

Title: Origin of strong and/or quantized optical properties of topological semimetals

Speakers: Joel Moore

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Abstract: This talk starts by reviewing known examples of how topological materials generate new kinds of electrodynamic couplings and effects. Three-dimensional topological insulators realize a particular electromagnetic coupling known as axion electrodynamics, and understanding this leads to an improved understanding of magnetoelectricity in all materials. We then turn to how topological Weyl and Dirac semimetals can show unique electromagnetic responses; we argue that in linear response the main observable effect solves an old problem via the orbital moment of Bloch electrons, and how in nonlinear optics there should be a new quantized effect, which may have been seen experimentally. This nonlinear effect has a natural quantum  $e^3/h^2$  and appears in chiral Weyl semimetals over a finite range of frequencies. We discuss interaction and disorder corrections to nonlinear responses in closing.

# Origin of strong and/or quantized (non-)linear optical properties of topological semimetals

Quantum Matter Seminar, 16 November 2020

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S. Zhong, J. E. Moore, and I. Souza, *Phys. Rev. Lett.* **116**, 077201 (2016).

F. de Juan, A. Grushin, T. Morimoto, J. E. Moore, *Nat. Comm.* **8**, 15995 (2017).

S. Patankar et al., *Phys. Rev. B* **98**, 165113 (2018).

D. Rees et al., *Science Advances* 2020

A. Avdoshkin, V. Kozii, *JEM, PRL* 2020

K. Takasan, T. Morimoto, *JEM, arXiv* 2020

$$j_{2D} = \frac{e^2 E}{h}; \quad \frac{dj_{3D}}{dt} = \frac{e^2 E}{h} \left( \frac{eE}{h} \right)?$$

Experimental collaborators:

Felser (Dresden), Orenstein (Berkeley), Torchinsky (Temple) groups



# Types of order

In 1980, the first ordered phase beyond symmetry breaking was discovered.

Electrons confined to a plane and in a strong magnetic field show, at low enough temperature, plateaus in the “Hall conductance”:

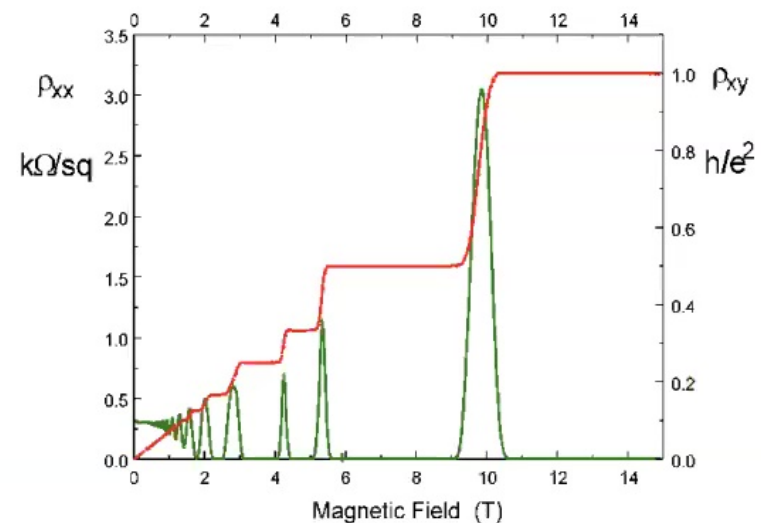
force  $I$  along  $x$  and measure  $V$  along  $y$

on a plateau, get

$$\sigma_{xy} = n \frac{e^2}{h}$$

at least within 1 in  $10^9$  or so.

Wavefunction topology  
causes this quantization



**Note I:** the AC Josephson effect between superconductors similarly allows determination of  $e/h$ .

**Note II:** there are also *fractional* plateaus in good (modulation-doped) samples.

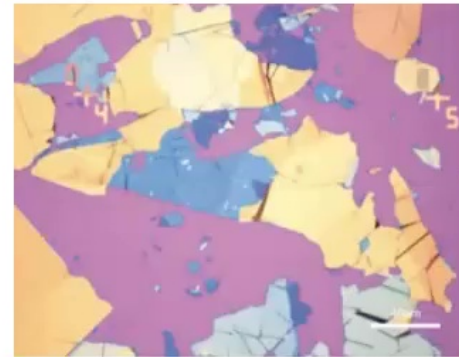
# Optical “quantization” in semimetals, linear version

Properties of the “semi-metallic” electrons in graphene:  
effective mass is zero

one layer of graphene attenuates 2.3% of light

( $\pi$  times the fine structure constant)

$$\frac{\pi e^2}{hc}$$



This effect is not particularly topological or well protected, but it shows what might be possible experimentally in metallic systems at nonzero frequency.

# Why study nonlinear optics of Weyls?

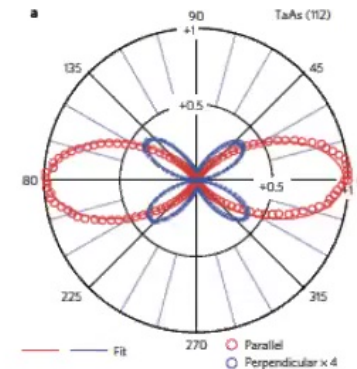
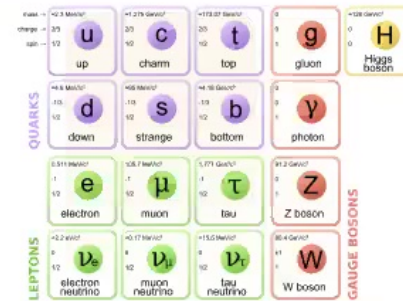
Weyl semimetals realize a kind of emergent particle not present in the “Standard Model” of particle physics.

Nonlinear optics is clearly exciting in these materials:

I. Nonlinear optics in Weyls such as TaAs is extremely strong

II. One nonlinear optical in mirror-free Weyls is not just strong but approximately *quantized*, for topological reasons

III. Linear optics isn't good enough...



# Outline

I. Intro: how do topological materials yield interesting electromagnetic responses? An example: magnetoelectric effects

II. Topological *semimetals*: what are topological semimetals in 3D? What are their unique electromagnetic responses?

Chiral magnetic effect. Chiral anomaly. *Chiral photocurrent*.

III. Add interactions in a toy model. Comparison to graphene.

IV. Out of equilibrium: induce nonlinear optical effects by current.

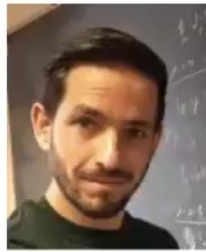
A general question to keep in mind:

How is quantization different in ideal semimetals than insulators?

# Acknowledgements (main part)



Fernando de Juan



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## Experimental groups



Claudia Felser



Joseph Orenstein



Darius Torchinsky

For projects at end:

**Vlad Kozii, Alex Avdoshkin, and Kazuaki Takasan.**

# Symmetry and (topological) response:

Symmetry tells us whether a response is *allowed*.  
("Everything that is allowed is mandatory.")

Some optically relevant examples:

**Inversion-breaking:** allows SHG along some directions (e.g., GaAs)

**Polar:** subset of inversion-breaking with a preferred direction

**Chiral:** another subset, with no mirrors: allows optical rotation without breaking time-reversal.

Geometric and topological mechanisms can lead to a *magnitude* that is largely independent of microscopic details.

(They also have forced us to think more carefully about what symmetry prohibits.)



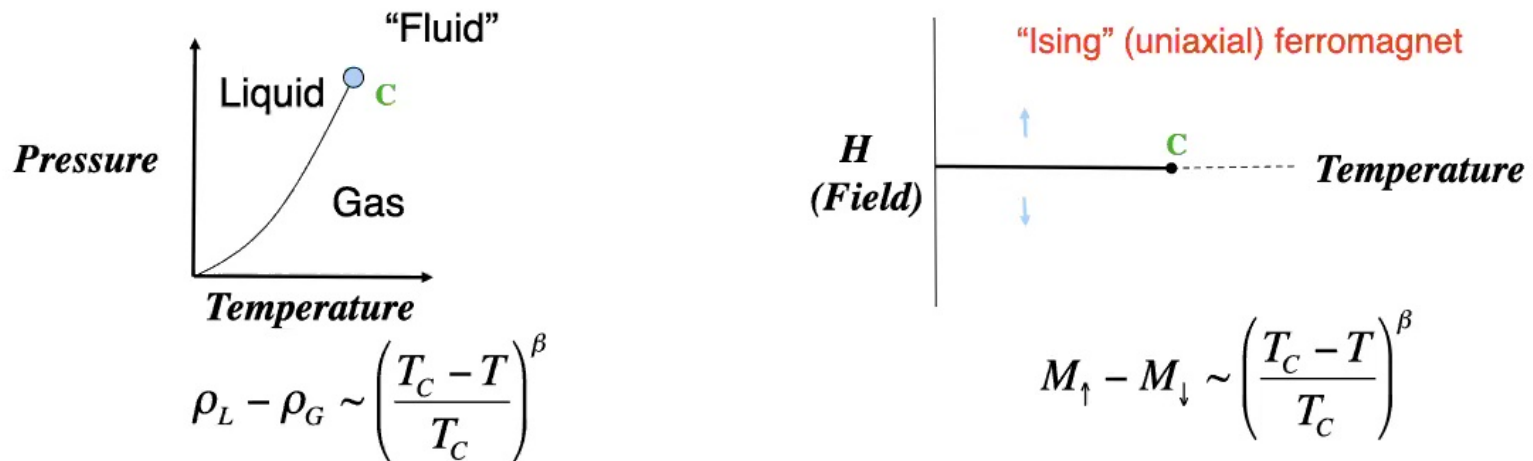
# Outline (in fewer words)

I. We now know that there are quantum-geometric effects in the basic theory of metals. Can measurable metallic effects be fully “topological” in the same sense as topological insulating phases?

# Types of order

At high temperature, entropy dominates and leads to a disordered state.  
At low temperature, energy dominates and leads to an ordered state.

In case this sounds too philosophical, there are testable results that come out of the “Landau theory” of symmetry-breaking:



Experiment :  $\beta = 0.322 \pm 0.005$

Theory :  $\beta = 0.325 \pm 0.002$

“Universality” at continuous phase transitions (Wilson, Fisher, Kadanoff, ...)

# Berry phase in solids

Every simple gauge-invariant object made from  $A$  and  $F$  seems to mean something physically. We can identify several types of Berry-phase phenomena of nearly free electrons:

Insulators:

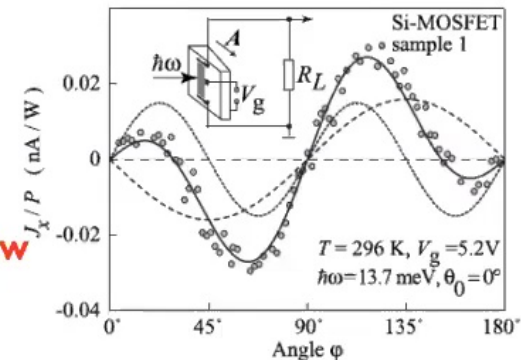
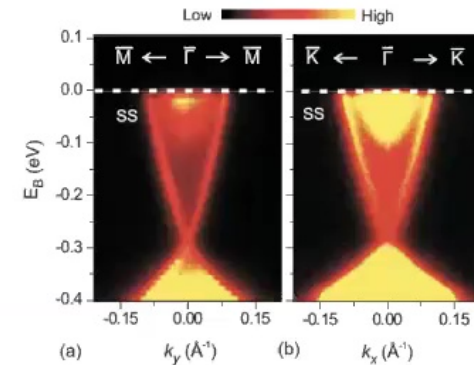
Topological phases independent of symmetry:  
Examples: 2D and 4D QHE (1982,1988)

Topological phases dependent on symmetry  
Examples: 2D and 3D Z<sub>2</sub> topological insulators (2005,2007)

The Berry-phase approach to understanding these leads to expressions that are physically meaningful without symmetries:

Examples: electrical polarization (1987-1990);  
magnetoelectric effect (2009-2010)

Metals: Several long-observed phenomena in metals are now believed to be Berry-phase effects, albeit not quantized. I will give a quick description of 3 (1999,2010,2012).



# Berry phase

What kind of “curvature” can exist for electrons in a solid?

Consider a quantum-mechanical system in its (nondegenerate) ground state.

The adiabatic theorem in quantum mechanics implies that, if the Hamiltonian is now changed slowly, the system remains in its time-dependent ground state.

But this is actually very incomplete (Berry).

When the Hamiltonian goes around a *closed loop*  $k(t)$  in parameter space, there can be an irreducible *phase*

$$\phi = \oint \mathcal{A} \cdot d\mathbf{k}, \quad \mathcal{A} = \langle \psi_k | -i \nabla_k | \psi_k \rangle$$

relative to the initial state.

Why do we write the phase in this form?

Does it depend on the choice of reference wavefunctions?



Michael Berry

# Berry phase in solids

In a solid, the natural parameter space is electron momentum.

The change in the electron wavefunction *within the unit cell* leads to a Berry connection and Berry curvature:

$$\psi(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{\mathbf{k}}(\mathbf{r})$$

$$\mathcal{A} = \langle u_{\mathbf{k}} | -i\nabla_{\mathbf{k}} | u_{\mathbf{k}} \rangle \quad \mathcal{F} = \nabla \times \mathcal{A}$$

We keep finding more physical properties that are determined by these quantum geometric quantities.

The first was that the integer quantum Hall effect in a 2D crystal follows from the integral of  $\mathcal{F}$  (like Gauss-Bonnet!). Explicitly,

$$n = \sum_{\text{bands}} \frac{i}{2\pi} \int d^2k \left( \left\langle \frac{\partial u}{\partial k_1} \middle| \frac{\partial u}{\partial k_2} \right\rangle - \left\langle \frac{\partial u}{\partial k_2} \middle| \frac{\partial u}{\partial k_1} \right\rangle \right) \quad \mathcal{F} = \nabla \times \mathcal{A}$$

$$\sigma_{xy} = n \frac{e^2}{h}$$

TKNN (Thouless et al.), 1982

“first Chern number”



S. S. Chern

# Berry phase in solids

Every simple gauge-invariant object made from  $A$  and  $F$  seems to mean something physically. We can identify several types of Berry-phase phenomena of nearly free electrons:

## Insulators:

Topological phases independent of symmetry:

**Examples: 2D and 4D QHE (1982,1988), QAHE.**

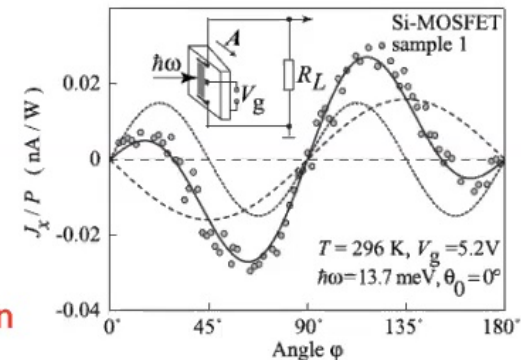
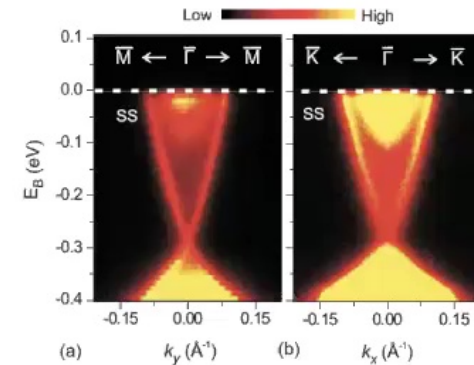
Topological phases dependent on symmetry

**Examples: 2D and 3D Z2 topological insulators (2005,2007). Antiferromagnetic TI (2019).**

The Berry-phase approach to understanding these leads to expressions that are physically meaningful without symmetries:

**Examples: electrical polarization (1987-1990); magnetoelectric effect (2009-2010)**

**Metals:** Several long-observed phenomena in metals are now believed to be Berry-phase effects. I will give a quick description of 3 (1999-2017).



# What is quantized in a 3D TI? Electrodynamics in insulators...

We know that the constants  $\epsilon$  and  $\mu$  in Maxwell's equations can be modified inside an ordinary insulator.

Particle physicists in the 1980s considered what happens if a 3D insulator creates a new term ("axion electrodynamics", Wilczek 1987)

$$\Delta\mathcal{L}_{EM} = \frac{\theta e^2}{2\pi h} \mathbf{E} \cdot \mathbf{B} = \frac{\theta e^2}{16\pi h} \epsilon^{\alpha\beta\gamma\delta} F_{\alpha\beta} F_{\gamma\delta}.$$

This term is a total derivative, unlike other magnetoelectric couplings. It is also "topological" by power-counting.

The angle  $\theta$  is periodic and odd under T.

A T-invariant insulator can have two possible values: 0 or  $\pi$ .

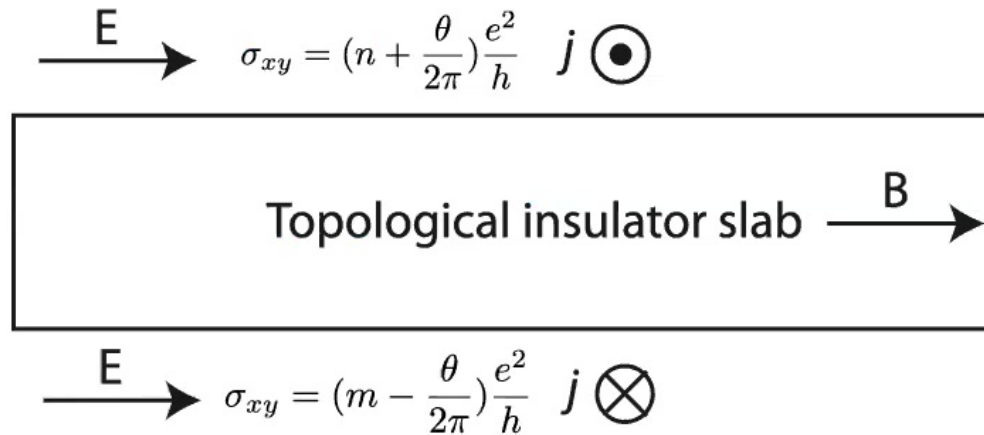
# Axion E&M

$$\Delta\mathcal{L}_{EM} = \frac{\theta e^2}{2\pi h} \mathbf{E} \cdot \mathbf{B} = \frac{\theta e^2}{16\pi h} \epsilon^{\alpha\beta\gamma\delta} F_{\alpha\beta} F_{\gamma\delta}.$$

This explains a number of properties of the 3D topological insulator when its surfaces become gapped by breaking T-invariance:

Magnetoelectric effect:

applying **B** generates polarization **P**, applying **E** generates magnetization **M**)

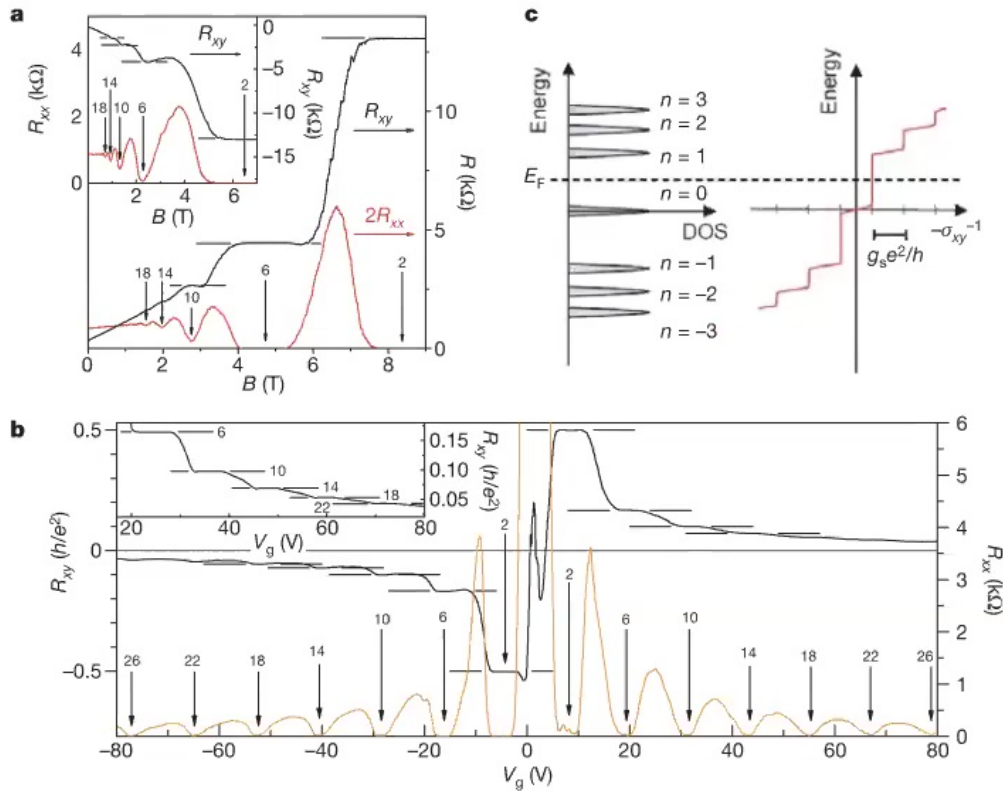




# Graphene QHE

The connection is that a single Dirac fermion contributes a *half-integer QHE*: this is seen directly in graphene if we recall the extra fourfold degeneracy.

Data shown below from Y. Zhang et al. (Kim group, Columbia)



# A non-Abelian example of electromagnetic response in insulators: axion electrodynamics

There is a “topological” part of the magnetoelectric term

$$\Delta\mathcal{L}_{EM} = \frac{\theta e^2}{2\pi h} \mathbf{E} \cdot \mathbf{B} = \frac{\theta e^2}{16\pi h} \epsilon^{\alpha\beta\gamma\delta} F_{\alpha\beta} F_{\gamma\delta}.$$

that is measured by the orbital magnetoelectric polarizability

$$\theta \frac{e^2}{2\pi h} = \frac{\partial M}{\partial E} = \frac{\partial}{\partial E} \frac{\partial}{\partial B} H = \frac{\partial P}{\partial B}$$

and computed by integrating the “Chern-Simons form” of the Berry phase

$$\theta = -\frac{1}{4\pi} \int_{\text{BZ}} d^3k \epsilon_{ijk} \text{Tr}[\mathcal{A}_i \partial_j \mathcal{A}_k - i \frac{2}{3} \mathcal{A}_i \mathcal{A}_j \mathcal{A}_k] \quad (*)$$

(Qi, Hughes, Zhang, 2008; Essin, JEM, Vanderbilt 2009; Experiments: Wu et al; Pimenov et al.)

This integral is quantized only in T-invariant insulators, but contributes in all insulators.

Has just the right gauge ambiguity under “large gauge transformations”.

Relevant to experiments by Armitage & Oh, among others.

# Non-Abelian Berry gauge fields phase in insulators

Note: If more than 1 degenerate state,  
the connection is *non-Abelian*:

$$A^{\alpha\beta} = \langle \psi_k^\alpha | -i \nabla_k | \psi_k^\beta \rangle$$

Two-fold degeneracies are automatic if nothing depends on spin.

1. Even with spin-orbit, certain momenta with  $k$  equal to  $-k$  still have degeneracies in non-magnetic materials, due to Kramers degeneracies;
2. Frequently, even if the occupied bands of a material are non-degenerate, calculation shows that physical properties depend on the non-Abelian connection as if they were degenerate.

So far, the orbital magnetoelectric polarizability is the only  $d \leq 3$  quantity I know of that depends on the non-Abelian Berry phase. It has an analogue in metallic dynamics (Varjas et al., PRL 2017).

# Topological response

**Many-body definition:** the Chern-Simons or second Chern formula does not directly generalize. However, the quantity  $dP/dB$  does generalize:  
a clue is that the “polarization quantum” combines nicely with the flux quantum.

$$\frac{\Delta P}{B_0} = \frac{e/\Omega}{h/e\Omega} = e^2/h.$$

So  $dP/dB$  gives a *bulk, many-body* test for a topological insulator.

(Essin, JEM, Vanderbilt 2009)

$$\frac{e^2}{h} \begin{aligned} &= \text{contact resistance in 0D or 1D} \\ &= \text{Hall conductance quantum in 2D} \\ &= \text{magnetoelectric polarizability in 3D} \end{aligned}$$

# Warmup for metals: polarization in insulators

Electrical polarization: “simple” Berry phase effect in solids (took about 50 years to understand how to calculate polarization of a solid from its unit cell)

Sum the integral of  $A$  over bands: in one spatial dimension,

$$P = \sum_v e \int \frac{dq}{2\pi} \langle u_v(q) | -i\partial_q | u_v(q) \rangle$$

Intuitive idea: think about the momentum-position commutation relation

$$A = \langle u_k | -i\nabla_k | u_k \rangle \approx \langle r \rangle$$

More seriously: relate changes in  $P$  to currents moving through the unit cell.

Polarization isn't quantized in general; it is just a simple physical observable determined by the Berry phase. **Note that there is an ambiguity  $ne$ .**

**Broader reason, in hindsight:**  $E(k)$ , the band structure, is  $k$ -symmetric with time-reversal, even with broken inversion. Anything related to inversion-breaking has to come from the wavefunction, and at low energy, *usually* from the Berry phase.

# What about metals?

Claim: the biggest omission in Ashcroft and Mermin (standard solids text) is a term in the semiclassical equations of motion, the (Karplus-Luttinger) ***anomalous velocity***.

$$\frac{dx^a}{dt} = \frac{1}{\hbar} \frac{\partial \epsilon_n(\mathbf{k})}{\partial k_a} + \mathcal{F}_n^{ab}(\mathbf{k}) \frac{dk_b}{dt}.$$

a “magnetic field” in momentum space.

The anomalous velocity results from changes in the electron distribution *within the unit cell*: **the Berry phase is connected to the electron spatial location.**

Example I: the intrinsic anomalous Hall effect in itinerant magnets (Fe, e.g.)

Example II: helicity-dependent photocurrents in optically active materials

Example III: optical rotation in gyrotropic/chiral materials with T symmetry

**Can we get anything quantized/interesting in a metal?**

# Anomalous Hall effect (100+ years)

From Nagaosa et al., RMP 2011

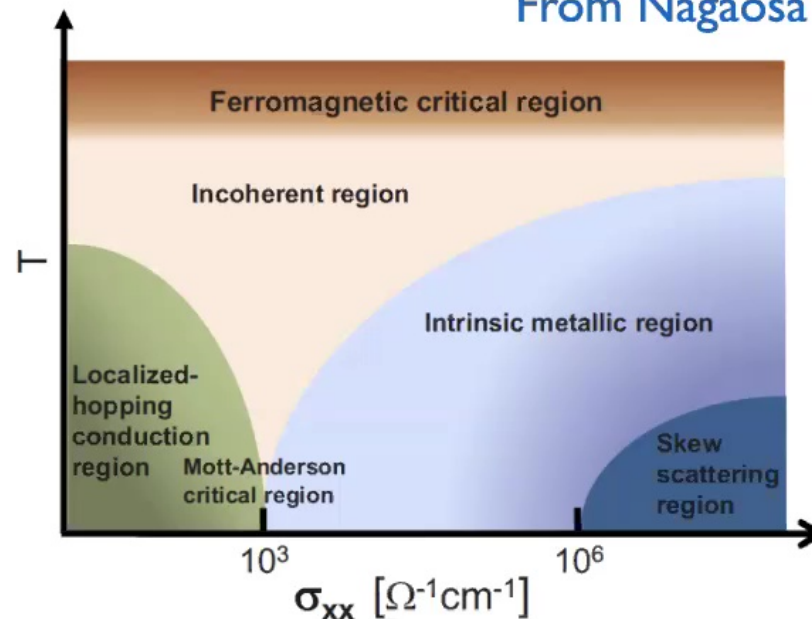


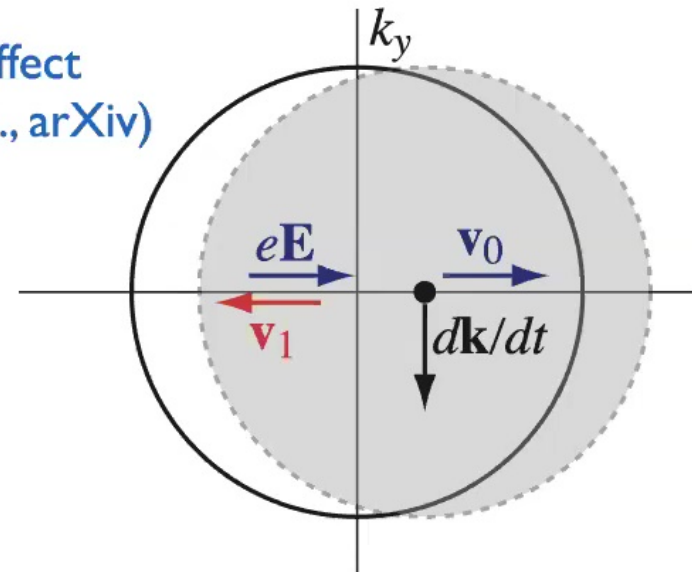
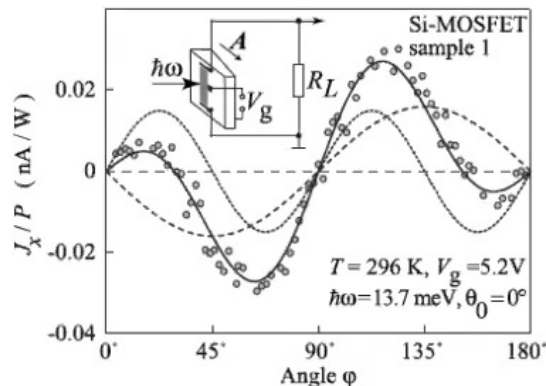
FIG. 47. (Color online) A speculative and schematic phase diagram for the anomalous Hall effect in the plane of the diagonal conductivity  $\sigma_{xx}$  and the temperature  $T$ .

cf. Sundaram and Niu, 1999 
$$\sigma_{xy} = \frac{e^2}{h} \int_{\text{FS}} d^2k \frac{F}{2\pi} + \text{extrinsic}$$

# Two “mystery” effects in *optics*

Here “mystery” means effect allowed by symmetry, and known to exist from experiment, but microscopic picture was unclear.

## I. Nonlinear optics: circular photogalvanic effect (JEM and J. Orenstein, PRL 2010; Deyo et al., arXiv)



Currents are switched by the sense of circular polarization, as previously observed in a series of experiments by S.D. Ganichev et al. We believe this is a semiclassical Berry-phase effect at low frequency.



# Another mystery effect: low-frequency optical activity

## 2. Linear optics:

Chiral materials (and sugar water!) can show optical rotation in transmission, similar to the Faraday effect but without time-reversal breaking.

**Surprise:** this problem is intimately connected to the “chiral magnetic effect” proposed in Weyl semimetals, although as sometimes described that effect is actually zero for topological reasons.

(Zhong, Moore, Souza, PRL 2016; Ma, Pesin, PRB 2016)

# 3D Dirac and Weyl metals

Can we find 3D materials that are massless semimetals like graphene?

Yes! There are two ways to generalize graphene's massless "Dirac electrons" to 3D.

In the early days of quantum mechanics, two alternatives were put forward that are "half" of Dirac's celebrated equation for the electron.



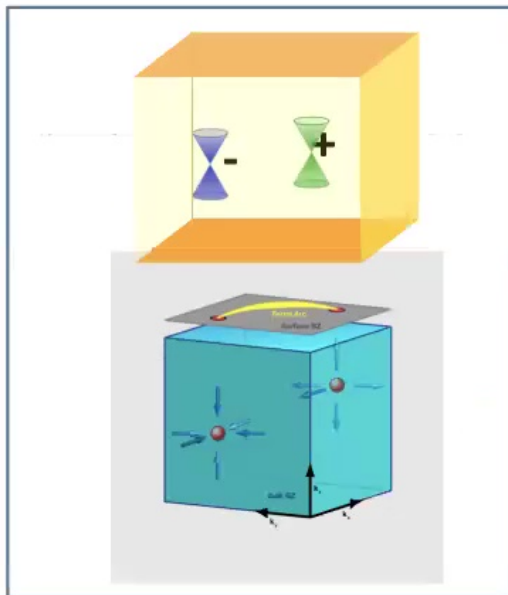
Dirac: 4 by 4 matrix equation describes the electron and the positron  
4-band semimetals found in  $\text{Na}_3\text{Bi}$ ,  $\text{Cd}_2\text{As}_3$ , 2013



Weyl: 2 by 2 matrix equation describes a particle with only one "handedness"  
Does not seem to exist in the standard model; neutrinos were a possibility  
2-band semimetals found in "inversion-breaking" TaAs, 2014-2015

# Weyl semimetals

old theory idea (Herring, Ph.D. thesis);  
trick is finding Weyl points at Fermi surface



*A Weyl point has topological charge: the Chern number from Berry flux through a small sphere around it is an integer. (Volovik; Murakami, 2008)*

There are surface Fermi arcs connecting Weyl points (Wan, Turner, Vishwanath, Savrasov, 2010), now seen in photoemission.

Unusual quantum oscillations (“Weyl wiggles”, Analytis et al.) from arcs.

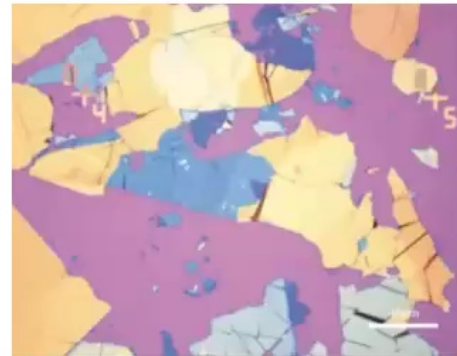
What are bulk consequences of Weyl topological charge?

# Optical quantization in semimetals, linear version

Properties of the “semi-metallic” electrons in graphene:  
effective mass is zero

one layer of graphene attenuates 2.3% of light

( $\pi$  times the fine structure constant)  $\frac{\pi e^2}{hc}$



This effect is not particularly topological or well protected, but it shows what might be possible experimentally in metallic systems at nonzero frequency.

*Could there be an interesting analogue in Weyl semimetals?*

## A well-known feature of Weyl fermions: “chiral anomaly”

Chiral anomaly: current conservation is anomalous for a single Weyl fermion coupled to a U(1) gauge field:

$$\partial^\mu J_\mu^W = \frac{g^2 C}{16\pi^2} \mathbf{E} \cdot \mathbf{B}$$

CME version of Nielsen-Ninomiya theorem:  
The total charge of Weyl points in a crystal is 0.

But Weyl points still have interesting consequences. It will turn out that one needs *second order* in EM fields ( $E^2$ ,  $EB$ , for example) for unique Weyl effects...

$$\dot{j}_{2D} = \frac{e^2 E}{h}; \quad \frac{dj_{3D}}{dt} = \frac{e^2 E}{h} \left( \frac{eE}{h} \right) ?$$

(ignore tensor indices for now)

## Outline of CME and GME

Many papers have been written on the possibility of a “chiral magnetic effect” in Weyl semimetals and other materials, also of the form

$$J_i = -\alpha_{ij}^{\text{gme}} B_j$$

This would be related to the chiral anomaly in particle physics, and to the Berry curvature around Weyl points.

Consensus now that it is zero at equilibrium (as “Bloch’s other theorem” says). It can be nonzero at nonzero frequency (non-commutation of  $q \rightarrow 0$  and  $\omega \rightarrow 0$  limits).

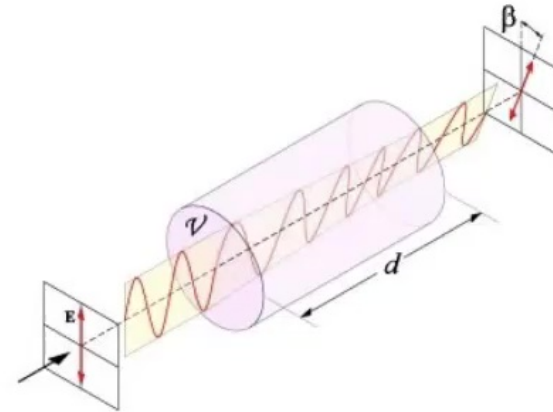
In its simplest form does not involve the Berry phase but something else:

(Zhong, JEM, Souza, PRL 2016; see also Goswami-Sharma-Tewari, Ma-Pesin PRB 2016)

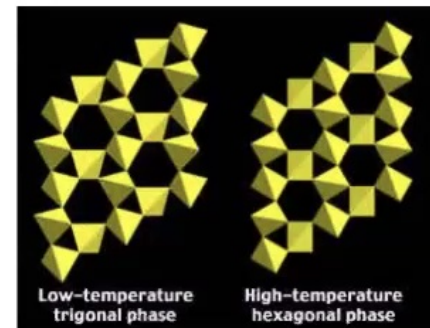
$$\alpha_{ij}^{\text{gme}} = -\frac{1}{(2\pi)^2} \frac{e}{h} \sum_{na} \int_{S_{na}} dS \hat{v}_{F,i} m_{n,j}(\mathbf{k}_f).$$

The *only* linear response CME is actually optical rotation! (“gyrotropic magnetic effect”) It comes from orbital moments of electrons.

Natural optical activity  
or optical gyrotropy:  
like a Faraday effect in a non-  
magnetic material



Occurs in materials with low spatial  
symmetry (intrinsic handedness),  
such as quartz or selenium



## Orbital moment of Bloch electrons

Something that is not always taught (at least by me) in solid state courses is that a Bloch electron has an orbital moment

$$m_{n,j} = \frac{e}{2\hbar} \epsilon_{jln} \text{Im} \langle \partial_l n | H - \epsilon_n | \partial_n n \rangle .$$

This modifies the group velocity that appears in the semiclassical equations:

$$v_{\text{group}} = \frac{1}{\hbar} \nabla_{\mathbf{k}} \epsilon_{\mathbf{k}} = \frac{1}{\hbar} \nabla_{\mathbf{k}} (\epsilon_{\mathbf{k}} + \mathbf{m}_{n\mathbf{k}} \cdot \mathbf{B}).$$

In other words, all the previous pieces found by us and other people come together in the full quantum Kubo formula in a very simple Fermi-surface expression that is pretty easy to calculate in actual materials.

*(This effect is the order- $q$  expansion of conductivity; for a recent paper on the order- $q^2$  term in crystals, see V. Kozii, A. Avdoshkin, S. Zhong, JEM arXiv 2020)*



## Nonlinear effects in metals

The point of the preceding was that the linear response to  $A(q, \omega)$  is not particularly quantized: for two Weyl nodes split in energy by  $E$ , the result is

$$\mathbf{j} = \frac{e^2}{3h^2} (\epsilon_R - \epsilon_L) \mathbf{B}.$$

This is scalar, like the proposed CME, but with a different magnitude. More to the point, it depends on the energy difference between nodes, so isn't quantized.

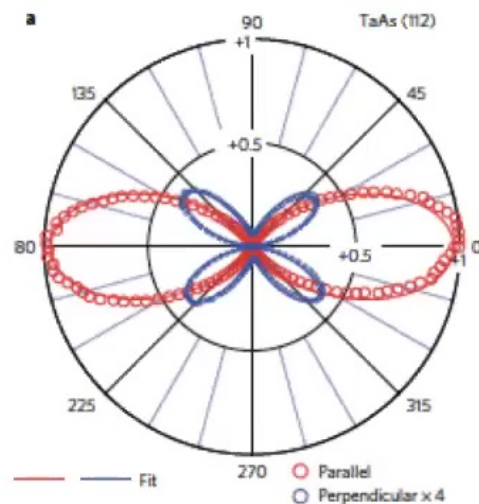
Can we ever see quantized responses in metals? Yes, and even in linear optics...

# Experimental evidence that *nonlinear* effects are interesting in Weyls

(This is in TaAs, the most well-studied Weyl, which has a polar direction; see also work in other talks. Lots of mirror symmetries yield 24 Weyl nodes.)

## Giant anisotropic nonlinear optical response in transition metal monopnictide Weyl semimetals

Liang Wu<sup>1,2\*</sup>, S. Patankar<sup>1,2</sup>, T. Morimoto<sup>1</sup>, N. L. Nair<sup>1</sup>, E. Thewalt<sup>1,2</sup>, A. Little<sup>1,2</sup>, J. G. Analytis<sup>1,2</sup>, J. E. Moore<sup>1,2</sup> and J. Orenstein<sup>1,2\*</sup>



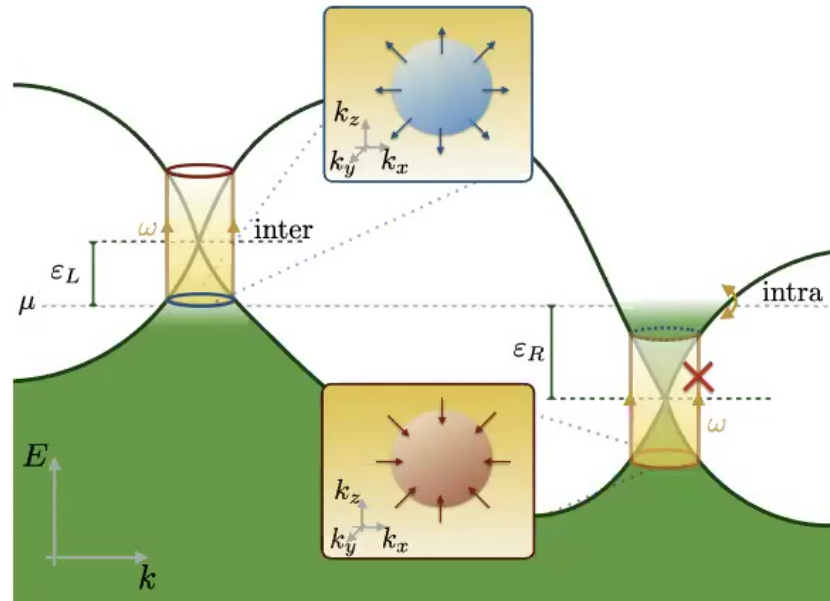
**Table 1 |** Second-harmonic generation coefficients of different materials at room temperature.

Material	$ d_{ij} $	$ d $ (pm V <sup>-1</sup> )	Fundamental wavelength (nm)	Reference
TaAs	$d_{33}$	3600 ( $\pm 550$ )	800	This work
GaAs	$d_{14}$	350*	810	Ref. 11
ZnTe	$d_{14}$	250, 450*	800, 700	Ref. 12
BaTiO <sub>3</sub>	$d_{33}$	15	900	Ref. 24
BiFeO <sub>3</sub>	$d_{33}$	15-19	1550, 800	Refs 26, 27 <sup>†</sup>
LiNbO <sub>3</sub>	$d_{33}$	26	852	Ref. 23
BiFeO <sub>3</sub>	$d_{33}$	130*	500	Ref. 27 <sup>†</sup>
BaTiO <sub>3</sub>	$d_{33}$	100*	170	Ref. 28 <sup>†</sup>
PbTiO <sub>3</sub>	$d_{33}$	200*	150	Ref. 28 <sup>†</sup>

Second-harmonic optical susceptibility can be calculated by  $\chi_{ijk} = 2d_{ij}$ . \*denotes the peak value of the material. <sup>†</sup>denotes theoretical calculation. The uncertainty of  $d_{33}$  in TaAs is determined by setting  $d_{31}$  and  $d_{32}$  in and out of phase with respect to  $d_{33}$  in the fit.

## Quantized CPGE

The quantum calculation of CPGE from a Weyl node gives a surprising result: there is a large quantized value, over a broad range of frequencies, that will dominate metallic contributions from other parts of the Brillouin zone.



(F. de Juan, A. Grushin, T. Morimoto, JEM, Nat. Comm. 2017)

## What is the “quantum”?

$$\frac{1}{2} \left[ \frac{dj_{\odot}}{dt} - \frac{dj_{\ominus}}{dt} \right] = \frac{2\pi e^3}{h^2 c \epsilon_0} I C_n,$$

### Selected pre- and post-history:

Earlier considerations of photocurrent at Dirac points in 2D (e.g., Hosur, PRB 2011).

For Weyls with mirror symmetry, need tilting/bending for an effect:  
C.-K. Chan, N. H. Lindner, G. Refael, P. A. Lee, *Phys. Rev. B* 95, 041104 (2017).

Discovery of a multifold Weyl material RhSi with the right symmetries:  
G. Chang, *et al.* (Hasan), *Phys. Rev. Lett.* 119, 206401 (2017).  
*Should give a factor of 4 enhancement*

Detailed understanding of GME and QCPGE in RhSi:  
F. Flicker *et al.*, *Phys. Rev. B* 98, 155145 (2018).

## Simplified picture of origin of quantization

$$\frac{1}{2} \left[ \frac{dj_{\uparrow}}{dt} - \frac{dj_{\downarrow}}{dt} \right] = \frac{2\pi e^3}{h^2 c \epsilon_0} I C_n,$$

Take the linear-response conductivity from Tabert and Carbotte, PRB 2016:

$$\sigma_1(\omega) = \frac{e^2}{12h} \frac{\omega}{v_F}$$

and imagine that at a Weyl point, every transition changes the current by the *change in velocity* between upper and lower band of the allowed transition.

However, this does not really capture the topological part: band bending, changes in chemical potential, etc. do not affect the result as long as the frequency is in the proper range.

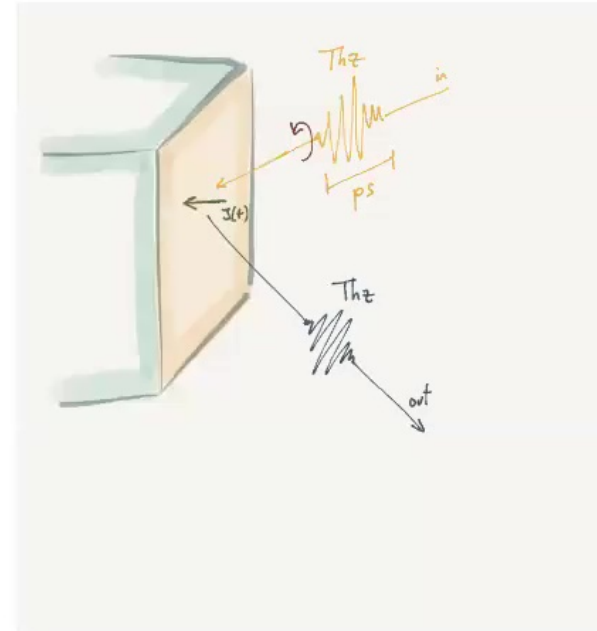
# Experimental detection

Setup in all-optical experiments:

Cubic symmetry in RhSi means that the current direction is into the sample.

The radiated signal from a short pulse comes out in frequencies determined by the pulse envelope, so that the signal becomes DC if the pulse becomes cw.

*Prediction: a plateau in response from low frequency up to around 0.7 eV*

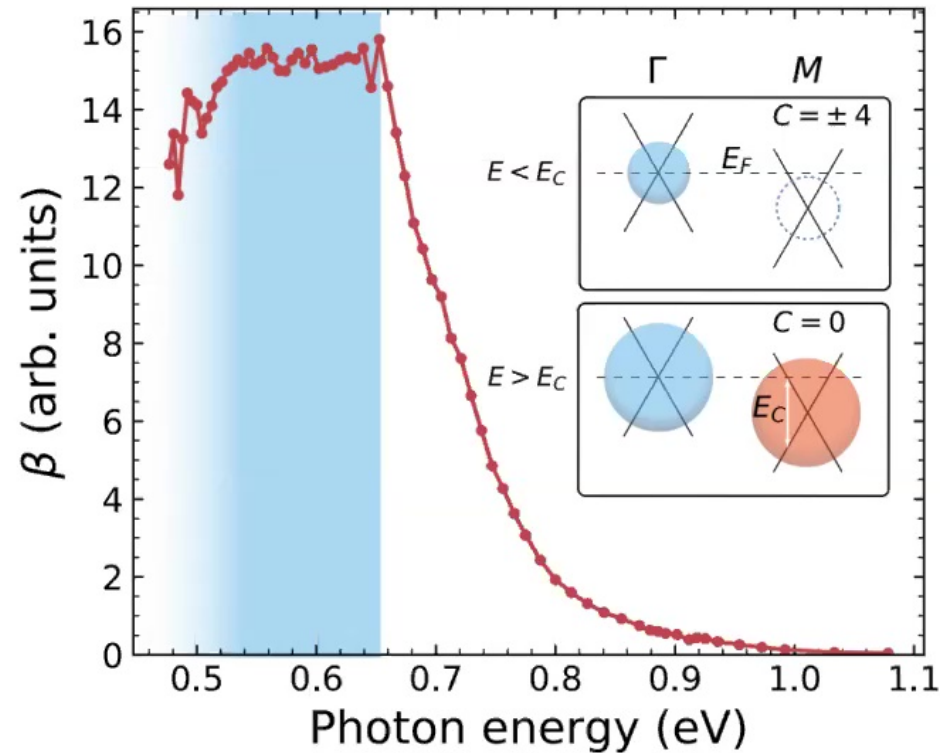


From *Science Advances*, 2020

Quantized Photocurrents in the Chiral Multifold Fermion System RhSi

**Authors:** Dylan Rees, Kaustuv Manna, Baozhu Lu, Takahiro Morimoto, Horst Borrmann, Claudia Felser, J. E. Moore, Darius H. Torchinsky, J. Orenstein

## Experimental detection



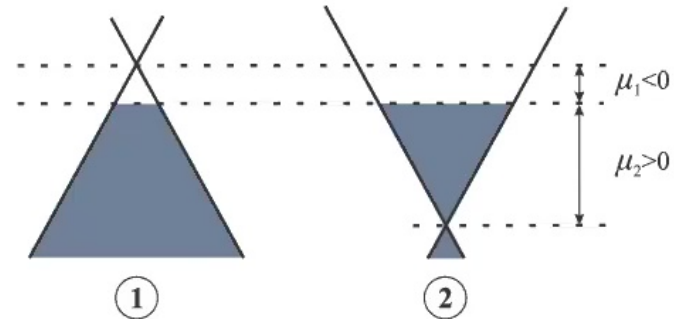
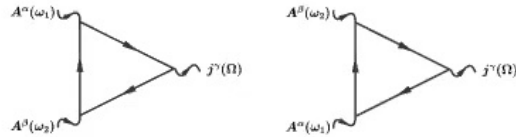
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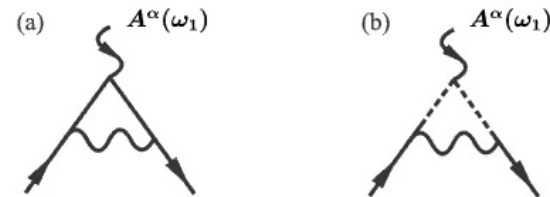
**Authors:** Dylan Rees, Kaustuv Manna, Baozhu Lu, Takahiro Morimoto, Horst Borrmann, Claudia Felser, J. E. Moore, Darius H. Torchinsky, J. Orenstein

# Interaction effects (with A. Avdoshkin, V. Kozii, PRL 2020)

We construct a continuum toy model of two Weyl points. This reproduces the quantization with no band corrections.



1. Add interactions (Hartree or Yukawa type). Self-energy only shifts frequency range.
2. We find that the *vertex correction* is frequency-dependent and regular except at the onset.



In the linear problem in graphene, the calculation is pathological (cutoff-dependent), but most groups agree that the interaction correction is small, in agreement with experiment.

This happens not for any deep reason: the coefficient is  $\frac{19 - 6\pi}{12} \approx 0.013$ .



# Current-induced second harmonic generation in Dirac semimetals

Start from Dirac points protected by both inversion and time-reversal.

Need to break inversion to generate SHG. Can do this by an applied current, as previously done in graphene.

Largely because the material is three-dimensional, the signal can be much larger in graphene, particularly if the material has a long relaxation time. Fortunately, cadmium arsenide is believed to have quite a long relaxation time, and hence a strong signal.

Experimental status:  
LANL group (Prasankumar, Yarotski et al.)  
have shown an effect in TaAs; Cd3As2  
currently underway

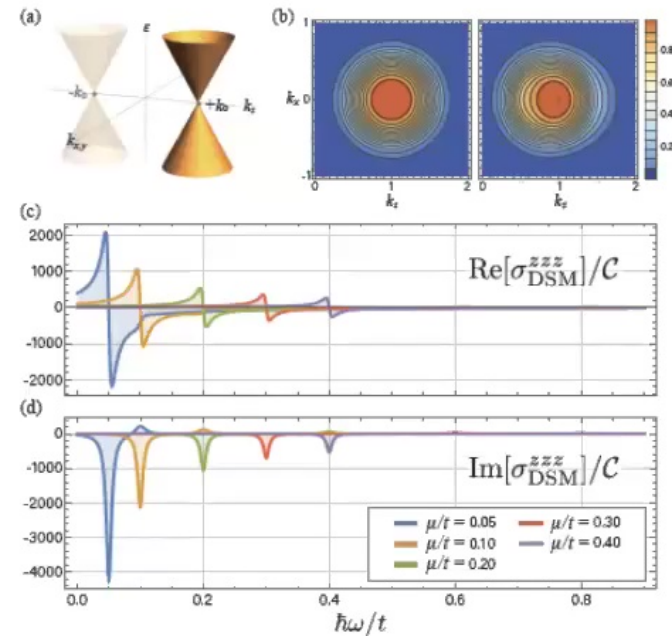


FIG. 2. (a) Energy dispersion of the Weyl Hamiltonian [Eq. (7)] whose gapless point is located at  $\pm k_0 = (0, 0, \pm k_0)$ . (b) Equilibrium distribution function  $f^{(0)}(\varepsilon(k))$  (left) and nonequilibrium distribution function  $f(\varepsilon(k))$  up to the order of  $\mathcal{E}_a^2$  (right) in momentum space ( $k_y$  is fixed to zero). Here,  $\varepsilon(k)$  denotes the larger eigenvalue of the Weyl Hamiltonian and the parameters are set as  $t = 1$ ,  $a = 1$ ,  $\mu = 0.5$ ,  $k_0 = 1.0$ ,  $\beta = 10$ , and  $(\mathcal{E}_x, \mathcal{E}_y, \mathcal{E}_z) = (0, 0, 0.1)$ . (c, d) Real and imaginary part of the CISHG response tensor of Dirac semimetals  $\sigma_{\text{DSM}}^{zzz}(\omega)$  calculated with the Weyl Hamiltonian. The values of the vertical axes are normalized by a constant  $C = (e\tau Ea/\hbar) \cdot (\hbar/t) \cdot (e^3/h^2)$ . Here, we set  $\gamma = 0.01(t/\hbar)$ .

# Conclusions

Nonlinear optics has unique features in Weyl semimetals.  
There is a decent range of frequencies where

(a) in the "conventional" Weyl material TaAs, second-order responses (SHG and photocurrent) are extremely strong;

(b) in the mirror-free multifold Weyl RhSi, chiral photocurrent (the CPGE) is quantized, with a small correction from other bands, and probably an additional correction from interactions like graphene.

“Integer” Quanta in electromagnetic response:

$e$

$e/h$ :  
Josephson frequency;  
(inverse) flux  
quantization

$e^2$

$e^2/h$ : Quantum Hall;  
point contact; QSHE;  
magnetoelectric effect

$e^2/(hc)$ : Graphene  
optical transmission

$e^3$

$e^3/h^2$ : QCPGE in mirror-  
split Weyl semimetals