KIAS workshop on Topology and Correlation in Quantum Materials
May 31, 2019

Quantized circular photogalvanic effect in multifold fermions

Takahiro Morimoto

University of Tokyo, Department of Applied Physics

Berkeley UNIVERSITY OF CALIFORNIA

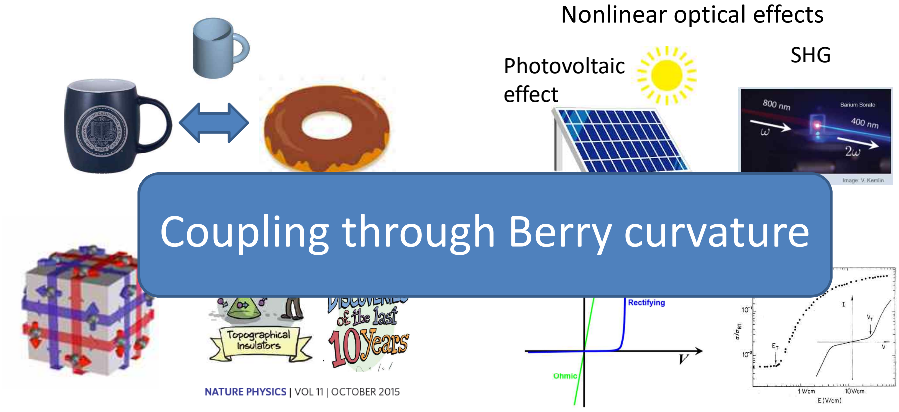
THE UNIVERSITY OF TOKYO

Geometry Topology ⟷ Nonlinear responses

Nonlinear optical effects

Photovoltaic effect

SHG



Coupling through Berry curvature

Plan of this talk

- Introduction
  - Geometry and topology in k space
- Quantized circular photogalvanic effect in Weyl semimetals
  - Generalization to chiral multifold fermions
  - Material realization in RhSi

de Juan, Grushin, Morimoto, Moore, Nat. Commun. (2017)
Flicker et al., PRB (2018)
Rees et al. arxivarXiv:1902.03230

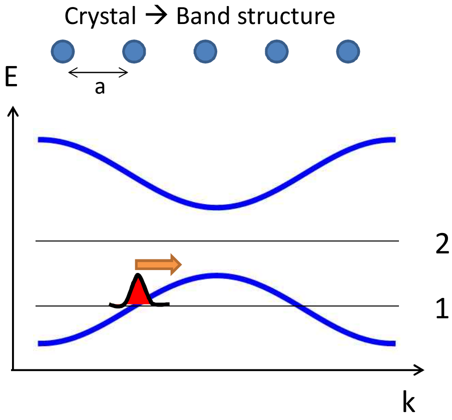
- Shift current as a geometric nonlinear response
  - Berry phase formula from Floquet theory
  - I-V characteristics and application to LLs

Morimoto, Nagaosa, Sci. Adv. (2016)
Morimoto, Nakamura, Kawasaki, Nagaosa, PRL (2018)

Electromagnetic responses in condensed matter

$$H_{\text{coupling}} = \underline{\boldsymbol{J}} \cdot \boldsymbol{A}$$

- *Intra*-band matrix element
- *Inter*-band matrix element



1. Metal

Lead

2. Insulator

Rock salt

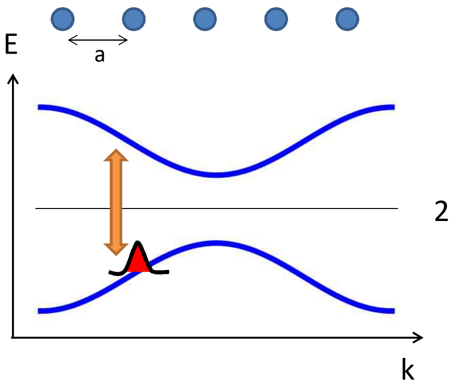
Electromagnetic responses in condensed matter

$$H_{\text{coupling}} = \boldsymbol{J} \cdot \boldsymbol{A}$$

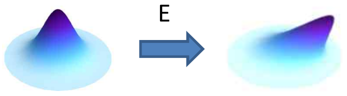
Intra-band matrix element

Inter-band matrix element

Crystal → Band structure



Distortion of wave function



- Quantum Hall current
- Anomalous Hall current
- Polarization current

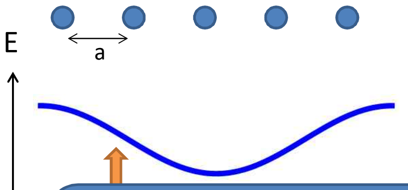
Electromagnetic responses in condensed matter

$$H_{\text{coupling}} = \boldsymbol{J} \cdot \boldsymbol{A}$$

Intra-band matrix element

Inter-band matrix element

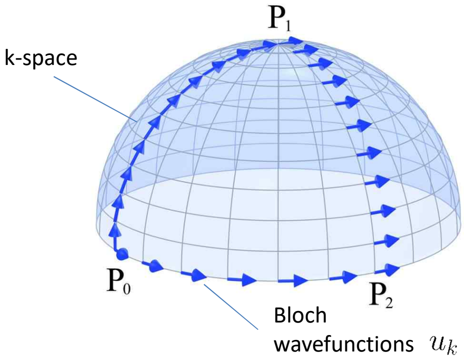
Crystal → Band structure

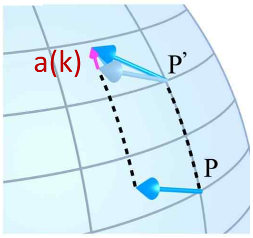


Distortion of wave function

Constraining a wave packet within a band
→ Geometry of the energy band

Geometry in k space





Berry connection: $\boldsymbol{a}(\boldsymbol{k}) = i\langle u_{\boldsymbol{k}}|\boldsymbol{\nabla}_{\boldsymbol{k}}|u_{\boldsymbol{k}}\rangle$

Berry curvature: $\boldsymbol{F}(\boldsymbol{k}) = \boldsymbol{\nabla}_{\boldsymbol{k}} \times \boldsymbol{a}(\boldsymbol{k})$

Position operator in k space rep.: $\hat{\boldsymbol{r}} = i\nabla_{\boldsymbol{k}} + \boldsymbol{a}(\boldsymbol{k})$ Intracell coordinate

Current response and band geometry

$$\frac{dx}{dt} = -i[x, H]$$

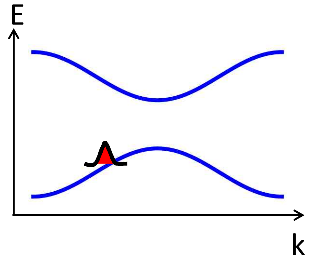
$$\Bigg\downarrow \quad \left[\begin{array}{l} x = i\partial_{k_x} + a_x(\boldsymbol{k}) \\ H = \dfrac{\boldsymbol{k}^2}{2m} + V(\boldsymbol{r}) \end{array}\right.$$

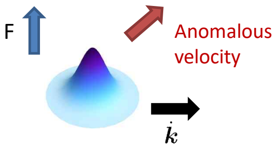
$$\frac{dx}{dt} = \frac{k_x}{m} - i[x, y]\frac{\partial V}{\partial y}$$

$$\Bigg\downarrow \quad [x, y] = i[\partial_{k_x} a_y(\boldsymbol{k}) - \partial_{k_y} a_x(\boldsymbol{k})] = iF(\boldsymbol{k})$$

Berry curvature in k-space

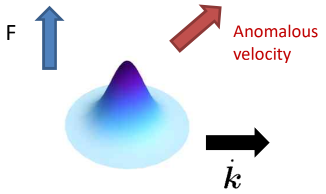
$$\frac{dx}{dt} = \underbrace{\frac{k_x}{m}}_{\text{Group velocity}} + \underbrace{F(k)\frac{\partial V}{\partial y}}_{\text{Anomalous velocity}}$$

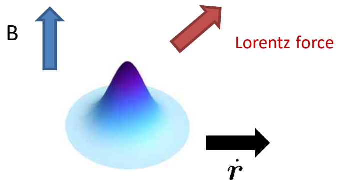




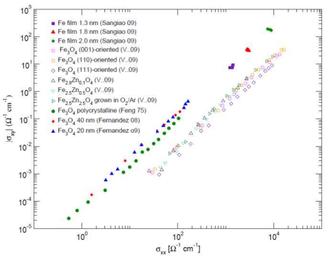
Anomalous velocity and anomalous Hall effect

$$\dot{\boldsymbol{r}} = \nabla_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}} - \underline{\dot{\boldsymbol{k}} \times \boldsymbol{F}} \qquad \hbar\dot{\boldsymbol{k}} = -e\boldsymbol{E} - \underline{e\dot{\boldsymbol{r}} \times \boldsymbol{B}}.$$





- Anomalous Hall effect
  - Hall effects without B



Nagaosa et al., RMP (2010)

Plan of this talk

- Introduction
  - Geometry and topology in k space
- Quantized circular photogalvanic effect in Weyl semimetals
  - Generalization to chiral multifold fermions
  - Material realization in RhSi

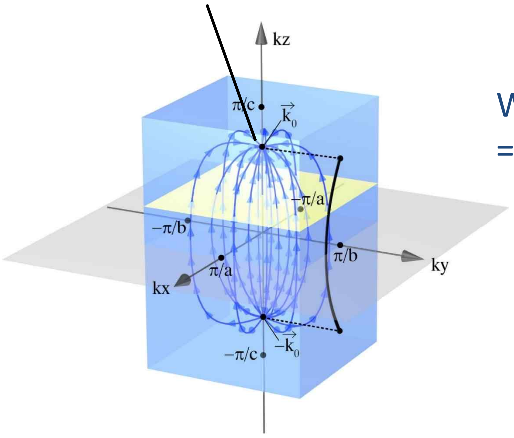
de Juan, Grushin, Morimoto, Moore, Nat. Commun. (2017)
Flicker et al., PRB (2018)
Rees et al. arxivarXiv:1902.03230

- Shift current as a geometric nonlinear response
  - Berry phase formula from Floquet theory
  - I-V characteristics and application to LLs

Morimoto, Nagaosa, Sci. Adv. (2016)
Morimoto, Nakamura, Kawasaki, Nagaosa, PRL (2018)

Weyl semimetal: Monopole in k space

$$H_0 = k_x\sigma_x + k_y\sigma_y + k_z\sigma_z,$$

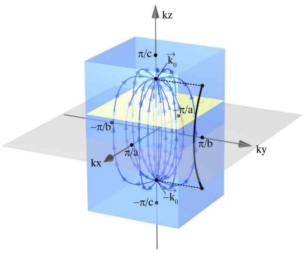


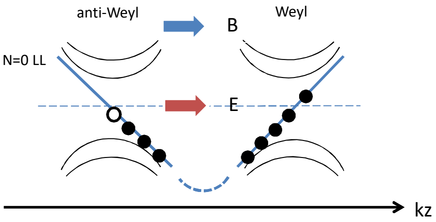
Weyl point
= Berry curvature monopole

Monopole charge: $\frac{1}{2\pi}\oint d\boldsymbol{S}\cdot\boldsymbol{\Omega}$

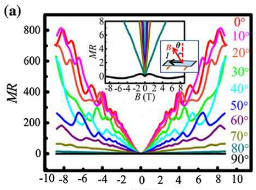
Chiral anomaly

- Chiral anomaly
  – $\frac{dQ^5}{dt} = \frac{2\nu}{(2\pi)^2}\frac{e^2}{\hbar^2}\boldsymbol{E}\cdot\boldsymbol{B},$





- Negative magnetoresistance
  – Not quantized

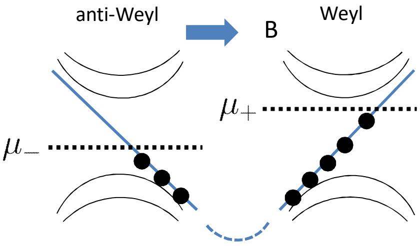


Huang et al. PRX (2015)

Search for quantized response in Weyl semimetals

- Chiral magnetic effect
  - $J \propto (\mu_L - \mu_R)B$
  - Zero in the equilibrium

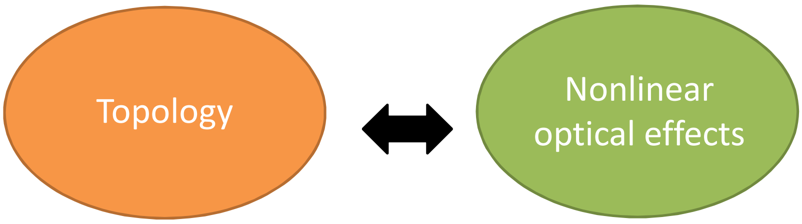
Fukushima et al, PRD (2018)



- Gyrotropic magnetic effect
  - ac effect
  - $J(\omega) \propto (\varepsilon_L - \varepsilon_R)B(\omega)$
  - not quantized, material dependent

Zhong, Moore, Souza, PRL (2016)

Accessing monopole charge has been difficult in linear responses.



- Quantized circular photogalvanic effect in Weyl semimetals


Fernando de Juan


Adolfo Grushin

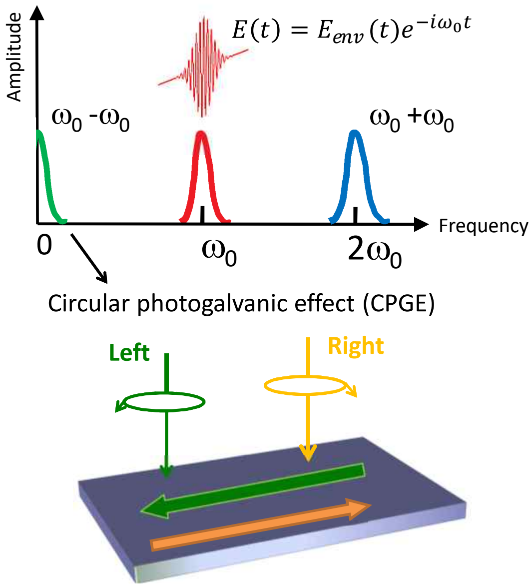

Joel Moore

de Juan, Grushin, Morimoto, Moore
Nat. Commun. (2017)

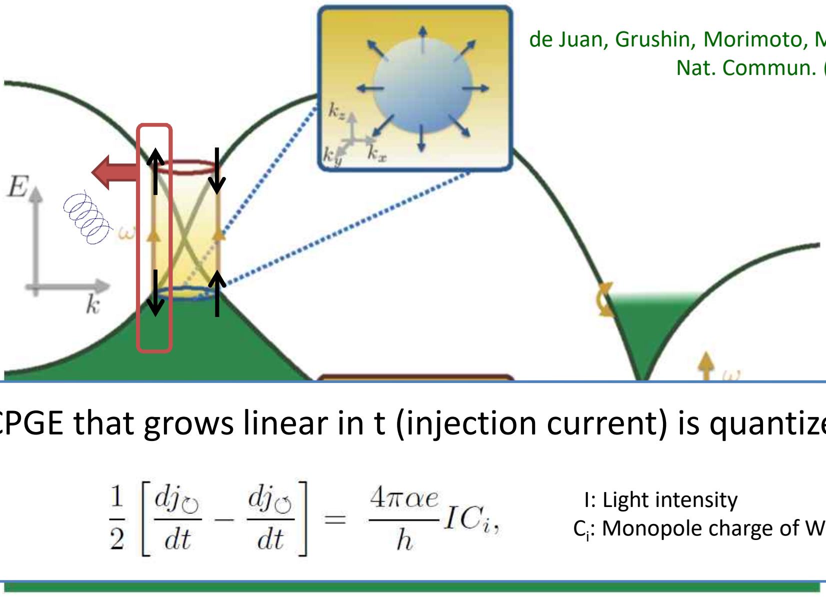
Multifold fermion RhSi
Felix Flicker et al., Phys. Rev. B (2018)
Dylan Rees, et al., arXiv:1902.03230

How about nonlinear responses?

Second order nonlinear optical effects



Quantized CPGE in Weyl semimetals



de Juan, Grushin, Morimoto, Moore, Nat. Commun. (2017)

CPGE that grows linear in t (injection current) is quantized.

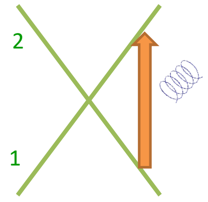
$$\frac{1}{2}\left[\frac{dj_{\circlearrowright}}{dt}-\frac{dj_{\circlearrowleft}}{dt}\right]=\frac{4\pi\alpha e}{h}IC_i,$$

I: Light intensity
C_i: Monopole charge of WF

Derivation by Fermi's golden rule

- Difference of transition rate

  ~ Berry curvature



$$(|v_{x,12}+iv_{y,12}|^2-|v_{x,12}-iv_{y,12}|^2)\frac{E^2}{\omega^2}\delta(\Delta E-\hbar\omega)$$

$$= \frac{2\mathrm{Im}[v_{x,12}v_{y,21}]}{\omega^2}E^2\delta(\Delta E-\hbar\omega) = \underline{\Omega_z}E^2\delta(\Delta E-\hbar\omega)$$

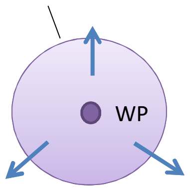
Ωz:Berry curvature

- Current = transition rate x group velocity

  = Berry flux

$$J_z \propto \int dk v_z \Omega_z \delta(\Delta E - \hbar\omega) = \int \Omega_z dS_z$$

$$\mathrm{tr}\beta = \sigma_{xyz} + \sigma_{yzx} + \sigma_{zxy} \propto \int dS \cdot \Omega = 2\pi C_i$$



Surface of optical transition $\Delta E = \hbar\omega$

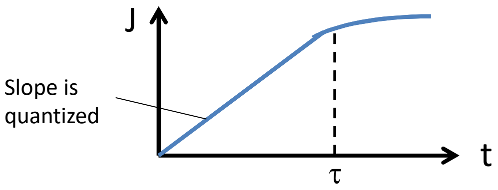
- Order estimate

$$J = 4\pi\alpha\frac{e}{h}\tau I = 22.17\frac{\tau}{\mathrm{ps}}\frac{\mathrm{I}}{\mathrm{W/cm^2}}\frac{\mathrm{A}}{\mathrm{cm^2}}.$$

→ 2nA/(W/cm^2)
for t=1ps,
10nm x 1mm^2

- Corrections from higher energy bands ~(w/ΔE)^2
  - Negligible in low frequency

- Saturation with relaxation time

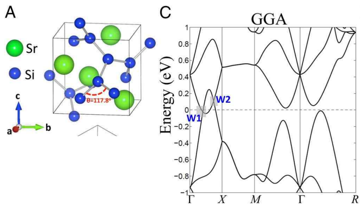


All optical measurement?

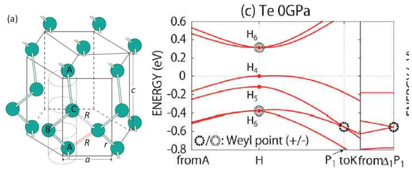
Candidate materials

- Mirror free Weyl semimetals
  - Otherwise Weyl and anti-Weyl points appear at the same energy
  - TaAs has mirror planes and doesn't support quantized CPGE

▪ SrSi2 Huang et al., PNAS (2016)



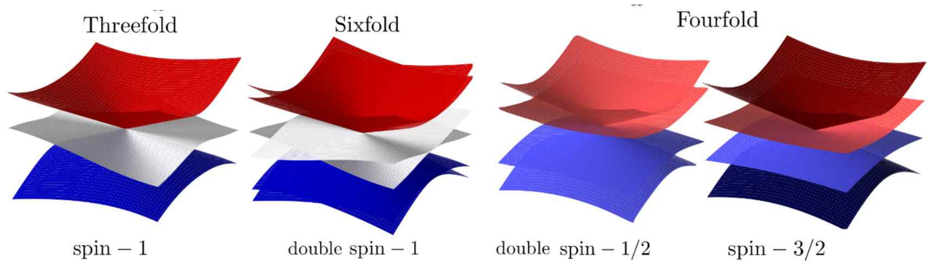
▪ doped Te Hirayama et al., PRL (2015)



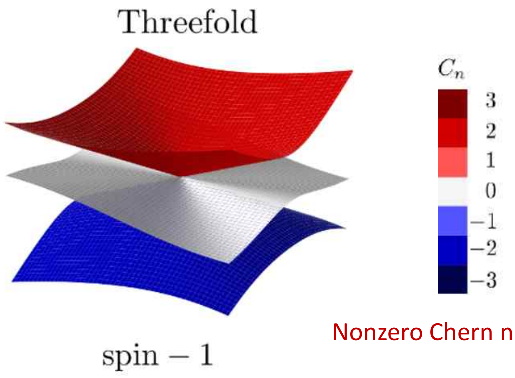
Quantized CPGE is not limited to conventional Weyl semimetals

Multifold fermions

- Bulk gapless excitations with more than twofold degeneracy at gapless points
  - Higher spin versions of Weyl semimetals



Multifold fermions



Nonzero Chern number C_n

$$H = \boldsymbol{k}\cdot\boldsymbol{S} = \hbar v_F \begin{pmatrix} 0 & e^{i\phi}k_x & e^{-i\phi}k_y \\ e^{-i\phi}k_x & 0 & e^{i\phi}k_z \\ e^{i\phi}k_y & e^{-i\phi}k_z & 0 \end{pmatrix}$$

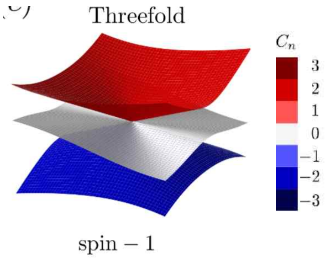
Pauli matrix σ is replaced by spin 1 operator S

CPGE is also quantized in multifold fermions

When the lowest band 1 is filled:

$$\begin{aligned}\beta(\omega) &= 4\pi^2\beta_0\left(\int d\vec{S}_{12}\cdot\vec{R}_{12} + \int d\vec{S}_{13}\cdot\vec{R}_{13}\right)\\ &= 4\pi^2\beta_0\Bigg(-i\int d\vec{S}_{12}\cdot\vec{\Omega}_1\\ &\qquad + \left[-\int d\vec{S}_{12} + \int d\vec{S}_{13}\right]\cdot\vec{R}_{13}\Bigg)\\ &= i\beta_0 C_1\end{aligned}$$

Cancel out



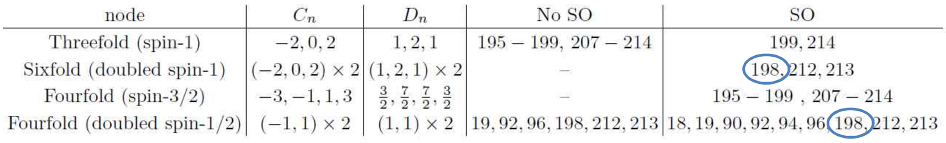
where $\vec{R}_{nm} = \vec{v}_{nm}\times\vec{v}_{mn}/(E_n - E_m)^2,$ S_{nm}: surface of optical transition

$\Omega_n^c = i\sum_{m\neq n} R_{nm}^c$ and $\beta_0 = \frac{\pi e^3}{h^2}$,

CPGE is quantized into the Chern number of the occupied bands.

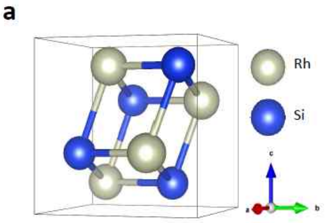
Realization of multifold fermion in RhSi

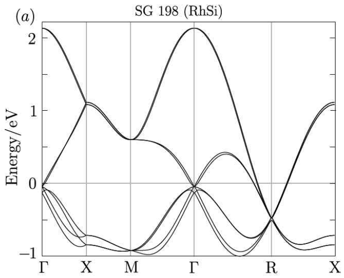
- Space group condition for multifold fermions



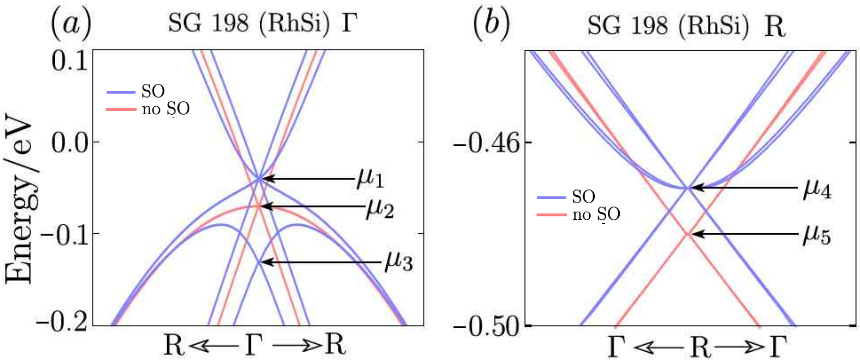
node	C_n	D_n	No SO	SO
Threefold (spin-1)	$-2, 0, 2$	$1, 2, 1$	$195 - 199,\ 207 - 214$	$199, 214$
Sixfold (doubled spin-1)	$(-2, 0, 2) \times 2$	$(1, 2, 1) \times 2$	–	$198, 212, 213$
Fourfold (spin-3/2)	$-3, -1, 1, 3$	$\frac{3}{2}, \frac{7}{2}, \frac{7}{2}, \frac{3}{2}$	–	$195 - 199\ ,\ 207 - 214$
Fourfold (doubled spin-1/2)	$(-1, 1) \times 2$	$(1, 1) \times 2$	$19, 92, 96, 198, 212, 213$	$18, 19, 90, 92, 94, 96, 198, 212, 213$

- RhSi: Space group 198, Chiral group





Multifold fermions in RhSi

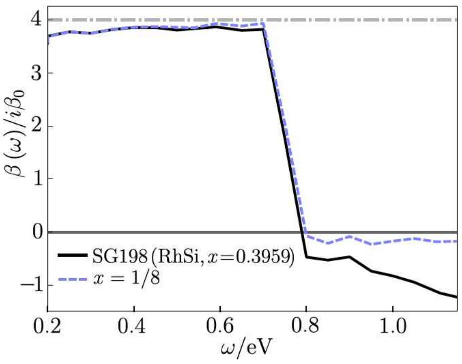


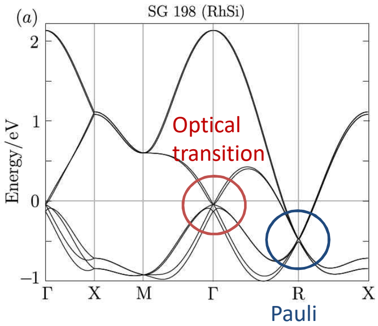
Degenerate threefold
→ Fourfold(S=3/2) + Weyl

Eightfold degeneracy
→ Sixfold + Weyl

Flicker, de Juan, Bradlyn, Morimoto, Vergniory, Grushin, Phys. Rev. B 98, 155145 (2018)

Theory of quantized CPGE in RhSi



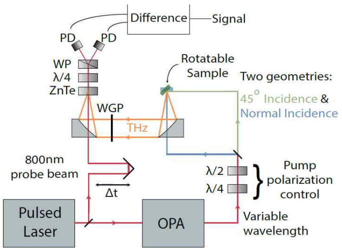


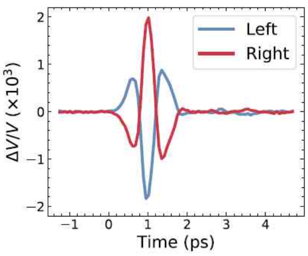
Optical transition

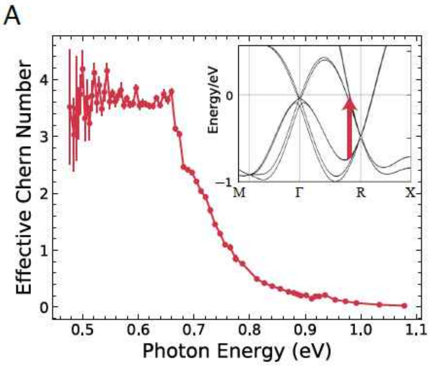
Pauli blocked

Flicker, de Juan, Bradlyn, Morimoto, Vergniory, Grushin, Phys. Rev. B 98, 155145 (2018)

THz measurement of quantized CPGE in RhSi



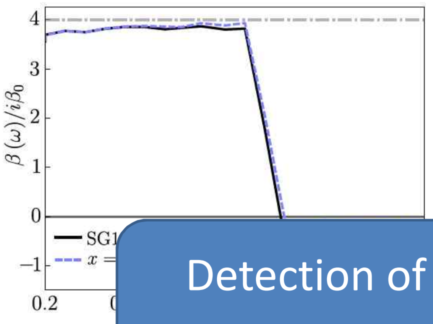




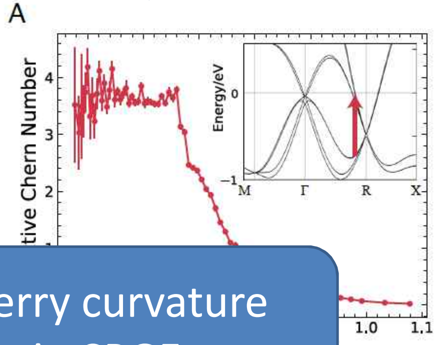
Rees, Manna, Lu, Morimoto, Borrmann, Felser, Moore, Torchinsky, Orenstein, arXiv:1902.03230

THz measurement of quantized CPGE in RhSi

Theory



Experiment for RhSi



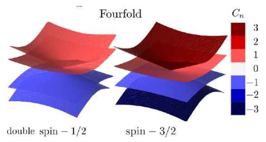
Detection of Berry curvature monopole via CPGE

Rees, Manna, Lu, Morimoto, Borrmann, Felser, Moore, Torchinsky, Orenstein, arXiv:1902.03230

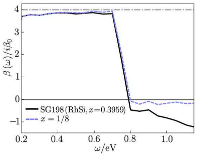
Summary (Part I)

- Quantized CPGE in multifold fermions

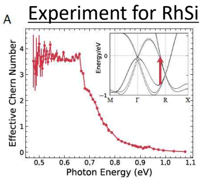
- Quantized CPGE has been experimentally confirmed in RhSi with THz measurement .



Theory



Experiment for RhSi



[Theory] de Juan e al., Nat. Commun. (2017)
Felix Flicker et al., Phys. Rev. B (2018)
[Experiment] Dylan Rees, Joe Orenstein et al., arXiv:1902.03230

Collaborators

Quantized CPGE in Weyls


Fernando de Juan


Adolfo Grushin


Joel Moore

Quantized CPGE in multifold fermions


Felix Flicker


Barry Bradlyn


Maia Vergniory

THz measurement of RhSi


Dylan Rees


Joe Orenstein


Darius Torchinsky

Plan of this talk

- Introduction
  - Geometry and topology in k space

- Quantized circular photogalvanic effect in Weyl semimetals
  - Generalization to chiral multifold fermions
  - Material realization in RhSi

de Juan, Grushin, Morimoto, Moore, Nat. Commun. (2017)
Flicker et al., PRB (2018)
Rees et al. arxivarXiv:1902.03230

- Shift current as a geometric nonlinear response
  - Berry phase formula from Floquet theory
  - I-V characteristics and application to LLs

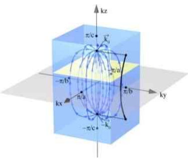
Morimoto, Nagaosa, Sci. Adv. (2016)
Morimoto, Nakamura, Kawasaki, Nagaosa, PRL (2018)

Geometry
Topology

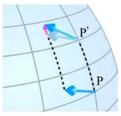
⟷

Nonlinear
responses

- Coupling through
  - ☑ Berry curvature: Quantized CPGE



  - ☐ Berry phase?



Geometry
Topology

⟷

Nonlinear
responses

- Geometrical formulas for shift current from Floquet theory

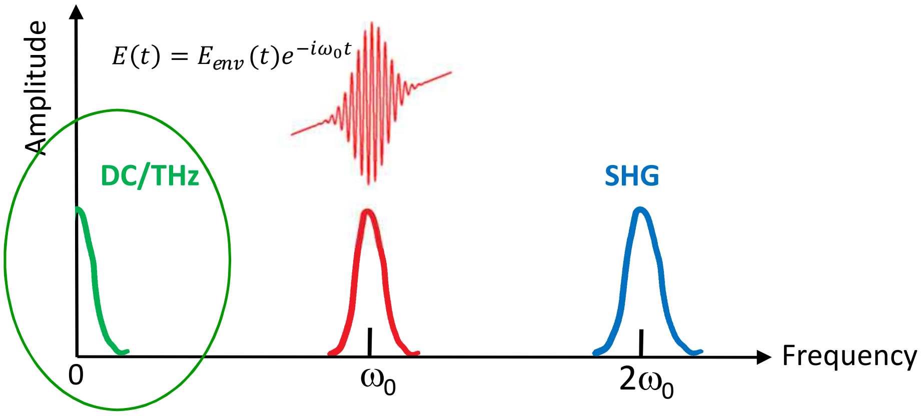
Naoto Nagaosa

Morimoto, Nagaosa, Sci. Adv. (2016)
Morimoto, Nakamura, Kawasaki, Nagaosa, PRL(2018)

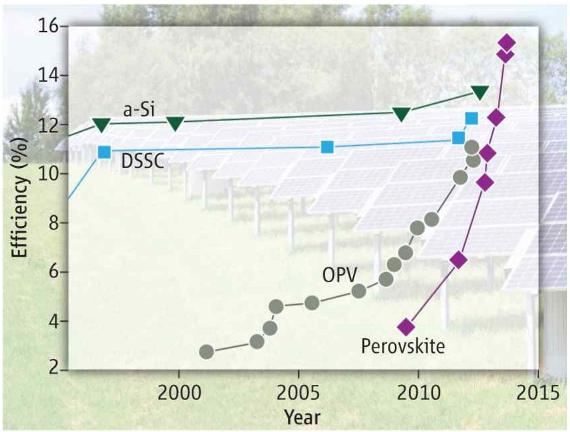
Second order nonlinear optical effects

$J(\omega+\omega') = \sigma(\omega+\omega'; \omega,\omega')\ E(\omega)\ E(\omega')$

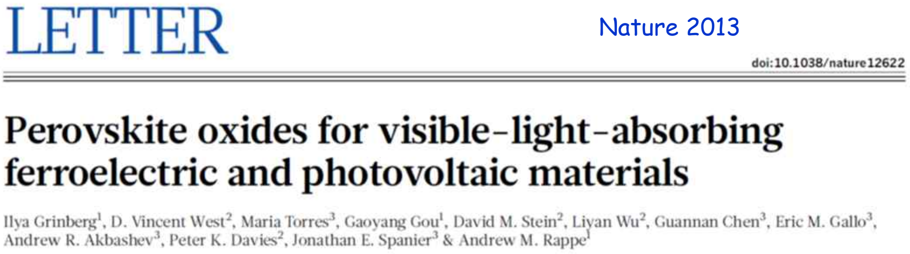
Vanishes under inversion symmetry (σ=0): $J = \sigma\ E(\omega)\ E(\omega') \Rightarrow -J = \sigma\ (-E(\omega))\ (-E(\omega'))$



Photovoltaic effect



Hodes, Science (2013)



LETTER

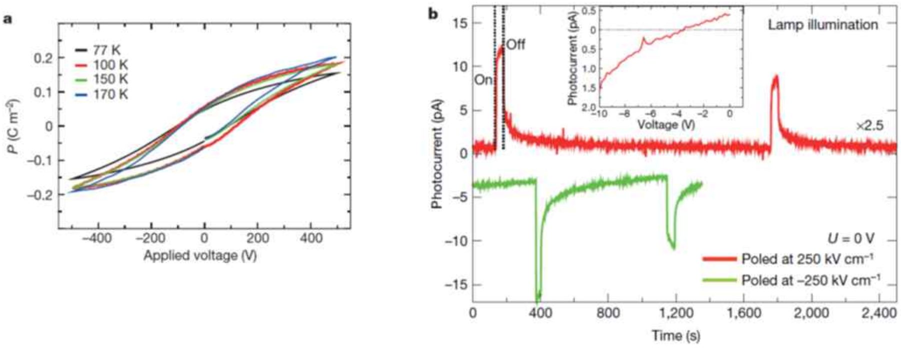
Nature 2013

doi:10.1038/nature12622

Perovskite oxides for visible-light-absorbing ferroelectric and photovoltaic materials

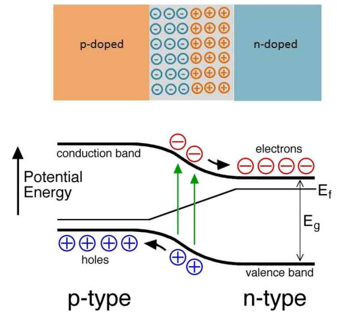
Ilya Grinberg[1], D. Vincent West[2], Maria Torres[3], Gaoyang Gou[1], David M. Stein[2], Liyan Wu[2], Guannan Chen[3], Eric M. Gallo[3], Andrew R. Akbashev[3], Peter K. Davies[2], Jonathan E. Spanier[3] & Andrew M. Rappe[1]

$[KNbO_3]_{1-x}[BaNi_{1/2}Nb_{1/2}O_{3-\delta}]_x$ (KBNNO)

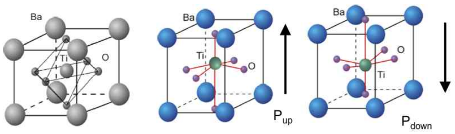


Mechanism of photovoltaic effect

- p-n junction
  - Potential gradient by artificial structure

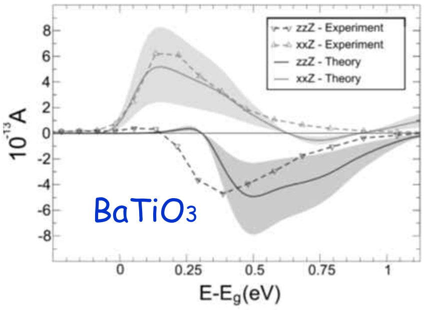


- Perovskite solar cell
  - Bulk crystal
  - Noncentrosymmetric

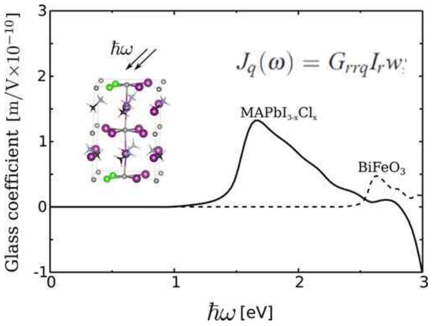


Shift current

- dc current proportional to E^2 Sipe, Shkrebtii, PRB (2000)
- Nonvanishing in noncentrosymmetric crystals

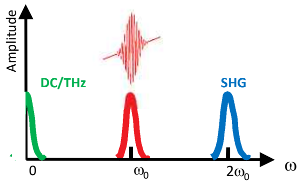


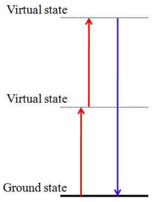
Young and Rappe, PRL (2015)

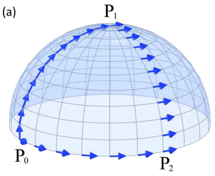


Rappe group, J. Phys. Chem. Lett., (2015)

Motivation



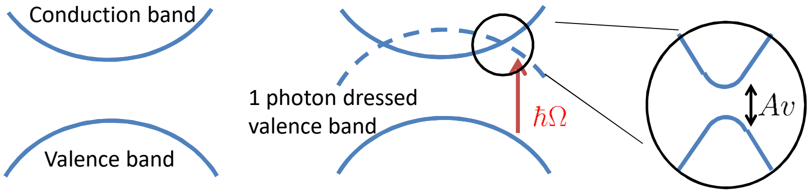




- Concise descriptions of second order nonlinear optical effects
- Not a complicated perturbation theory
- Connection to k-space geometry

Floquet theory describes nonlinear optical effects

Floquet formalism: an analog of Bloch theorem in the time direction



- Floquet two band model $H_F = \begin{pmatrix} \epsilon_1^0 + \hbar\Omega & Av_{12}^0 \\ Av_{21}^0 & \epsilon_2^0 \end{pmatrix}$ with $A = E/\Omega$
- Nonequilibrium steady state = Anticrossing of Floquet bands
- Photocurrent = sum of velocities of occupied Floquet states

  ← Keldysh Green's function

Compact expression of shift current

Joint DOS

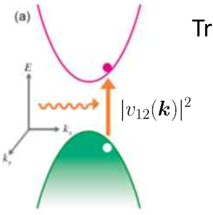
Saturation effect

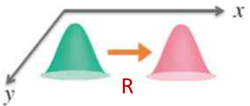
$$J = \frac{\pi E^2}{\omega^2}\int d\boldsymbol{k}\delta(\epsilon_1(\boldsymbol{k})-\epsilon_2(\boldsymbol{k})+\omega)\underline{\frac{\Gamma}{\sqrt{\frac{E^2}{\omega^2}+\Gamma^2}}}$$

$$\times|v_{12}(\boldsymbol{k})|^2\underline{[\nabla_{\boldsymbol{k}}\varphi_{12}(\boldsymbol{k})+\boldsymbol{a}_1(\boldsymbol{k})-\boldsymbol{a}_2(\boldsymbol{k})]}$$

Transition rate

Shift vector: R

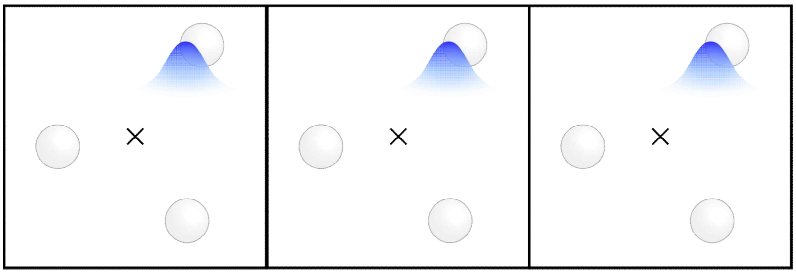


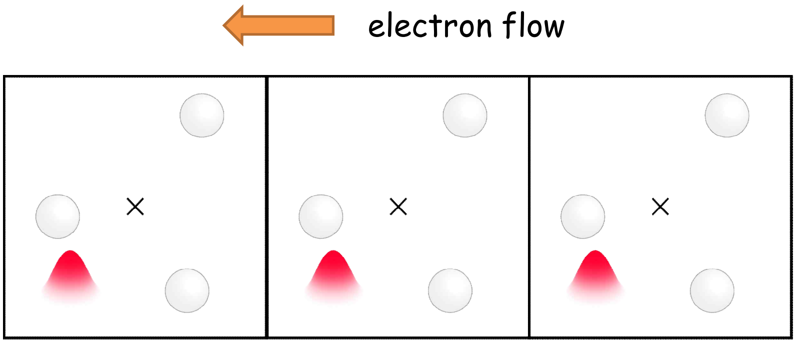


$$\begin{cases} \varphi_{12} = \mathrm{Im}(\log v_{12}^0) \\ \boldsymbol{a_i} = -i\langle u_k|\partial_k|u_k\rangle \end{cases}$$ Berry connection

Morimoto, Nagaosa, Sci. Adv. (2016)

[cf. perturbation theory : Sipe, Shkrebtii, PRB (2000)]

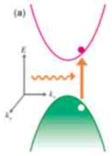



electron flow

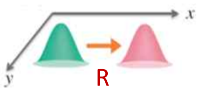
Semiclassical picture of shift current

- Photoexcitation creates electron-hole pairs that have polarization.
- Constant photoexcitation induces dc current.

Momentum space

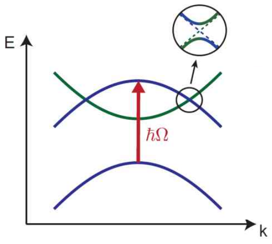


Real space



Advantage of Floquet theory description

$$J = \frac{\pi E^2}{\omega^2}\int d\boldsymbol{k}\delta(\epsilon_1(\boldsymbol{k})-\epsilon_2(\boldsymbol{k})+\omega)\frac{\Gamma}{\sqrt{\frac{E^2}{\omega^2}+\Gamma^2}} \times|v_{12}(\boldsymbol{k})|^2[\nabla_{\boldsymbol{k}}\varphi_{12}(\boldsymbol{k})+\boldsymbol{a}_1(\boldsymbol{k})-\boldsymbol{a}_2(\boldsymbol{k})]$$



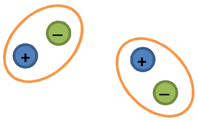
1. Requires information of only 2 bands
   - c.f. perturbation theory including all bands

Von Baltz, Kraut, PRB (1981); Sipe, Shkrebtii, PRB (2000)

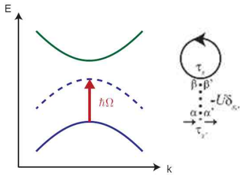
2. Floquet → Saturation effect (nonperturbative in E)
3. Keldysh → Interaction effect, relaxation effect

Applications of the Floquet description of shift current

1. Shift current of excitons

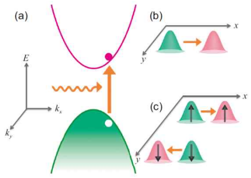


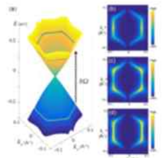
No free electron hole pair



Morimoto, Nagaosa,
PRB (2016)

2. Proposal of shift spin current



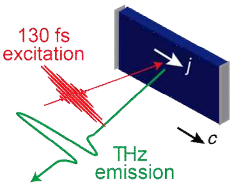


Application to TI

Kim, **Morimoto**, Nagaosa,
PRB (2016)

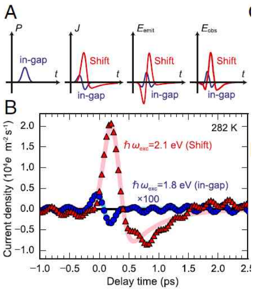
THz spectroscopy of shift current in SbSI

All optical measurement

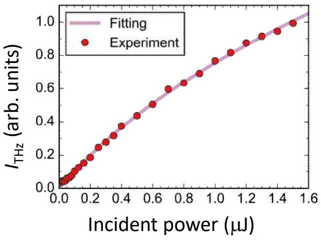


$E^{\mathrm{THz}}(\omega) \propto dj/dt$

Ultrafast dynamics



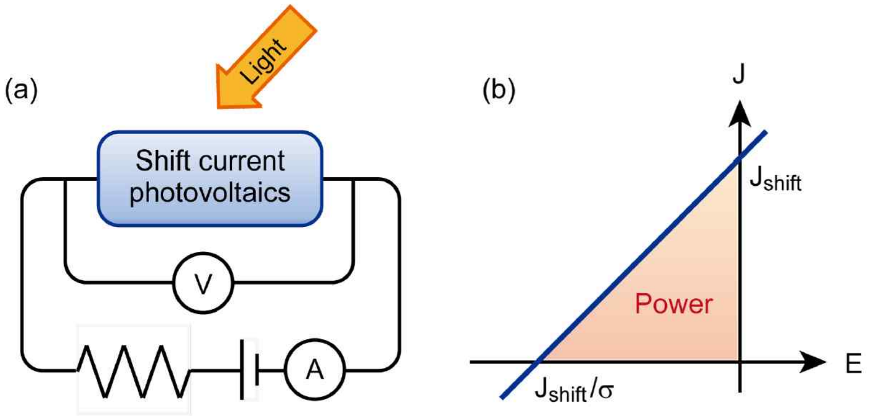
Saturation effect



Floquet theory: $J \propto \dfrac{\Gamma}{\sqrt{\frac{E^2}{\omega^2}+\Gamma^2}}$

Sotome, Nakamura, Fujioka, Ogino, Kaneko, Morimoto, Zhang,
Kawasaki, Nagaosa, Tokura, Ogawa, PNAS (2019)

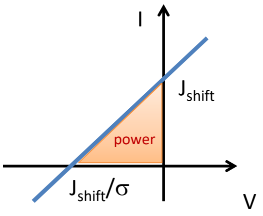
Application 1: I-V characteristics of shfit current photovoltaics



I-V characteristics from Floquet theory

- Floquet + Keldysh approach

$$J(E_{\rm dc}) = J_{\rm shift} + \sigma_E E_{\rm dc},$$



$$\sigma_E = \frac{4\pi e^4}{\hbar^3\omega^2}|E(\omega)|^2\tau^2 \int [dk]|v_{12}|^2(v_{11}-v_{22})R'\delta(\omega_{21}-\omega),$$

Relaxation time

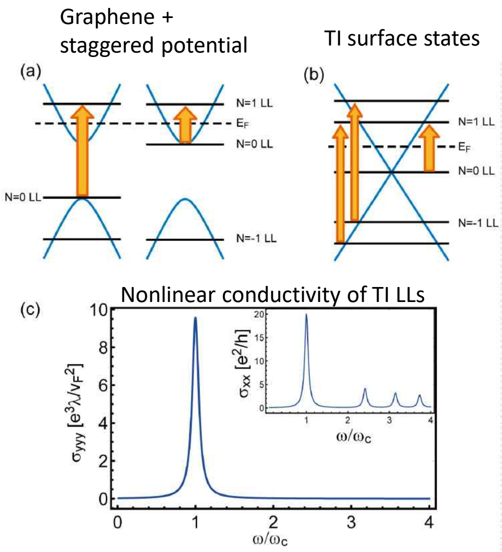
Group velocity difference

“Imaginary part” of shift vector

- σ: Slope of I-V
  - Metallic transport of photoexcited carriers
  - Flat band systems are suitable for application
    - Landau levels in graphene/ TI surface states

Morimoto, Nakamura, Kawasaki, Nagaosa, PRL (2018)

Energy harvesting in Landau levels

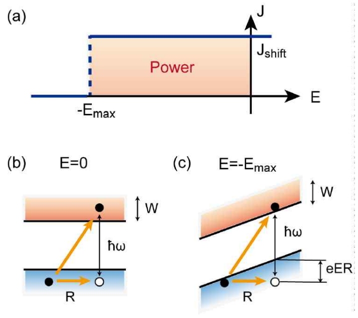


Graphene + staggered potential

TI surface states

Nonlinear conductivity of TI LLs

Cutoff E field determines conversion efficiency r

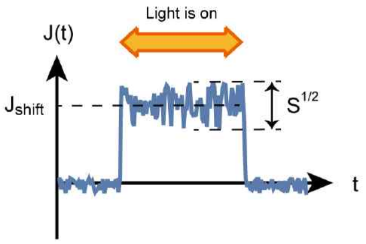


$$r \simeq \frac{W}{\hbar\omega} \simeq \frac{1}{\omega\tau}.$$

r =
- 11 % (graphene)
- 18% (MoS2)
- 29% (TI surface)

Application for photodetector

Shot noise of shift current



Noise formula from Floquet theory

$$S = \int dt(\langle v_{\rm loc}(t) v_{\rm loc}(0)\rangle - \langle v_{\rm loc}\rangle^2).$$

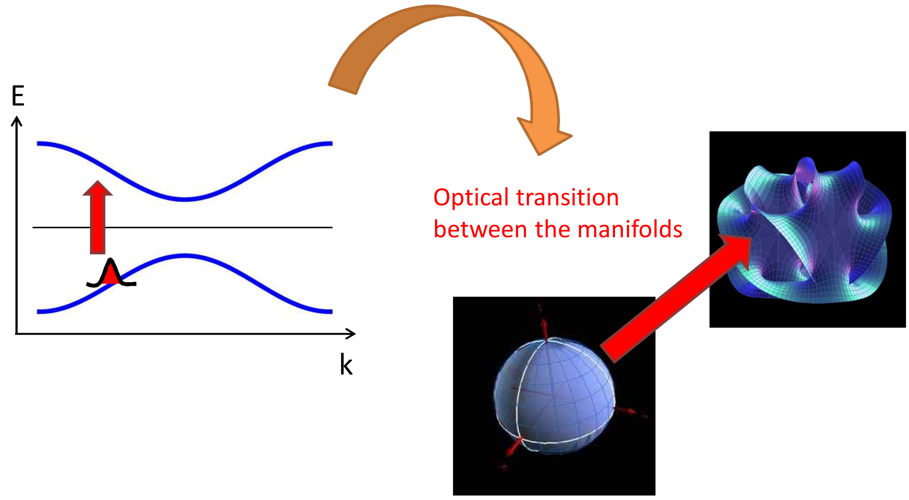
$$S = \frac{e^4}{\hbar^2\omega^2} E^2 \tau \int [dk] \underline{|v_{11} - v_{22}|} |v_{12}|^2 \delta(\omega_{21} - \omega).$$

Shot noise is also suppressed
for flat band systems

Landau levels may offer an efficient photodetector that is frequency tuanble

Morimoto, Nakamura, Kawasaki, Nagaosa, PRL (2018)

Geometry matters in nonlinear responses!



Optical transition
between the manifolds

Summary

- Quantized circular photogalvanic effect in Weyl semimetals
  - Observed in multifold fermion RhSi

de Juan, Grushin, Morimoto, Moore, Nat. Commun. (2017)
Flicker et al., PRB (2018)
Rees et al. arxivarXiv:1902.03230

- Shift current is a geometric nonlinear response
  - I-V characteristics suggests LLs may be good as a solar cell/photodetector.

Morimoto, Nagaosa, Sci. Adv. (2016)
Morimoto, Nakamura, Kawasaki, Nagaosa, PRL (2018)

- Outlook:
  - Observation of the quantized slope of CPGE at initial time and in other materials (Weyls)
  - Nonlinear responses in strongly correlated systems and its relationship to geometry