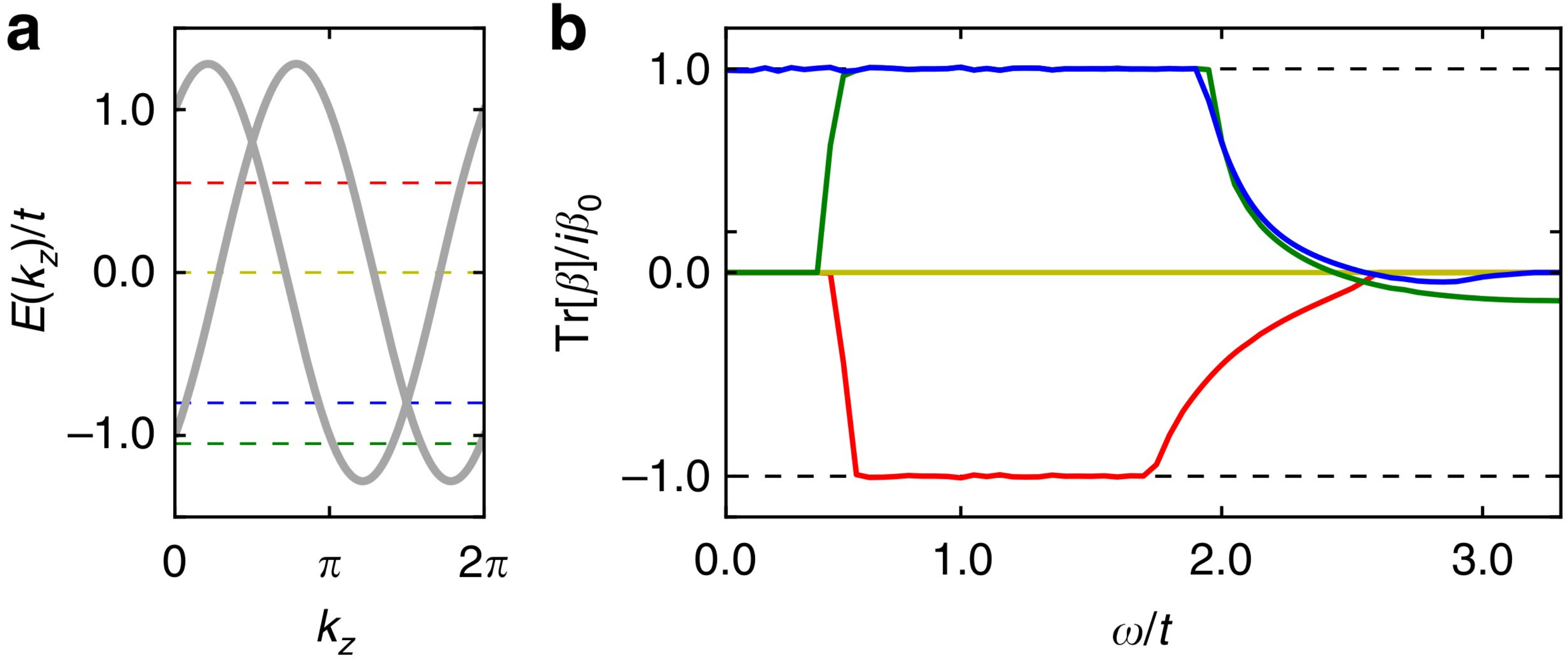

$$\beta_0 = \pi\frac{e^3}{\hbar^2}C_L$$

de Juan, Grushin, Morimoto and Moore, Nat. Comm. 8, 15995 (2017)

2. Quantized CPGE

Ideal case: Weyl semimetal with nodes at different energy

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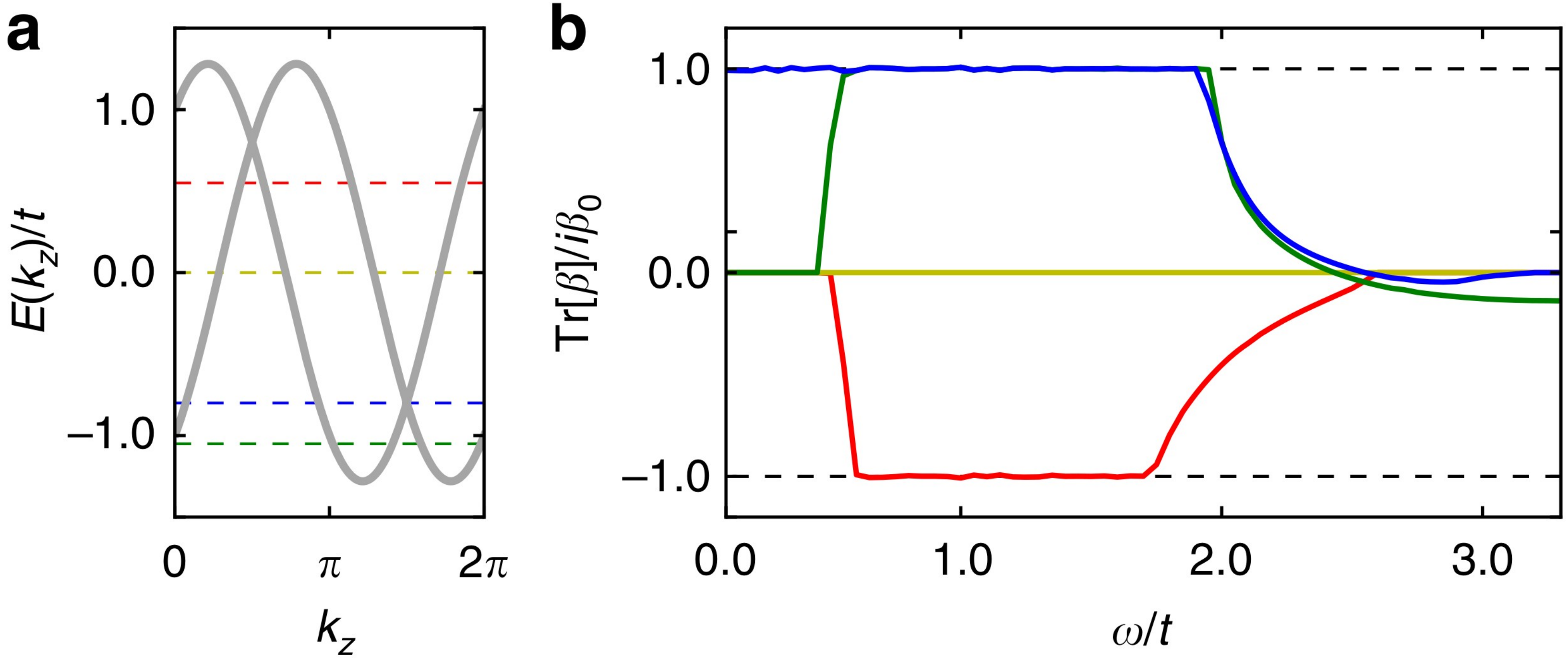


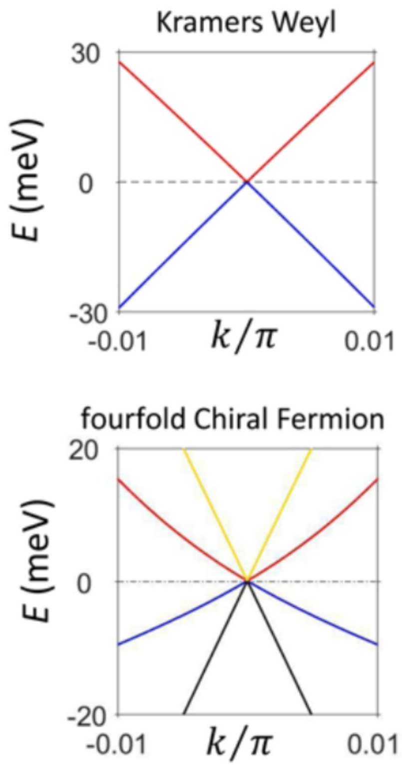
$$\beta_0 = \pi\frac{e^3}{\hbar^2}C_L$$

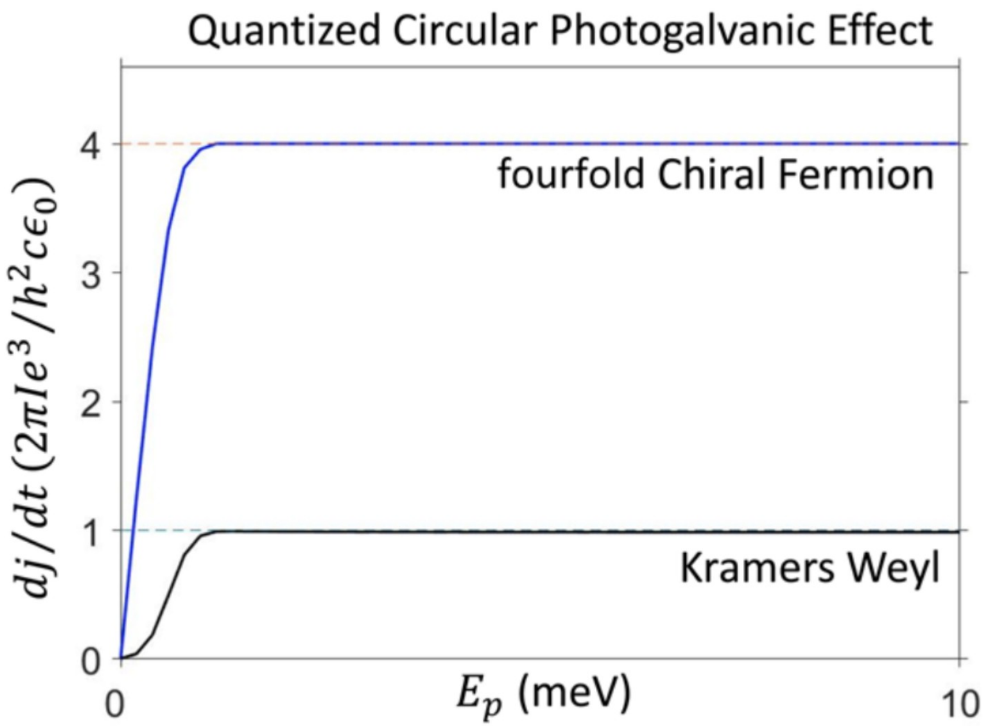
CPGE: $J_+ - J_- = \tau\cdot\frac{4I}{c\epsilon_0}\cdot\beta_0$

de Juan, Grushin, Morimoto and Moore, Nat. Comm. 8, 15995 (2017)

2. Quantized CPGE

- Generalization to multifold chiral Fermions, family of RhSi





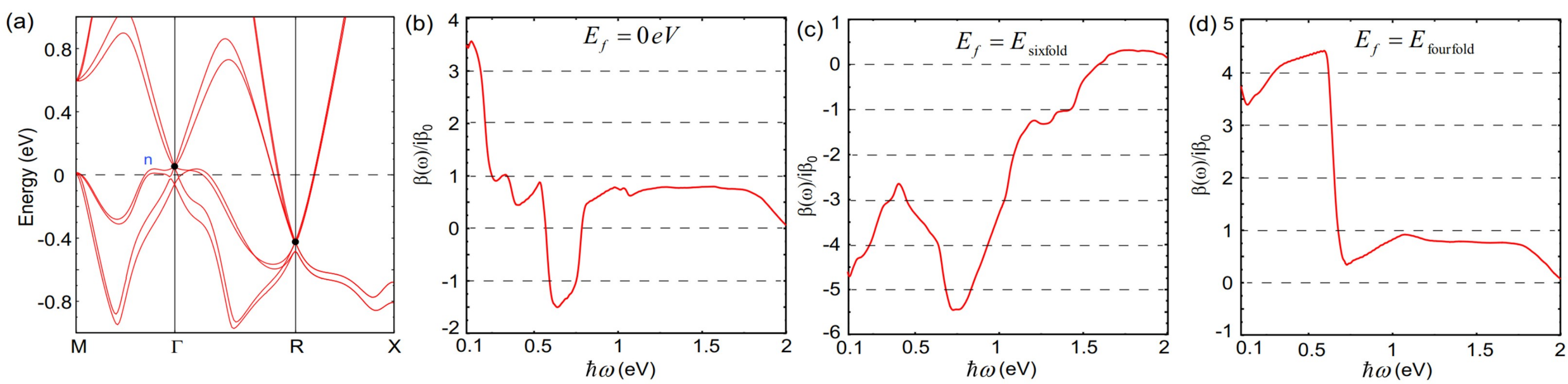
Chang et. al., PRL 119, 206401 (2017)

2. Quantized CPGE

Towards quantization in real materials. More tomorrow in Q. Ma's lecture.

- Ab-initio calculations on RhSi

Le, Zhang, Felser and Sun, PRB 102, 121111 (2020)



- Experiments

ARTICLE

Check for updates

https://doi.org/10.1038/s41467-020-20408-5 OPEN

Giant topological longitudinal circular photo-galvanic effect in the chiral multifold semimetal CoSi

Zhuoliang Ni[1], K. Wang[2], Y. Zhang[3,4], O. Pozo[5], B. Xu[6], X. Han[1], K. Manna[4,7], J. Paglione[2,8], C. Felser[4,8], A. G. Grushin[9], F. de Juan[10,11], E. J. Mele[1] & Liang Wu[1]✉

Literature

- Theory

  Two-band Weyl materials:

  de Juan, Grushin, Morimoto and Moore, Nat. Comm. 8, 15995 (2017)

  Multifold chiral fermions:

  Chang et. al., PRL 119, 206401 (2017)

  Flicker, de Juan, Bradlyn, Morimoto, Vergniory and Grushin PRB 98, 155145 (2018)

- Ab-initio calculations and experiments

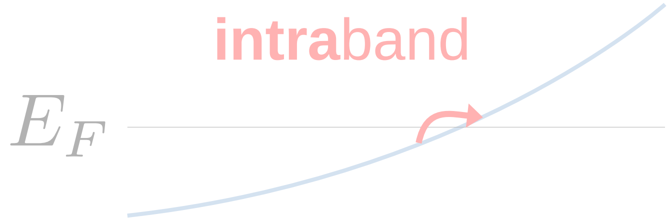
  Le, Zhang, Felser and Sun, PRB 102, 121111 (2020)

  Ni et. al., Nat. Comm. 12, 154 (2021)

3. Shift current across a TPT

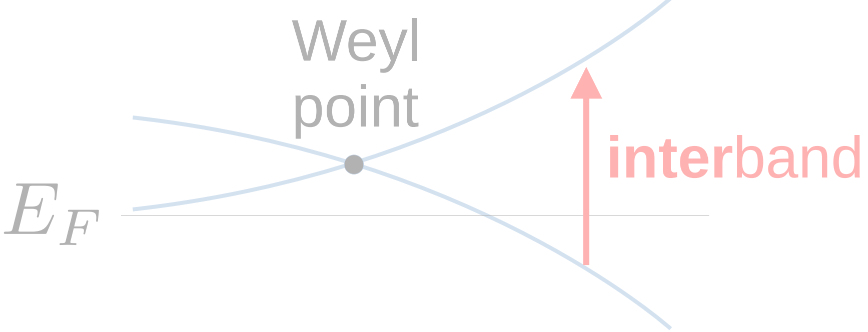
- Nonlinear Hall effect

$$J(0)\ \&\ J(2\omega)$$



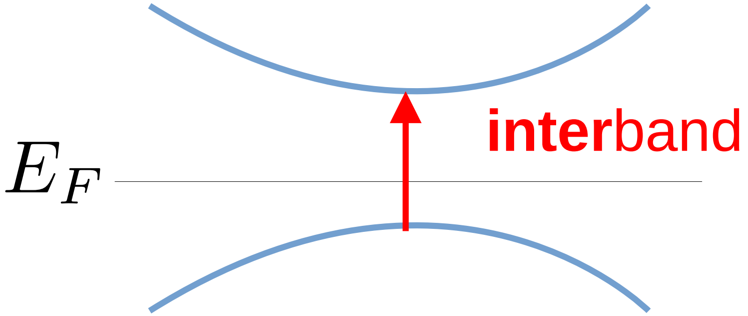
- Quantized circular photogalvanic effect

$$J(0)$$



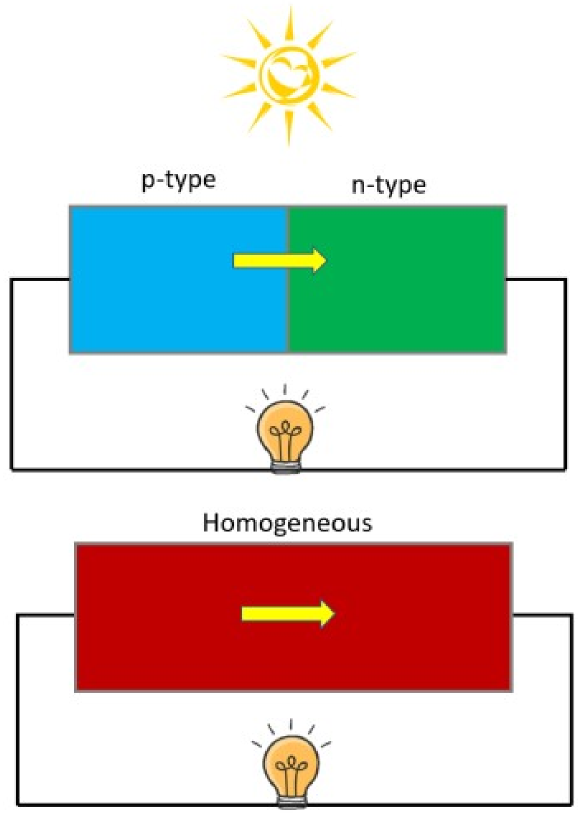
- Shift current across topological phase transition (TPT)

$$J(0)$$



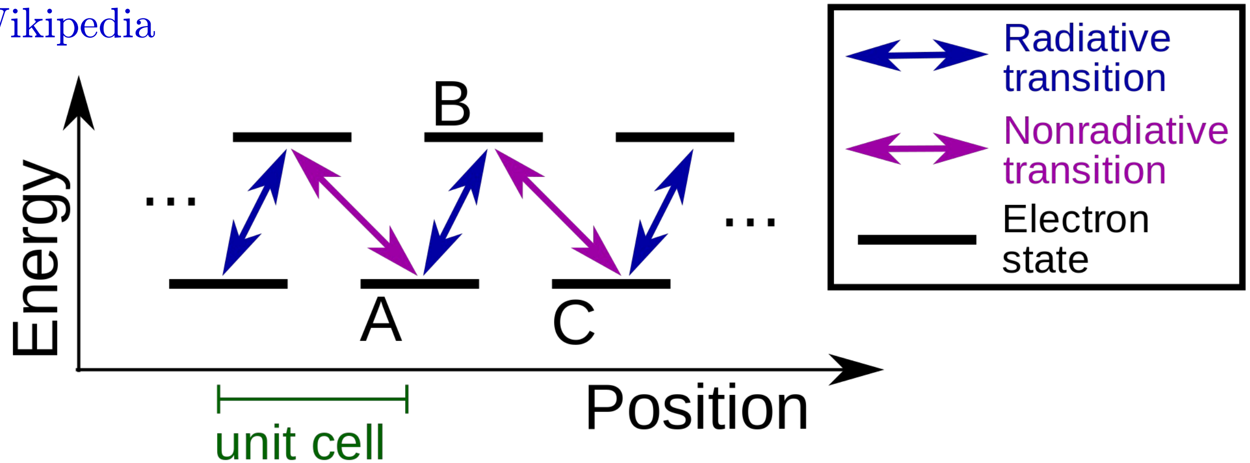
3. Shift current across a TPT

Part of the so-called *"Bulk photovoltaic effect"*



Dai, Zhenbang and Rappe, Chem. Phys. Rev. 4, 011303 (2023)

Wikipedia



Real-space centers of the bands are displaced or "shifted" upon photoexcitation

3. Shift current across a TPT

RESEARCH ARTICLE

OPTICS

Topological nature of nonlinear optical effects in solids

Takahiro Morimoto[1]* and Naoto Nagaosa[2,3]

Science Advances 2016

3. Shift current across a TPT

$$J_a(0;\omega,-\omega) = \sigma^{(2)}_{abc}(\omega)\cdot E_b(\omega)E_c(-\omega)$$

$$\sigma^{(2)}_{abc}(\omega) = \frac{\pi|e^3|}{4\hbar^2}\left(\int_k \sum_{n,m} f_{nm} R^a_{nm} r^b_{nm} r^c_{mn}\delta(\omega_{nm}-\omega) + b \leftrightarrow c\right)$$

WF phase: $r^a_{nm} = |r^a_{nm}|e^{-i\varphi_{nm}}$

Shift vector:
(length units)

$$R^a_{nm} = -\frac{\partial\varphi_{nm}}{\partial k_a} - \left(A^a_{nn} - A^a_{mm}\right)$$

Intraband Berry connections: $A^a_{nn} \equiv \langle u_{n\mathbf{k}}|\partial_{k_a} u_{n\mathbf{k}}\rangle$

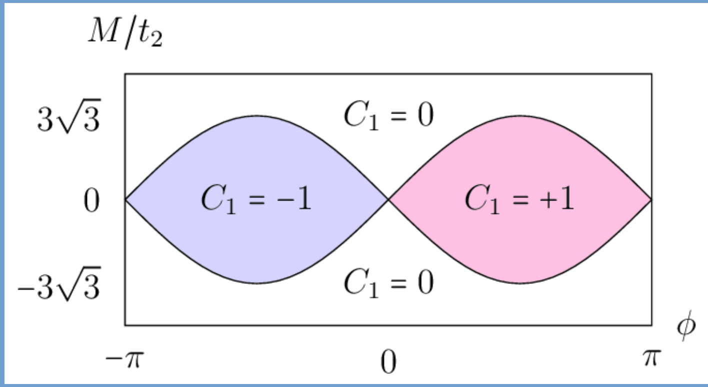
<u>Sensitive to the phase of Bloch states, geometry of the wavefunction</u>

3. Shift current across a TPT

$$J_a(0;\omega,-\omega) = \sigma^{(2)}_{abc}(\omega)\cdot E_b(\omega)E_c(-\omega)$$

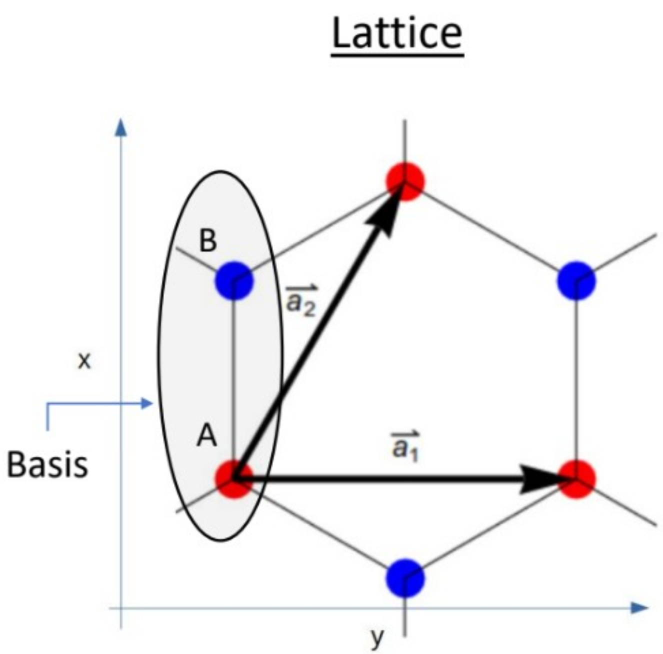
Haldane model

- Simple description of a topological phase diagram



- Simplified (analytical) two-band expression for the shift current

Haldane model: quick review

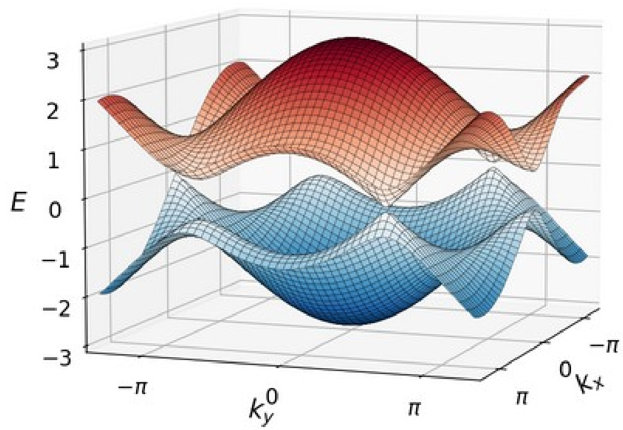


Breaks Inversion symmetry

Tight-binding parameters

- Onsite energies $\pm M$ +(-) for A(B)
- nn tunnelings t_1
- nnn tunnelings $t_2 e^{i\pm\phi}$ +(-) for A(B)→ A(B)

Breaks Time Reversal symmetry (TRS)

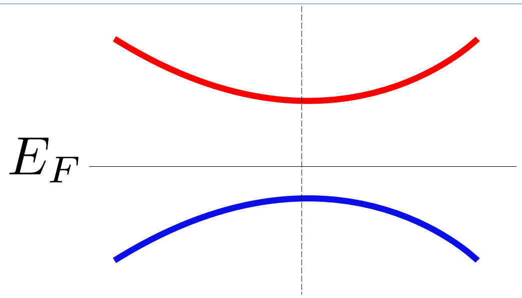


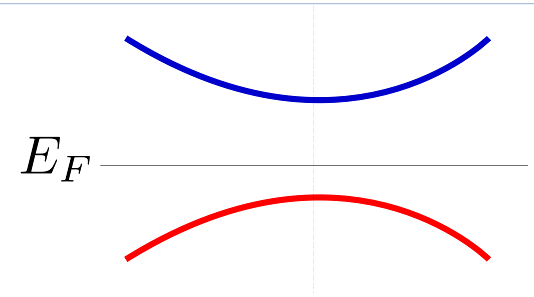
Band edge at the corner K or K′ of the Brillouin zone

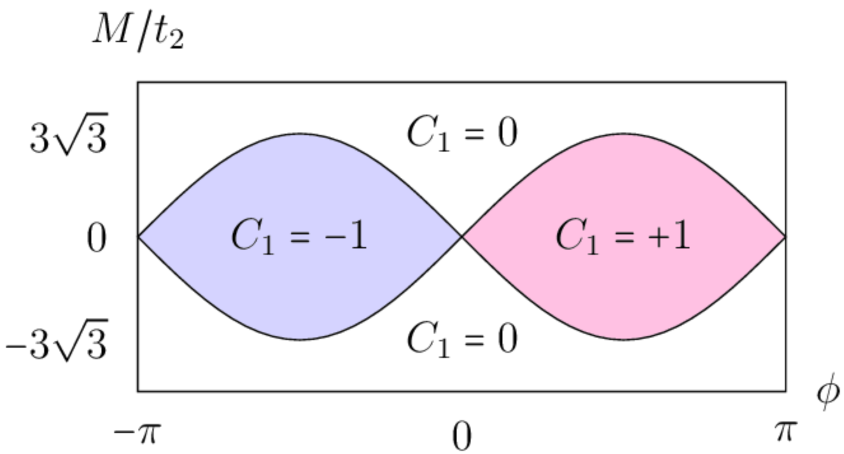
3. Shift current across a TPT

TPT driven by the **mass term:** $m = M + 3\sqrt{3}t_2 \sin\phi$

Trivial insulator (C=0)	Topological insulator (|C|=1)
$m > 0$	$m < 0$



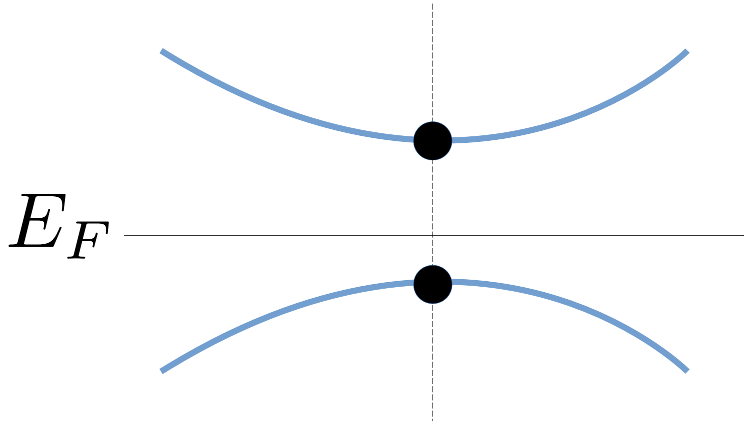




Fruchart and Carpentier, CRP (2013)

3. Shift current across a TPT

Description of the **band edge**:



- 2x2 Hamiltonian: $$H = \epsilon_0 \sigma_0 + \sum_i \sigma_i g_i$$

  with expansion coefficients: $$g_i = g_i(\mathcal{O}(k^2), m)$$

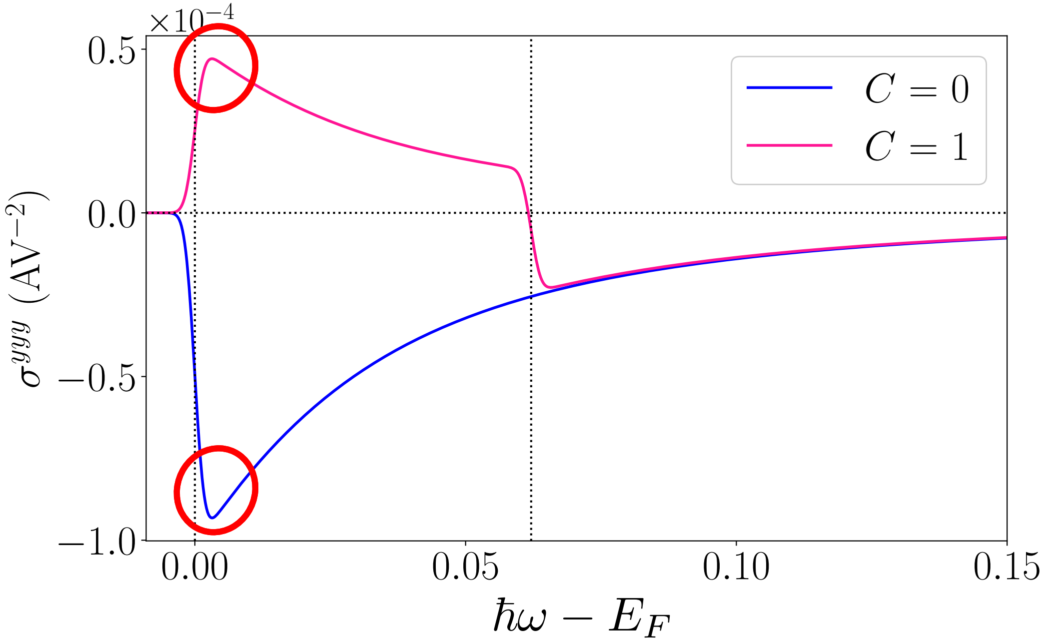
- Shift current: $$\sigma^{(2)}_{abb} \propto \int_k \sum_{ijm} \frac{1}{\omega_{12}} \left(g_m g_{i,b} g_{j,ab} \right) \epsilon_{ijm} \delta(\omega_{12} - \omega)$$

Cook, Fregoso, de Juan, Coh and Moore, Nat. Comm. (2017)

3. Shift current across a TPT

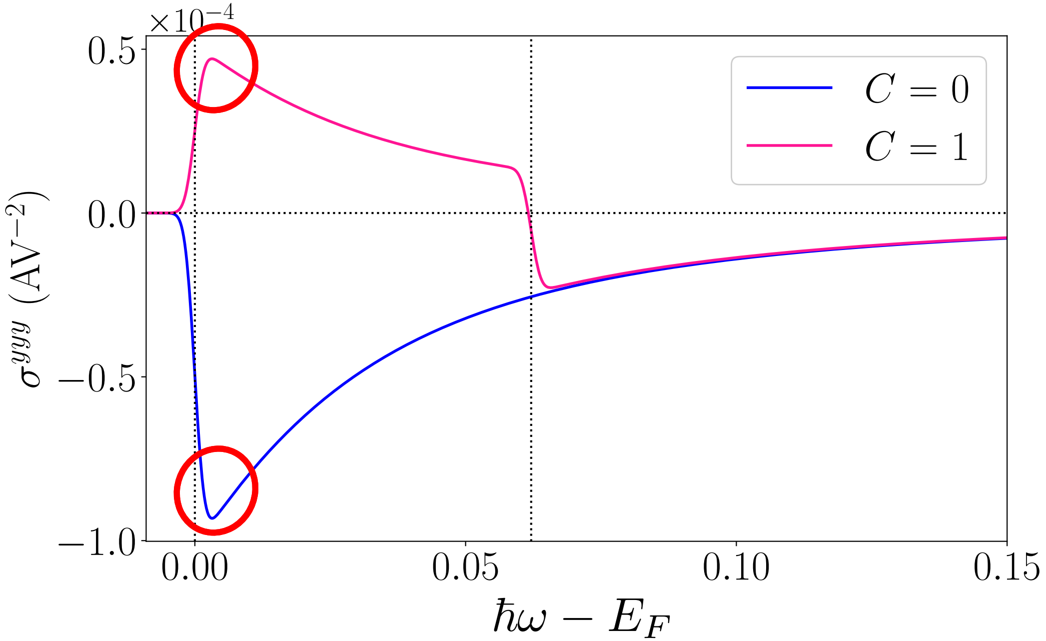
$$\sigma^{abb}(\omega) \propto \mathrm{sign}\,[m]$$

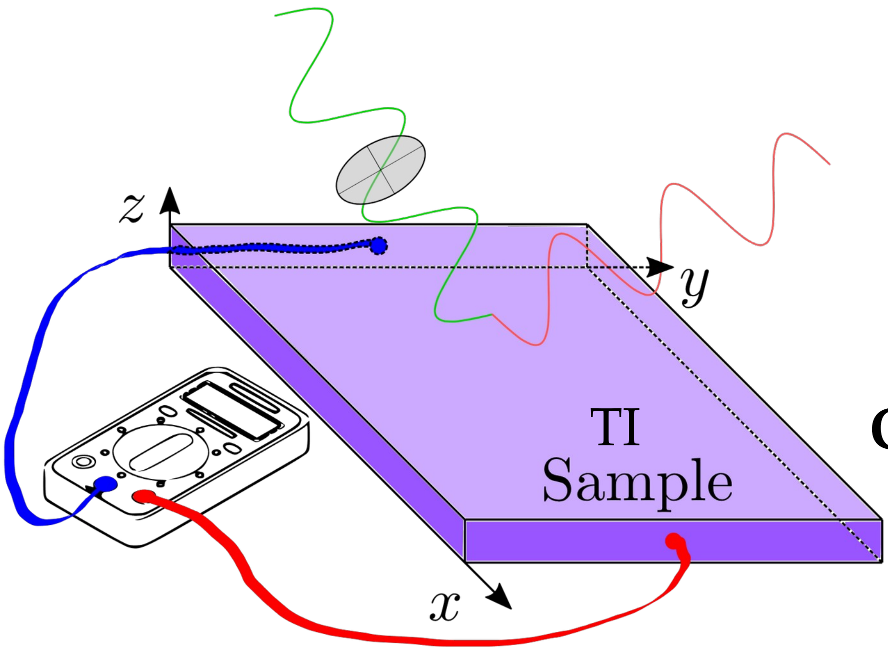
Band-edge shift photoconductivity switches sign across the TPT, driven by the mass term



$$\sigma^{abb}(\omega) \propto \text{sign}\,[m]$$

Band-edge shift photoconductivity switches sign across the TPT, driven by the mass term





$$\text{sign}[\mathbf{J}(C=0)] = -\text{sign}[\mathbf{J}(|C|=1)]$$

Optical determination of topological state!

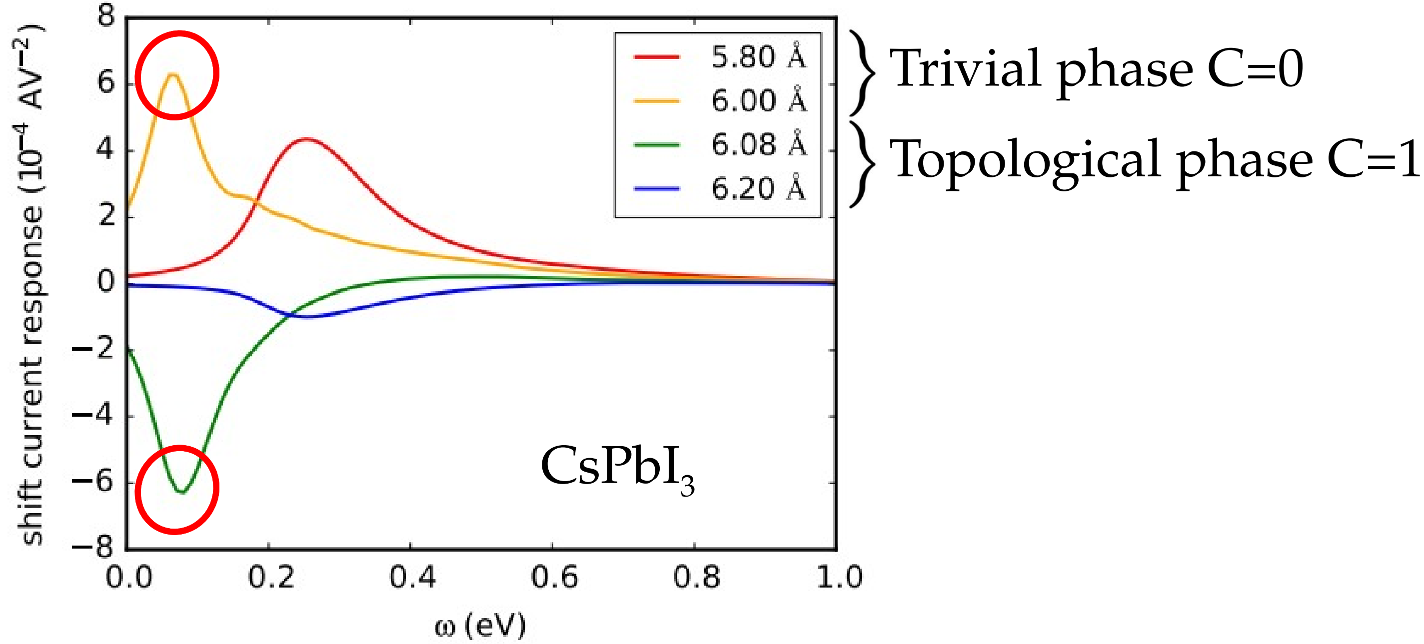
Ab-initio calculations

PRL **116,** 237402 (2016) PHYSICAL REVIEW LETTERS week ending 10 JUNE 2016

Enhancement of the Bulk Photovoltaic Effect in Topological Insulators

Liang Z. Tan and Andrew M. Rappe
The Makineni Theoretical Laboratories, Department of Chemistry, University of Pennsylvania, Philadelphia, Pennsylvania 19104-6323, USA
(Received 14 August 2015; published 8 June 2016)

TPT induced by **strain** (modified **lattice parameter**)



Literature

- Description of the effect

  Calculations on topological insulating materials:
  Tan and Rappe, PRL 116, 237402 (2016)

  Two-band model description:
  Z. Yan, arXiv:1812.02191 (2018)
  Sivianes and Ibañez-Azpiroz, arXiv:2305.17035 (2023)

- General theory

  von Baltz and Kraut, PRB 23, 10 (1981)
  Sipe and Shkrebtii, PRB 61, 5337 (2000)
  Fridkin Crystallogr. Rep. 46, 654 (2001)
  Morimoto and Nagaosa, Sci. Adv. 2, e1501524 (2016)

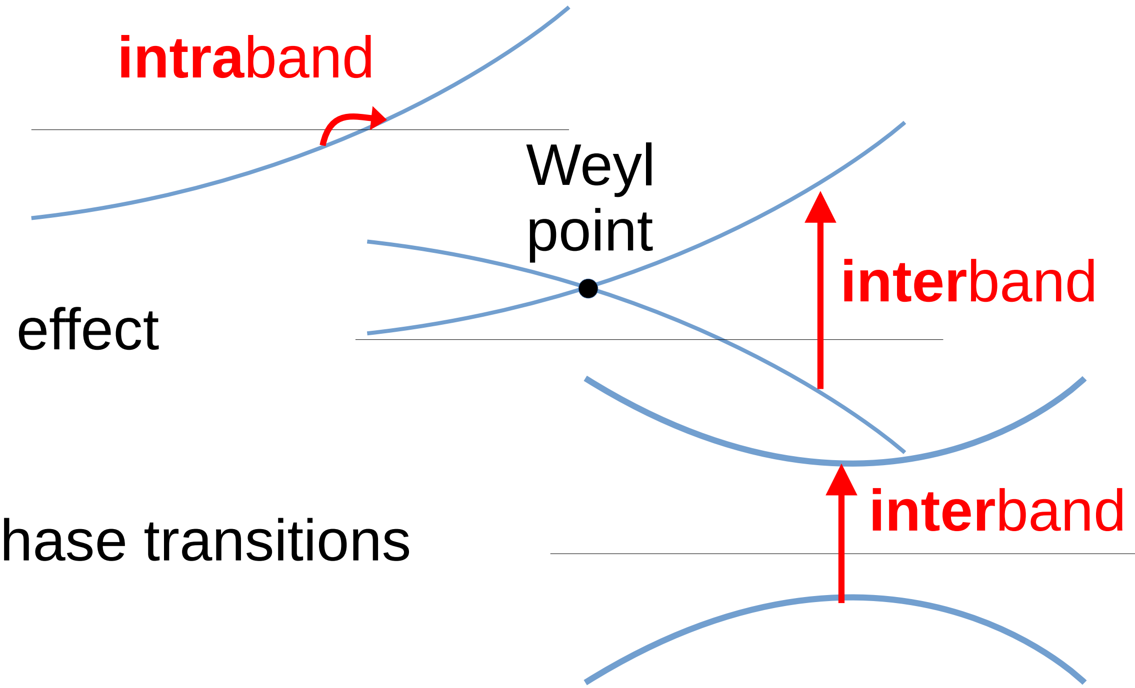
- Two-band tight-binding expression of the shift current

  Cook, Fregoso, de Juan, Coh and Moore, Nat. Comm. 8, 14176 (2017)

This afternoon tutorial by S. Tsirkin

WANNIER BERRI

- Nonlinear Hall effect
- Quantized circular photogalvanic effect
- Shift current across topological phase transitions



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