Contemporary physics: Lecture 1

Nonlinear Optics In Quantum Materials

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My research:
Theoretical condensed matter physics, *focusing on predicting material properties, with an emphasis on nonlinear optics and strain physics in quantum materials.*

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Outline

What are quantum materials?

Introduction: from early theory of metals to insulators
Quantum materials
Topological materials: graphene, topological insulators, TMDs, Weyl semimetals

Why does nonlinear optics matter?

Introduction to nonlinear optics
High harmonic and photocurrent generations
Sum and difference frequency effects
Self-focusing and optical switching
Multiphoton absorption

Nonlinear optics in topological materials

Example: Nonlinear charge and spin Hall photocurrent in WTe2

From Stone age to Quantum age

Stone age

Bronze and Iron ages

- Ancient Civilizations
- Industrial Revolution

Silicon age

- Quantum mechanics
- Transistor
- Computer

Information age

- Personal computer
- World Wide Web (Internet)
- AI

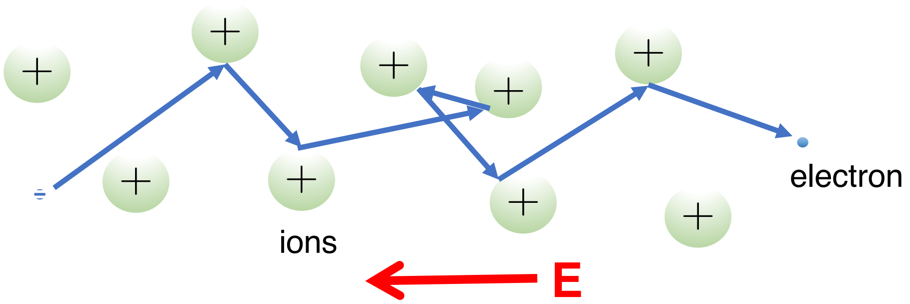
Quantum age

- Quantum Materials
- Quantum Computer
- Quantum Communication
- Quantum Sensing

Drude's model for metals (in 1900)

- Core electrons are bound to the core of atom
- Valence electrons freely move and don't belong to a particular atom

Trajectory of a conduction electron scattering off the ions according to Drude



Paul Drude
(1863-1906)

Mean free path: average distance that electron travels between consecutive collisions

According to Drude: Mean free path ~ interatomic distance

Mean free path $\sim 100 - 1000\text{Å} \gg$ interatomic distance

Element		Crystal structures	$\rho_{o,rt}$ ($\mu\Omega$ cm)	v_f (10^5 m/s)	$\tau \times \rho_o$ (10^{-22} Ω m s)	$\lambda \times \rho_o$ (10^{-16} Ω m^2)	τ_{rt} (fs)	λ_{rt} (nm)
Silver	Ag	fcc	1.587	14.48	5.84	8.46	36.8	53.3
Copper	Cu	fcc	1.678	11.09	6.04	6.70	36.0	39.9
Gold	Au	fcc	2.214	13.82	6.04	8.35	27.3	37.7

D. Gall, J. Appl. Phys.119, 085101 (2016)

Reminder: Electron as a wave

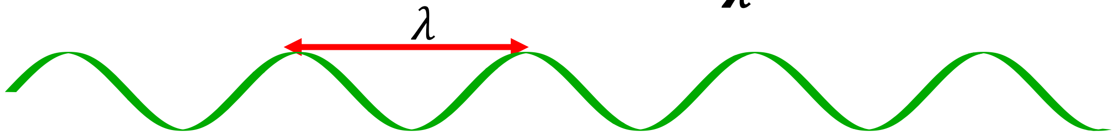
Louis de Broglie
(1892-1987)

In 1924, PhD thesis: all matter has wave properties

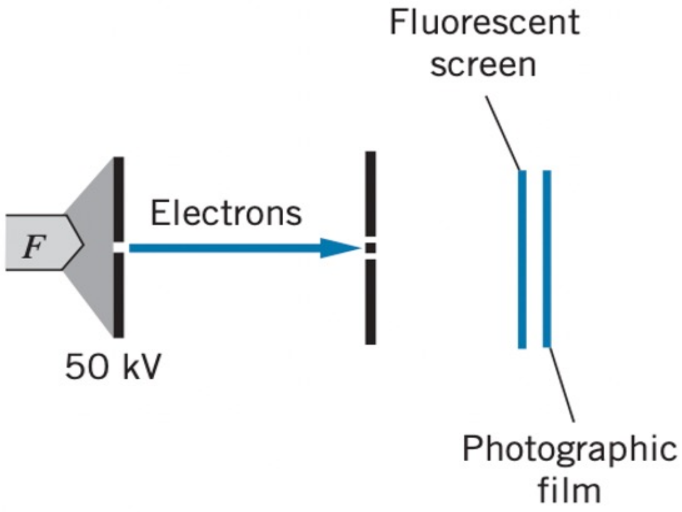
de Broglie wavelength: $\boldsymbol{\lambda = \frac{h}{p} = \frac{h}{m\,v}}$

Planck's constant: $h \approx 6.6{\times}10^{-34}\ \mathrm{J.s}$

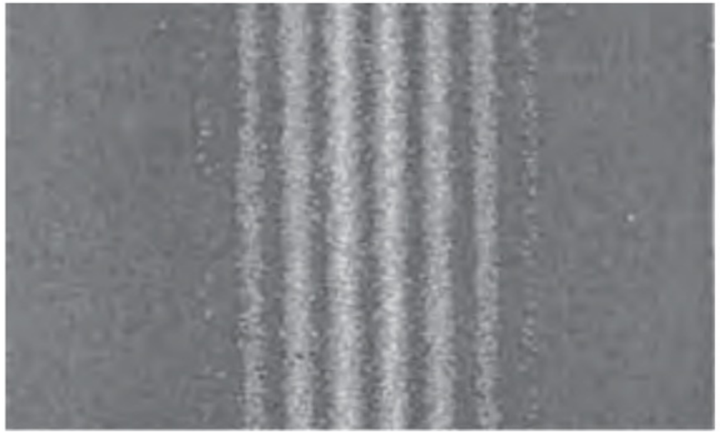
Wave vector: $\boldsymbol{k = \frac{2\pi}{\lambda}}$ $\quad \boldsymbol{p = \hbar k}$ $\quad \hbar = \frac{h}{2\pi}$



Because of the smallness of "*h*", only for particles of atomic or nuclear size the wave behavior will be observable



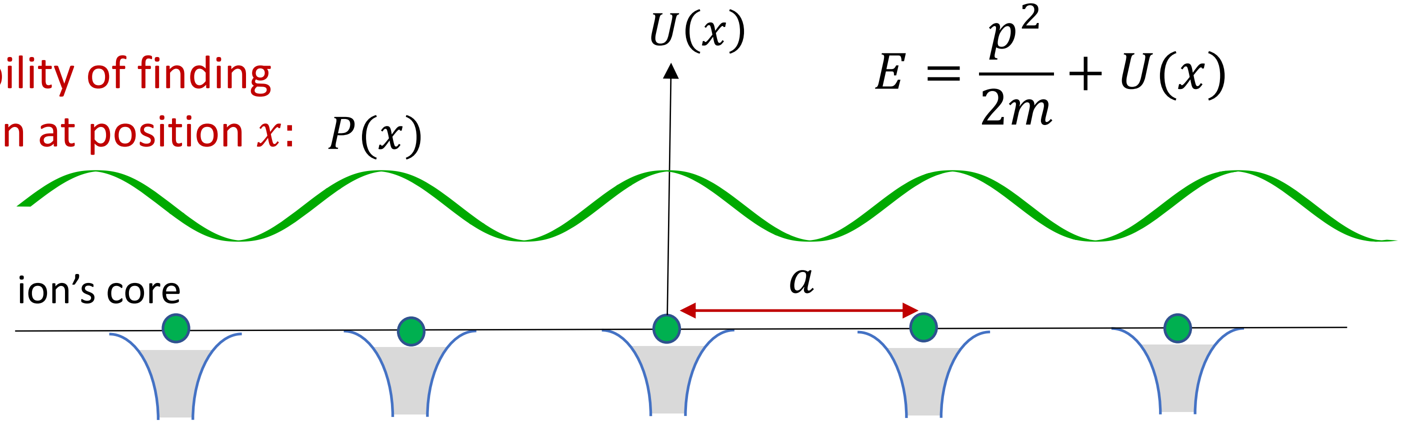
The first double-slit experiment with electrons was done in 1961



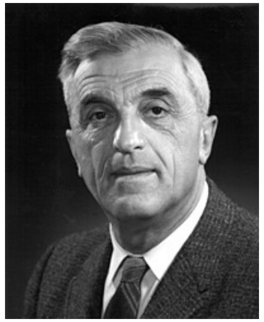
Electrons in periodic potential

Probability of finding electron at position x: $P(x)$

$$E = \frac{p^2}{2m} + U(x)$$



$$\frac{(-e)(+eZ)}{|\vec{r}_{ion} - \vec{r}_e|}$$



Felix Bloch
(1905-1983)

$U(x+R) = U(x)$ $\quad R = na \quad$ with $\quad n \in \mathbb{Z}$

$\rightarrow P(x+R) = P(x)$

Let us define:

$P(x) = |\psi(x)|^2$ $\quad \rightarrow \psi(x+R) = e^{ikR}\,\psi(x)$ $\quad$ Phase factor: $|e^{ikR}| = 1$

exercise: $\rightarrow \psi(x) = e^{ik\,x} u(x)$ $\quad$ with $\quad u(x+R) = u(x)$

Free electron wave function (U=0):

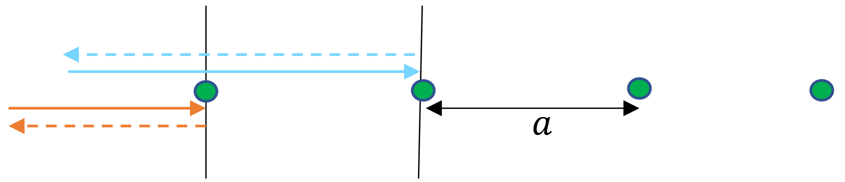
$u(x) = \text{constant} \rightarrow$ $\quad \psi(x) = \frac{1}{\sqrt{L}} e^{ik\,x}$

The "k" is the electron wave vector:

$$k = \frac{2\pi}{\lambda} = \frac{p}{\hbar}$$

Electron Bragg scattering → Energy forbidden gap

Bragg scattering of electrons in 1D lattice



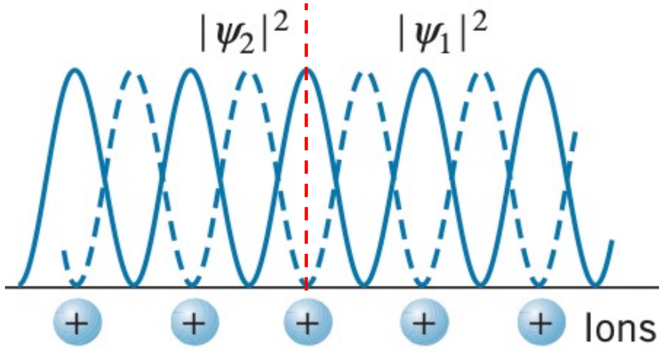
$$2a = n\lambda \text{ where } n = 1,2,\ldots$$

Constructive interference

Bragg condition

$$k = \frac{2\pi}{\lambda} = \frac{n\pi}{a}$$

For "k" that satisfy the Bragg condition, the reflected and incident waves add to produce standing waves

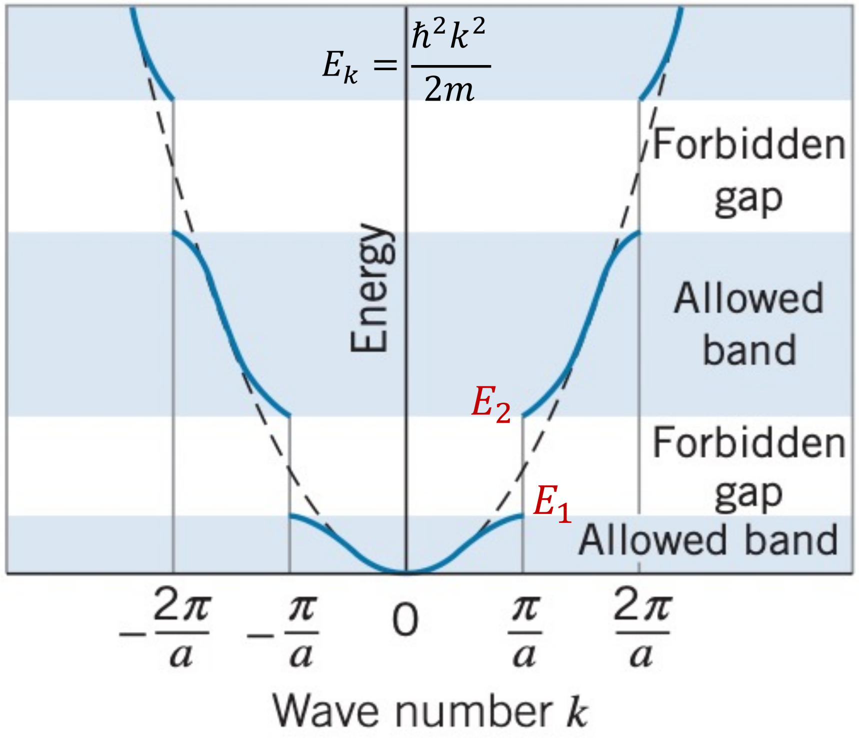


$$\psi_{\pm}(x) = \frac{1}{\sqrt{L}} e^{\pm ik\,x} \rightarrow$$

$$\psi_1 \sim \psi_+ + \psi_- \sim \cos kx$$

$$\psi_2 \sim \psi_+ - \psi_- \sim \sin kx$$

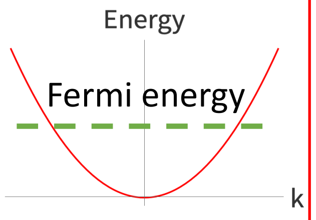
Electrons in a weak periodic potential



Conductor

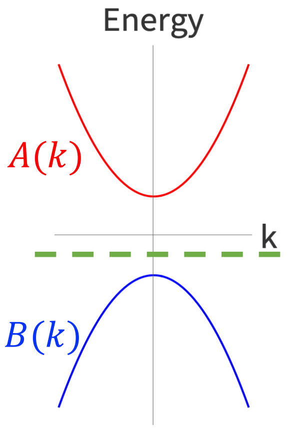
$$E = \frac{\hbar^2 k^2}{2m}$$

$$\psi \sim e^{i\,\boldsymbol{k}\cdot\boldsymbol{r}}$$



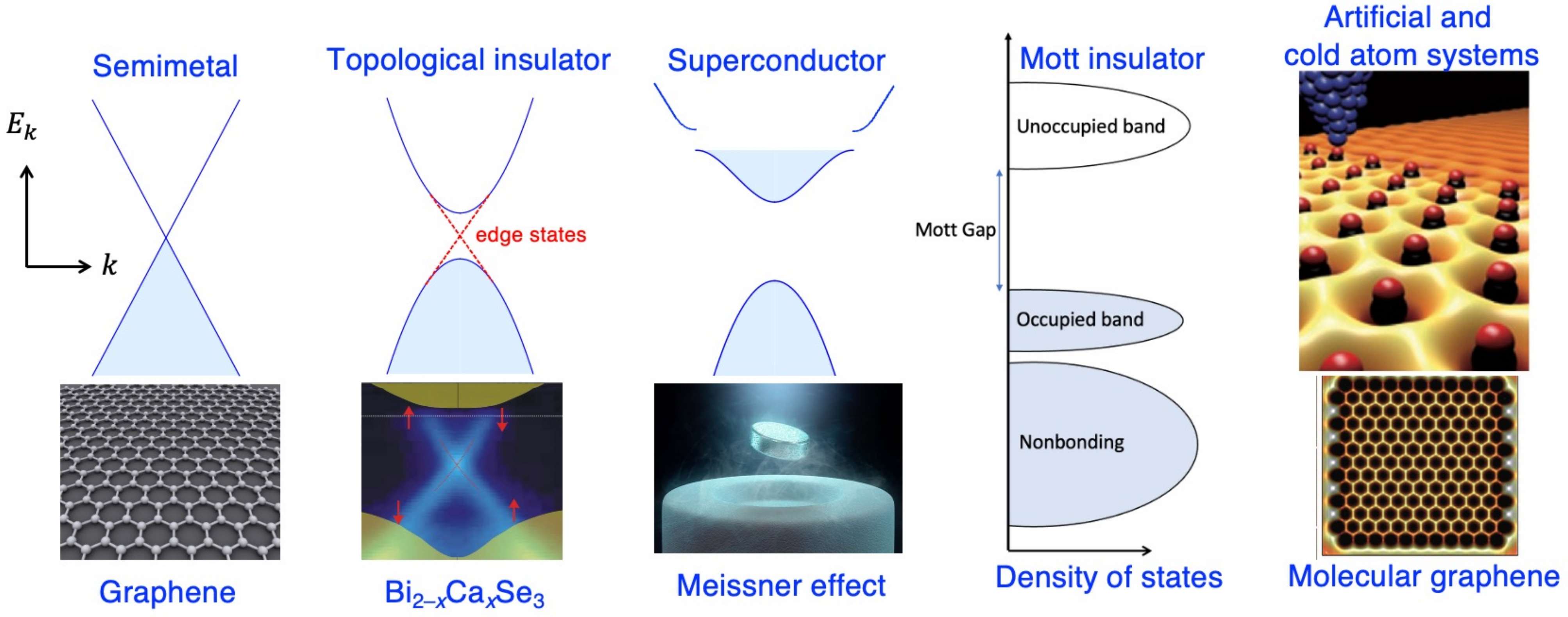
Insulator

$$\psi \sim \begin{bmatrix} A(k) \\ B(k) \end{bmatrix} e^{i\,\boldsymbol{k}\cdot\boldsymbol{r}}$$



Quantum Materials

Quantum materials reveal properties with nontrivial topology, coherence, entanglement, and many-body correlations



Quantum Materials

Many-body correlation

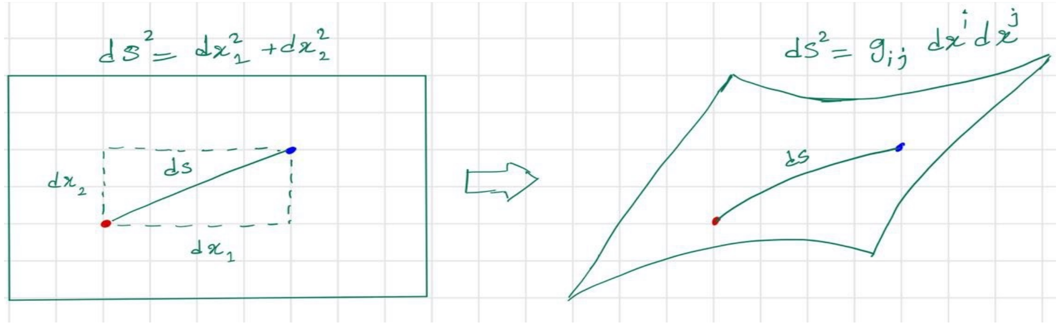
Emergence (symmetry breaking)

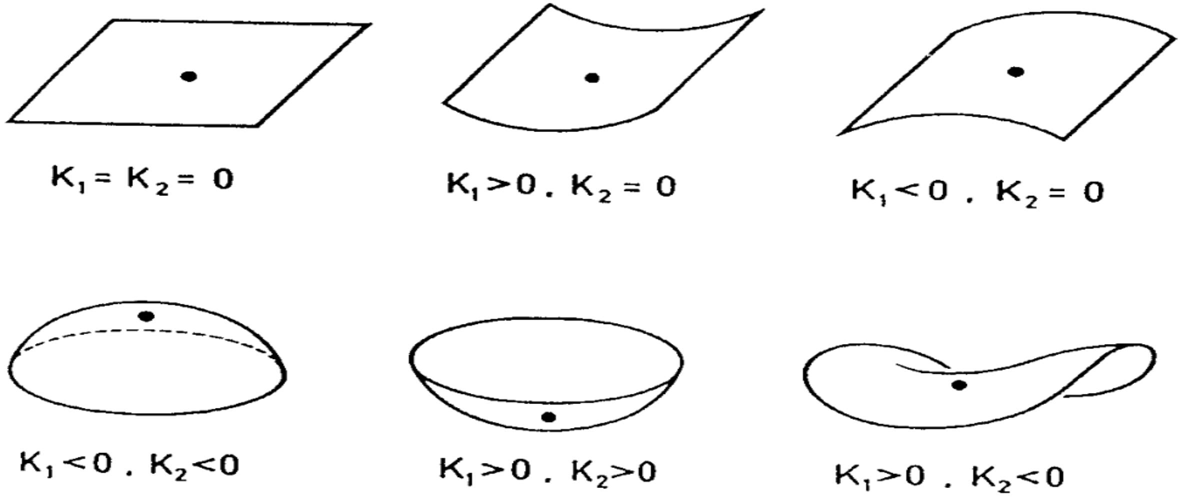
Topological entanglement

Recall: curvature and metric

Metric tensor g

$$ds^2 = g_{ij}dx^i dx^j$$





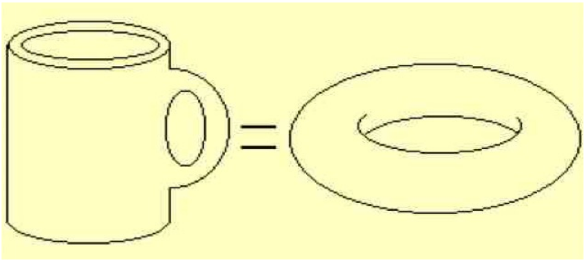
Gaussian Curvature

$$\Omega = K_1 K_2 = \frac{1}{R_1 R_2}$$

Geometry

Mean curvature

$$H = \frac{K_1 + K_2}{2} = \frac{1}{2}\left(\frac{1}{R_1} + \frac{1}{R_2}\right)$$



Gauss–Bonnet theorem

Topology

$$\frac{1}{2\pi}\int_S d\sigma\, \boldsymbol{\Omega} = 2(1-g),$$

Berry curvature and quantum metric

$$H(\boldsymbol{k})|\psi_n(\boldsymbol{k})\rangle = E_n(\boldsymbol{k})|\psi_n(\boldsymbol{k})\rangle$$

Distance between quantum states

$$S^2(\boldsymbol{k}_1, \boldsymbol{k}_2) = 1 - |\langle\psi_0(\boldsymbol{k}_1)|\psi_0(\boldsymbol{k}_2)\rangle|^2$$

Quantum metric

$$dS^2 = g_{ij}(\boldsymbol{k}) dk^i dk^j$$

$$g_{ij}(\boldsymbol{k}) = \mathrm{Re}[\langle\partial_i\psi_0|\,\hat{Q}(\boldsymbol{k})|\partial_j\psi_0\rangle]$$

$$\hat{Q}(\boldsymbol{k}) = 1 - |\psi_0(\boldsymbol{k})\rangle\,\langle\psi_0(\boldsymbol{k})|$$

Berry curvature

$$\Omega_{ij}(\boldsymbol{k}) = -2\mathrm{Im}\left[\langle\partial_i\psi_0|\,\hat{Q}(\boldsymbol{k})|\partial_j\psi_0\rangle\right] = \partial_i A_j(\boldsymbol{k}) - \partial_j A_i(\boldsymbol{k})$$

Berry phase (Geometric phase)

$$\psi \rightarrow \psi e^{i\gamma}$$

$$\gamma = \oint \boldsymbol{A}(\boldsymbol{k}) \cdot d\boldsymbol{k} = \int \boldsymbol{\Omega}(\boldsymbol{k}) \cdot \hat{\boldsymbol{n}}\, d\sigma = \frac{1}{2}\int dk^i dk^j\, \Omega_{ij}$$

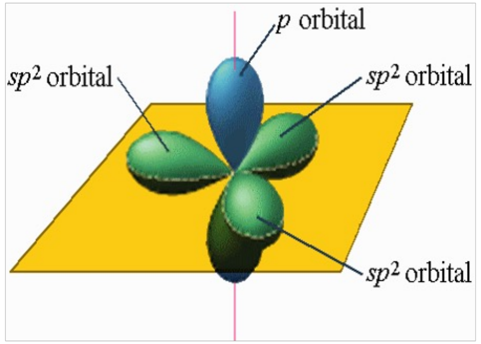
Topological state

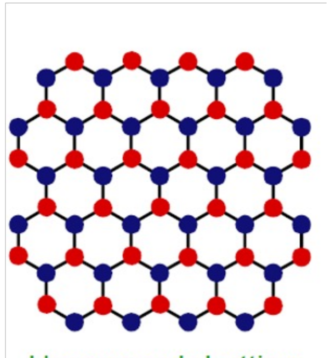
$$\gamma \neq 2n\pi = 0, \pm 2\pi, \pm 4\pi \ldots$$

Graphene

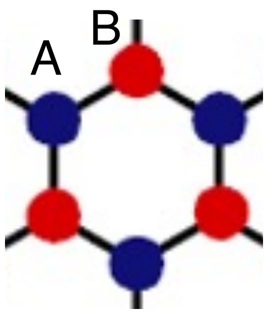
Carbon atom : $1s^2 2s^2 2p^2$

: $(two) 1s^2 + (three) sp^2 + (one) 2p_z$

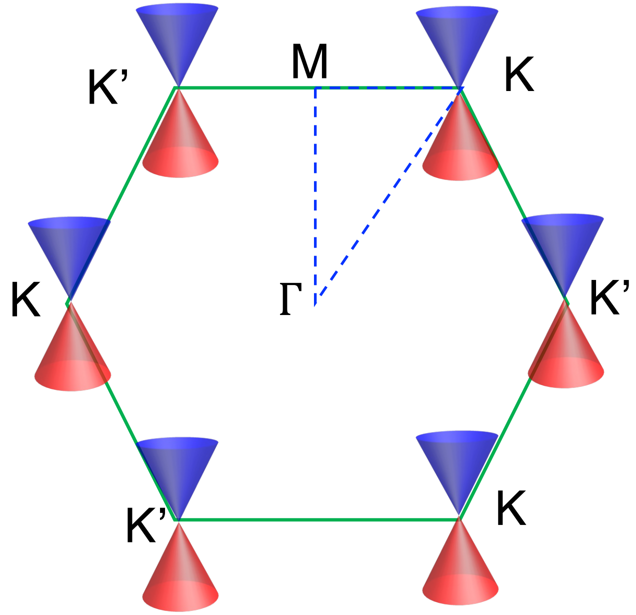




sublattice



Dirac cone



Conventional metal

$$H = \frac{p^2}{2m}$$

Berry phase =0

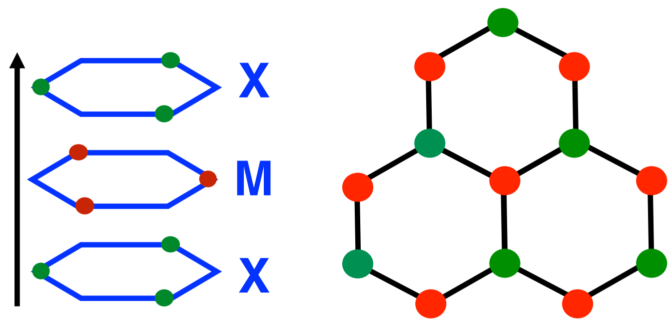
Dirac metal

$$\hat{H} = v(p_x\hat{\sigma}_x + p_y\hat{\sigma}_y) = v\begin{pmatrix} 0 & p\,e^{-i\theta} \\ p e^{i\theta} & 0 \end{pmatrix} \qquad |\psi_\pm\rangle = \frac{1}{\sqrt{2}}\begin{pmatrix} 1 \\ \pm e^{i\theta} \end{pmatrix} e^{ip\cdot r}$$

$$E = vp$$

Berry phase = π

Transition metal dichalcogenides

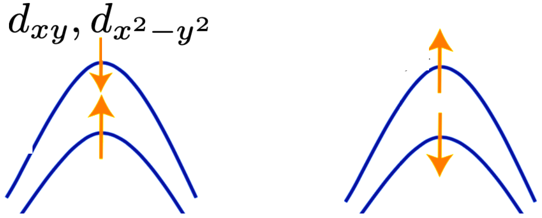


M: Mo, W

X: S, Se

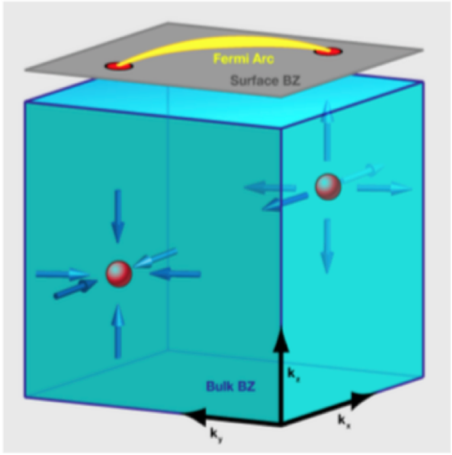
Non-zero Berry curvature

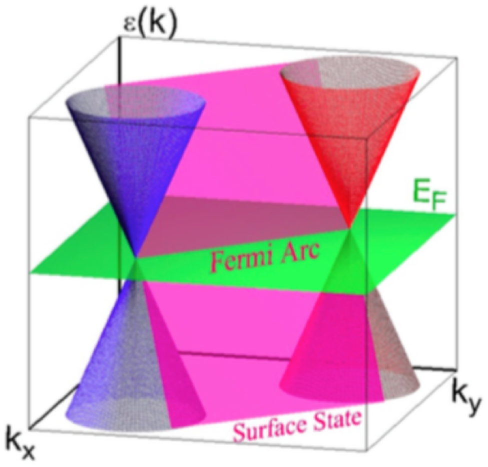
Spin-valley entanglement



Weyl semimetals

Example: TaAs, NbP



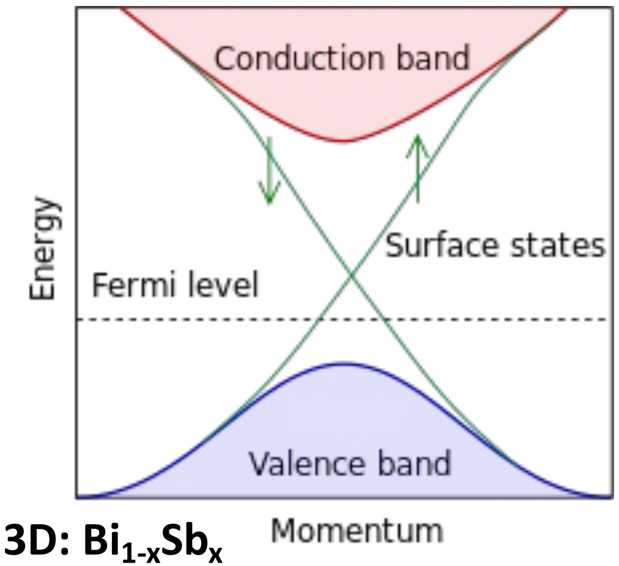


Monopole
Berry curvature

$$\boldsymbol{\Omega}^{(\chi)}(\boldsymbol{p}) = \frac{\chi}{2}\frac{\boldsymbol{p}}{p^3}$$

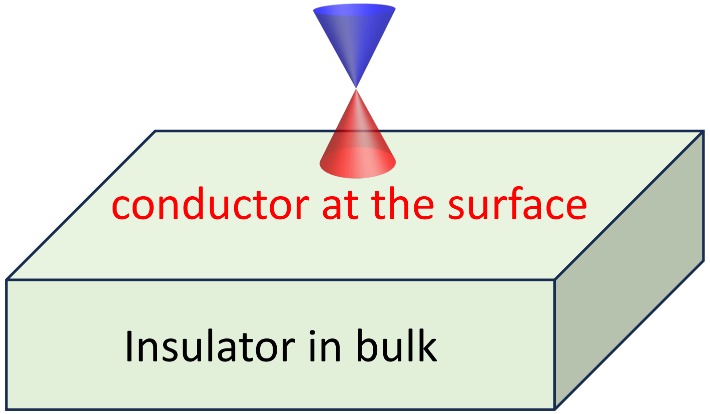
Topological insulator

From Wikipedia



3D: $Bi_{1-x}Sb_x$

2D (quantum spin Hall): HgTe, WTe2

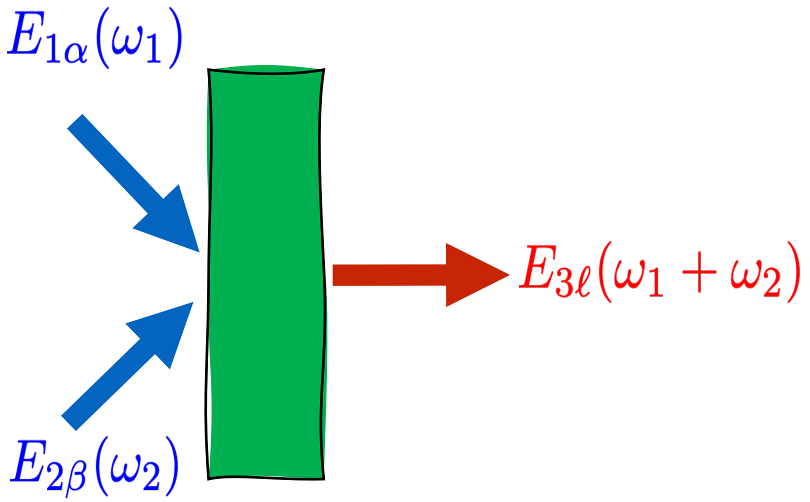


Why nonlinear optics?

Application importance

- High-Harmonic Generations
- Saturable Absorbance
- All-Optical Modulators
- Optical Amplifications
- Optical Fibers
- Harvesting Solar Energy
- Quantum computing

Multi-photon physics



Fundumental importance

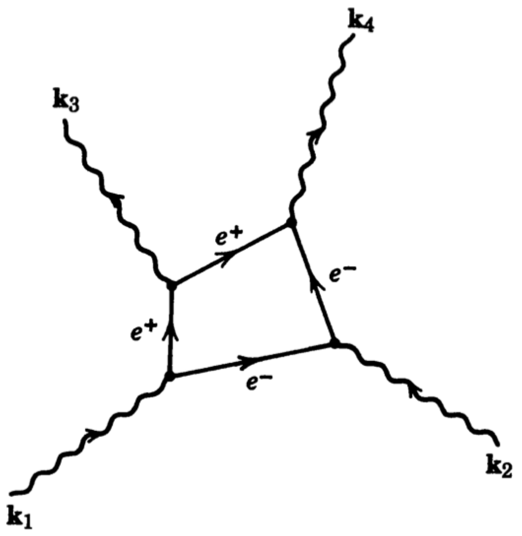
- Light-Light scattering and multi-photon physics
- Nonlinear spectroscopy: hidden symmetry/topology
- Perturbative modelling of non-equilibrium state

$$\langle ABC \ldots \rangle$$
$$G(t_1, t_2; \mathbf{A})$$

Nonlinear optics

Nonlinear optics in vacuum is a result of photon-photon interaction

$$\epsilon_{ik} = \delta_{ik} + \frac{e_G^4 \hbar}{45 \pi m^4 c^7} \left[2(E^2 - c^2 B^2)\delta_{ik} + 7\, c^2 B_i B_k \right] + \cdots$$



electron-positron mediated photon-photon scattering

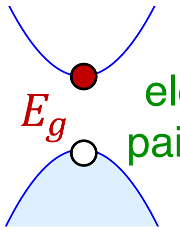
Vacuum becomes nonlinear beyond a threshold (**Schwinger limit**)

$$E_c = \frac{1}{e} \frac{\text{rest energy} = m_e c^2}{\text{Compton wavelength} = \hbar / m_e c} \sim 10^{18} \mathrm{V/m}$$

→ Linear Maxwell's equations in free space

Analogous threshold in materials is **Landau-Zener limit:**

$$E_c = \frac{1}{e} \frac{\text{bandgap} = E_g}{\text{mean free path} = \ell_e} \sim 10^{8}\ \mathrm{V/m}$$

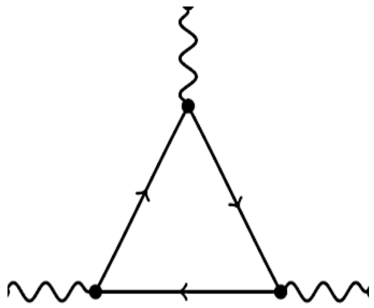


electron-hole pair production

Further nonlinearity in the pertubative regime:

$$P(E) = \chi^{(1)} E + \chi^{(2)} EE + \chi^{(3)} EEE + \cdots$$

Finite only in inversion-broken systems



Second-order photon-photon interaction in parity-breaking materials

Mechanisms of nonlinear optics

- Anharmonicity of potential

$$V(x) = \frac{1}{2}\omega_0^2 x^2 + \frac{1}{3} a\, x^3 + \frac{1}{4} b\, x^4 + \cdots$$

- Hydrodynamics

$$\frac{d\vec{v}}{dt} + \vec{v}\cdot\vec{\nabla}\vec{v} = -\frac{e}{m}\left(\vec{E} + \frac{1}{c}\vec{v}\times\vec{B}\right)$$

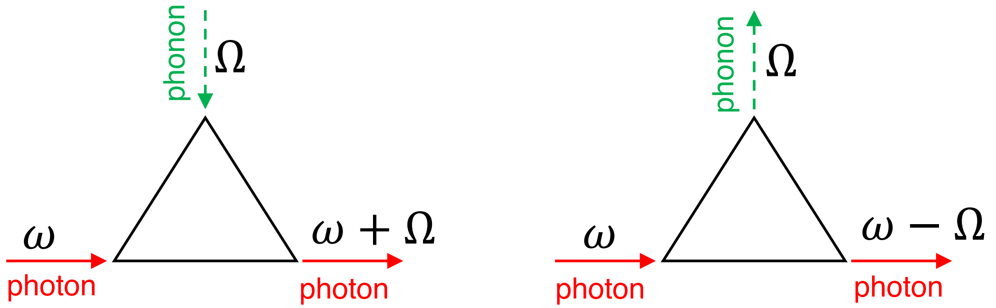
- Thermal mechanism

$$\epsilon_{ij}(T_\mathrm{i}, T_\mathrm{e}) \rightarrow \delta T_\mathrm{i} \propto E^2,\ \delta T_\mathrm{e} \propto E^2$$

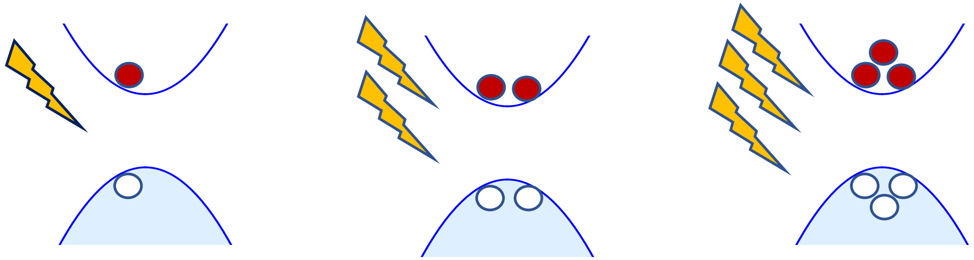
- Orientational mechanism

$$U = \frac{1}{2}\alpha_{ij} E_i E_j \rightarrow \mathrm{P} \propto e^{\Delta\alpha \frac{E^2}{T}} \rightarrow \delta\epsilon_{ij} \propto E^2$$

- Inelastic photon scattering



- Phase-space filling (Saturable absorption)



reduced avilable phase-space

Symmetry and nonlinear response

$$\vec{P} = \epsilon_0 \chi^{(1)} \cdot \vec{E} + \epsilon_0 \chi^{(2)} : \vec{E}\vec{E} + \epsilon_0 \chi^{(3)} \vdots \vec{E}\vec{E}\vec{E} + \cdots \quad 1$$

$$P_a = \epsilon_0 \chi_{ab}^{(1)} E_b + \epsilon_0 \chi_{abc}^{(2)} E_b E_c + \epsilon_0 \chi_{abcd}^{(3)} E_b E_c E_d + \cdots$$

Spatial inversion symmetry

$$I\,\vec{P}\,I^{-1} = -\vec{P}$$

$$I\,\vec{E}\,I^{-1} = -\vec{E} \quad \Rightarrow \quad -\vec{P} = -\epsilon_0 \chi^{(1)} \cdot \vec{E} + \epsilon_0 \chi^{(2)} : \vec{E}\vec{E} - \epsilon_0 \chi^{(3)} \vdots \vec{E}\vec{E}\vec{E} + \cdot \quad 2$$

$$I\,\chi^{(n)}\,I^{-1} = \chi^{(n)} \quad \Rightarrow \quad \chi^{(2)} = 0$$

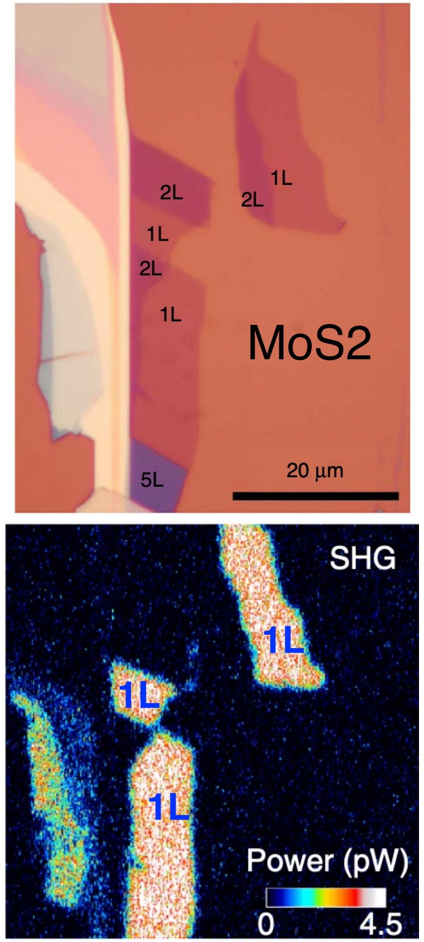
Discrete crystal symmetry

Rank-n tensorial transformation: $\tilde{\chi}^{(n)} = S^{\dagger} \chi^{(n)} S S \ldots S$

For example, rotation tensor in 2D: $S(\theta) = \begin{bmatrix} \cos\theta & \sin\theta \\ \sin\theta & -\cos\theta \end{bmatrix}$

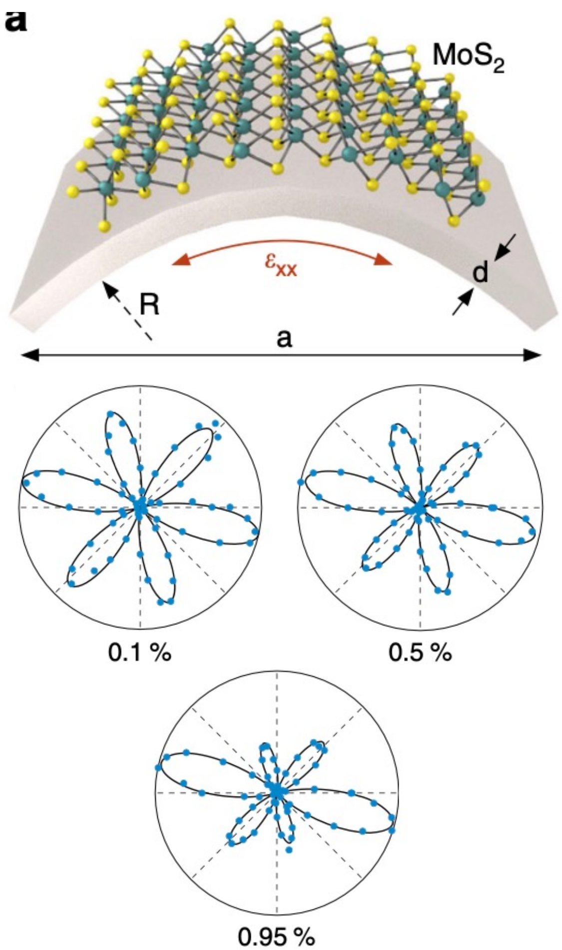
Nonlinear spectroscopy

Nonlinear crystal imaging



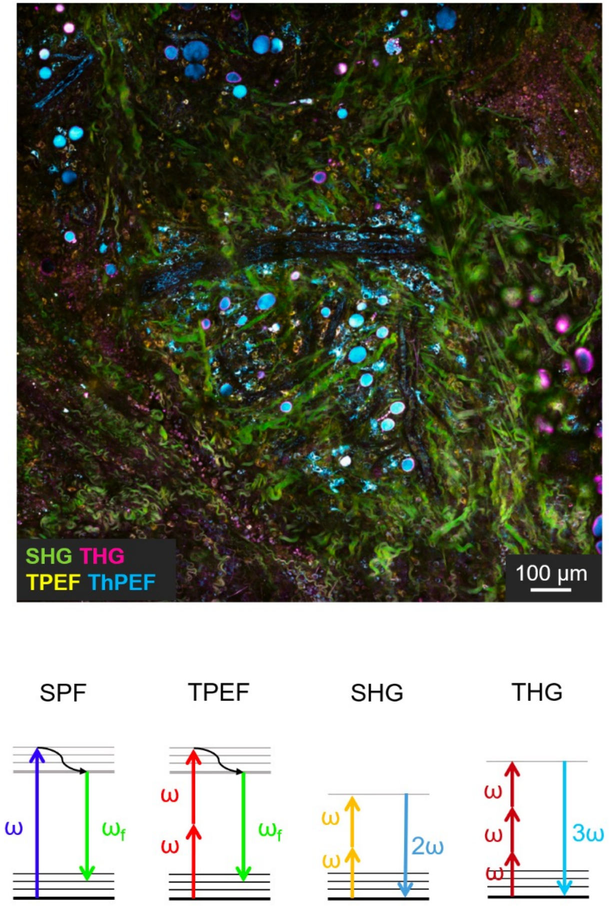
Nature Communications 8, Article number: 893 (2017)

Strain measurement



Nature Communications 9, Article number: 516 (2018)

Biomedical imaging



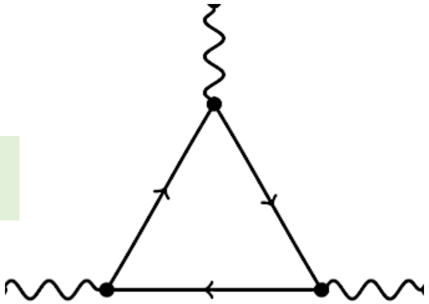
Nature Communications 9, Article number: 2125 (2018)

The Zoo of Nonlinear Effects

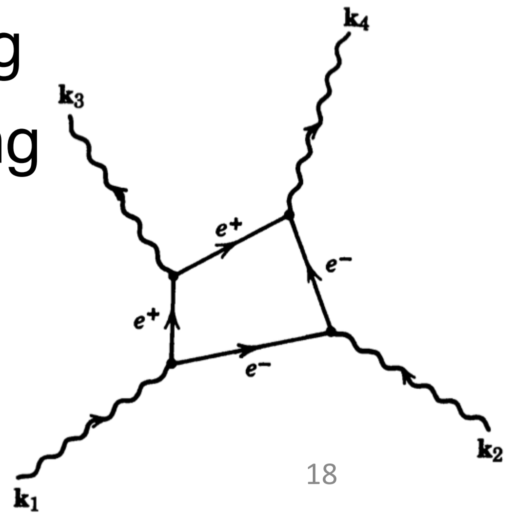
- Second-harmonic generation
- Photogalvanics effect (optical rectification effect)
- Pockels effect (electrooptic effect)
- Sum and difference frequency
- Optical parametric amplification
- Parametric down-conversion (quantum effect)

- Third-harmonic generation
- d.c. Kerr effect
- d.c.-induced second-harmonic generation
- Optical Kerr effect (optically-induced birefringence)
- Two-photon absorption
- General four-wave mixing
- Raman/Brillouin scattering

Three-photon process

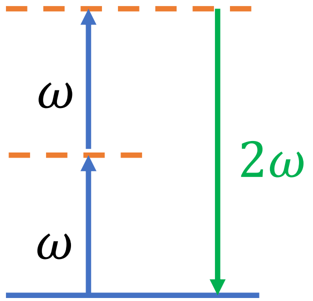


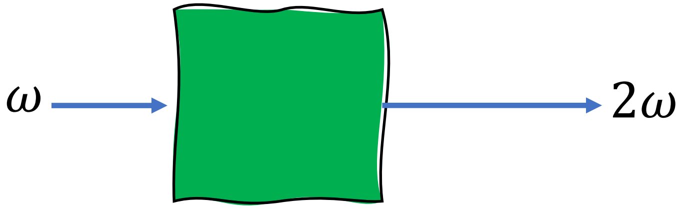
Four-photon process



Second-harmonic generation

$$P_a^{(2)}(2\omega) = \chi_{abc}^{(2)} E_b(\omega) E_c(\omega)$$





- ❑ Green laser
- ❑ Infrared single-photon detector
- ❑ Biological imaging
- ❑ Crystallography
- ❑ Strain measurement

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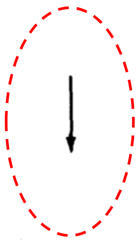
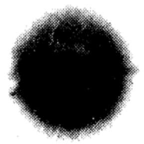
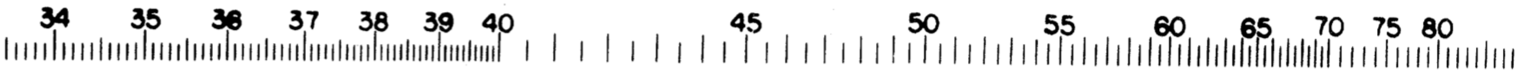


Generation of Optical Harmonics

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
Phys. Rev. Lett. **7**, 118 – Published 15 August 1961





SHG in single crystal of quartz

Optical rectification

$$P_a^{(2)}(0) = \chi_{abc}^{(2)} E_b(\omega) E_c(-\omega)$$

$\omega \rightarrow$ static polarization

$$J_a^{(2)}(0) = \sigma_{abc}^{(2)} E_b(\omega) E_c(-\omega)$$

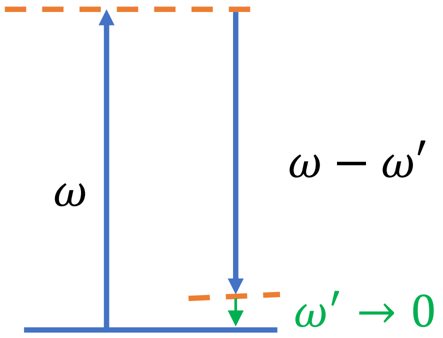
$\omega \rightarrow$ Dc current

$$M_a^{(2)}(0) = \lambda_{abc}^{(2)} E_b(\omega) E_c(-\omega)$$

$\omega \rightarrow$ static magnetization

$$F_a^{(2)}(0) = \pi_{abc}^{(2)} E_b(\omega) E_c(-\omega)$$

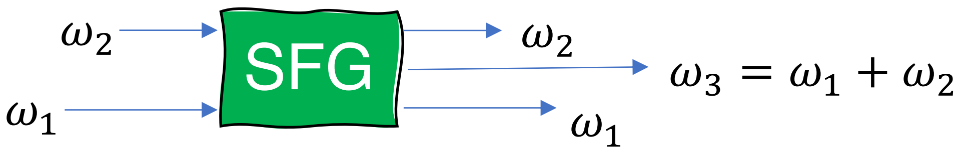
$\omega \rightarrow$ static force

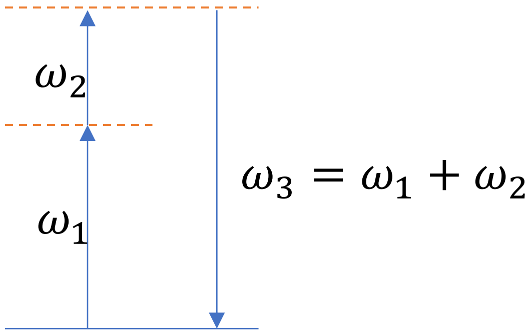


- Photodetection
- Photogalvanic effect
- Inverse Faraday effect
- Displaciv Raman force

Sum and difference frequency effects

Sum-frequency generation

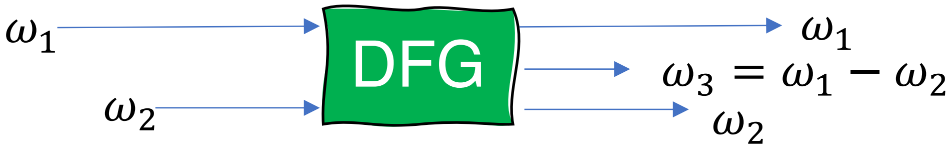


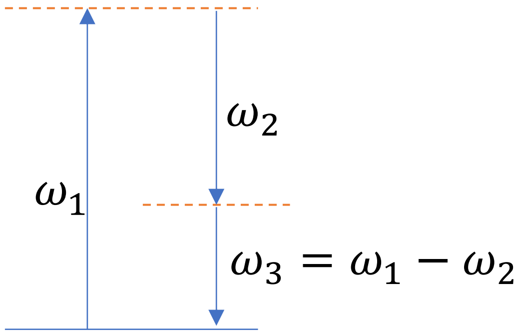


infrared detection

$$\omega_{\mathrm{IR}} \rightarrow \omega_{\mathrm{IR}} + \omega_{\mathrm{pump}}$$

Difference-frequency generation





optical amplification

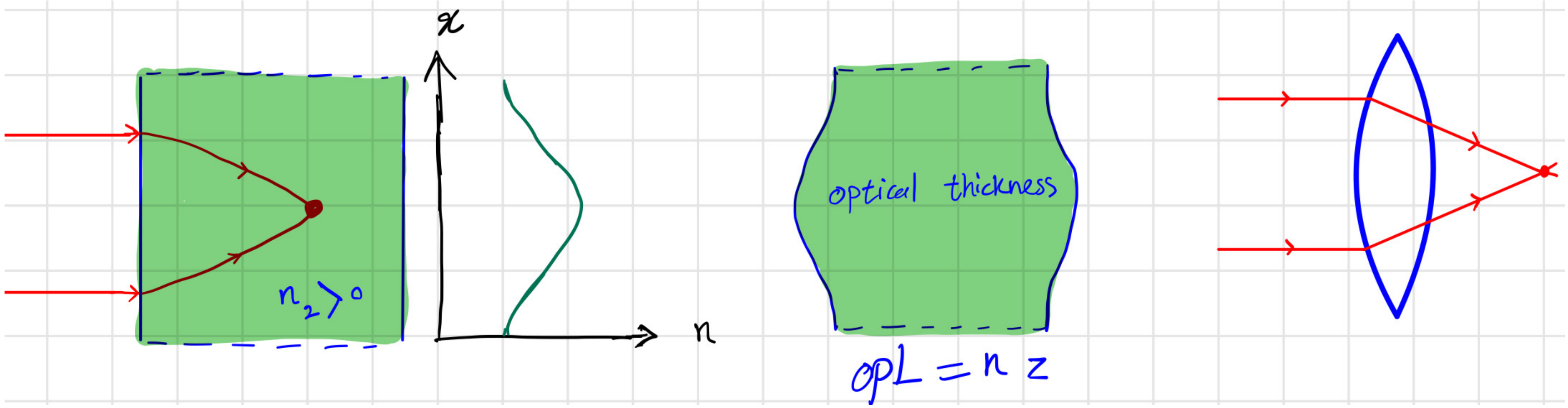
$$I_2 \rightarrow I_2\, e^{\alpha\, I_1\, L}$$

Self-focusing and optical switching

$$\epsilon(\omega) = \epsilon_0\left(1 + \chi^{(1)} + \chi^{(3)}_{\mathrm{Kerr}}|E(\omega)|^2\right)$$

Nonlinear refractive index:

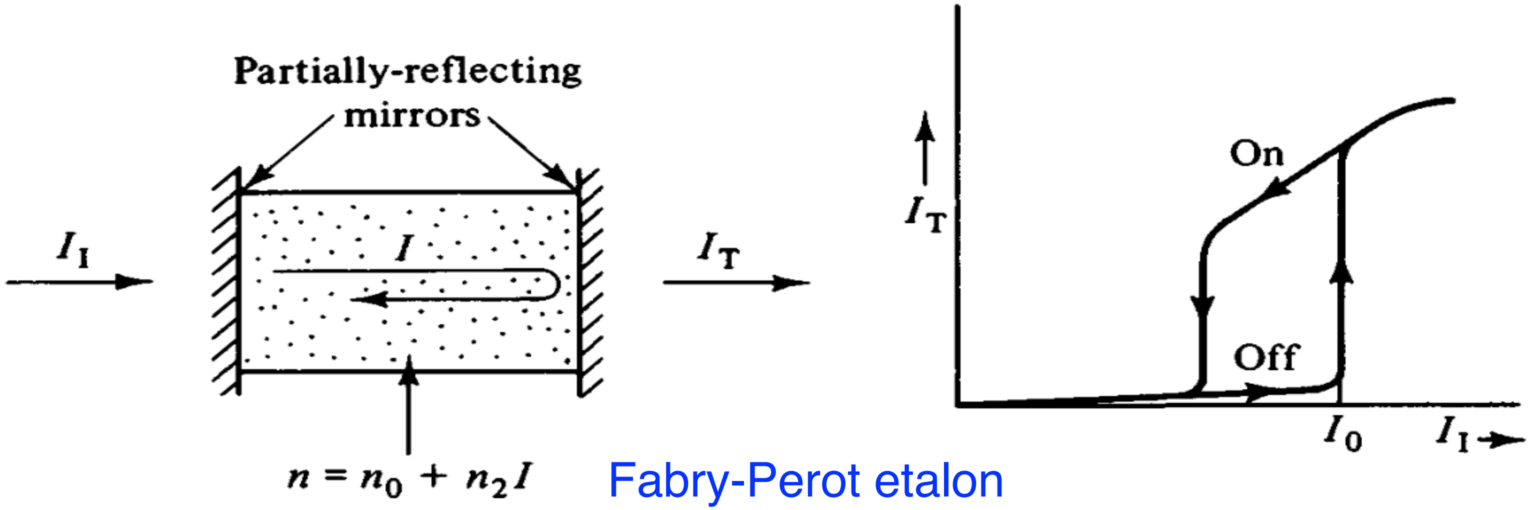
$$n = n_0 + n_2 I \qquad n_2 \sim \mathrm{Re}[\chi^{(3)}_{\mathrm{Kerr}}]$$



Optical switching

$$\frac{2nd}{\lambda} = \frac{2(n_0 + n_2 I)d}{\lambda} = 1,2,3, \ldots$$

constructive interference



Fabry-Perot etalon

Multiphoton absorption

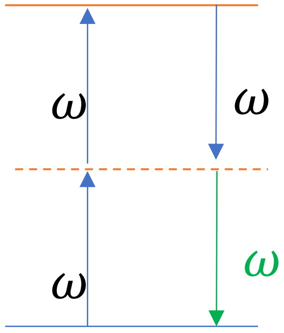
$$\epsilon(\omega) = \epsilon_0 \left(1 + \chi^{(1)} + \chi^{(3)}_{\text{Kerr}} |E(\omega)|^2\right)$$

Two-photon absorption coefficient:

$$\alpha = \alpha_0 + \alpha_2 I$$

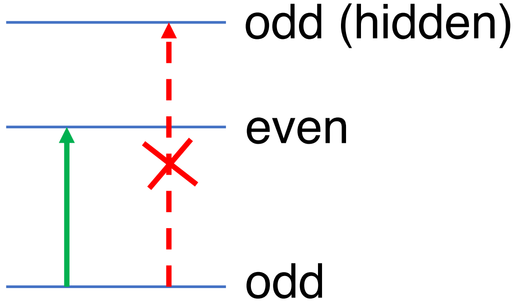
$$\frac{dI}{dz} = -\alpha\, I = -\alpha_0\, I - \alpha_2 I^2$$

$$\alpha_2 \sim \text{Im}\left[\chi^{(3)}_{\text{Kerr}}\right]$$



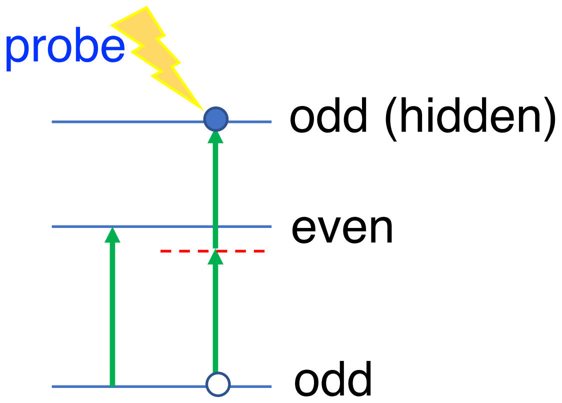
Since one-photon and two-photon transitions follow different selection rules, they are complementary spectroscopic tools.

Two-photon pump-probe spectroscopy:



$\langle \text{even}|r|\text{odd}\rangle \neq 0$

$\langle \text{odd}|r|\text{odd}\rangle = 0$



$\langle \text{odd}|r|\text{even}\rangle\langle \text{even}|r|\text{odd}\rangle \neq 0$

Wave-packet dynamics and Berry-phase effects

$$H_c(\boldsymbol{r}_c,t)\big|\phi_{\boldsymbol{q}}(\boldsymbol{r}_c,t)\big\rangle = E_c(\boldsymbol{r}_c,\boldsymbol{q},t)|\phi_{\boldsymbol{q}}(\boldsymbol{r}_c,t)\rangle$$

wave-pocket center-of-mass momentum

$$\boldsymbol{q}_c = \langle\Psi|\boldsymbol{q}|\Psi\rangle = \int d\boldsymbol{q}\,|a(\boldsymbol{q},t)|^2$$

wave-pocket center-of-mass position

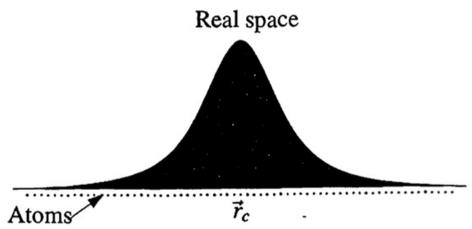
$$\boldsymbol{r}_c = \langle\Psi|\boldsymbol{r}|\Psi\rangle = \int d\boldsymbol{q}\,|a(\boldsymbol{q},t)|^2\left[\frac{\partial\gamma(\boldsymbol{q},t)}{\partial\boldsymbol{q}} + \left\langle u(\boldsymbol{r}_c,\boldsymbol{q},t)\middle|i\frac{\partial u(\boldsymbol{r}_c,\boldsymbol{q},t)}{\partial\boldsymbol{q}}\right\rangle\right]$$

$$\boldsymbol{r}_c = \frac{\partial\gamma(\boldsymbol{q}_c,t)}{\partial\boldsymbol{q}_c} + \underbrace{\left\langle u(\boldsymbol{r}_c,\boldsymbol{q}_c,t)\middle|i\frac{\partial u(\boldsymbol{r}_c,\boldsymbol{q}_c,t)}{\partial\boldsymbol{q}_c}\right\rangle}_{\mathcal{A}}$$

Berry curvature

$$\Omega = \partial_{\mathbf{q}}\times\mathcal{A}$$

$\mathcal{A}$ Berry connection



$$|\Psi\rangle = \int d\boldsymbol{q}\,a(\boldsymbol{q},t)|\phi_{\boldsymbol{q}}(\boldsymbol{r}_c,t)\rangle$$

$$|u(r_c,q,t)\rangle = e^{-i\,\boldsymbol{q}\cdot\boldsymbol{r}_c}|\phi_{\boldsymbol{q}}(\boldsymbol{r}_c,t)\rangle$$

$$a(\boldsymbol{q},t) = |a(\boldsymbol{q},t)|e^{-i\gamma(\boldsymbol{q},t)}$$

$$\int d\boldsymbol{q}\,|a(\boldsymbol{q},t)|^2 = 1$$

Lagrangian

$$L = \left\langle\Psi\middle|i\frac{d}{dt} - H\middle|\Psi\right\rangle$$

Semiclassical equations of motion

$$\frac{d\boldsymbol{r}_c}{dt} = \frac{\partial(E_c - \boldsymbol{M}\cdot\boldsymbol{B})}{\partial\boldsymbol{q}_c} - \frac{d\boldsymbol{q}_c}{dt}\times\boldsymbol{\Omega}$$

Anomalous velocity

$$\frac{d\boldsymbol{q}_c}{dt} = -e\boldsymbol{E} - e\frac{d\boldsymbol{r}_c}{dt}\times\boldsymbol{B}$$

Time reversal and spatial inversion

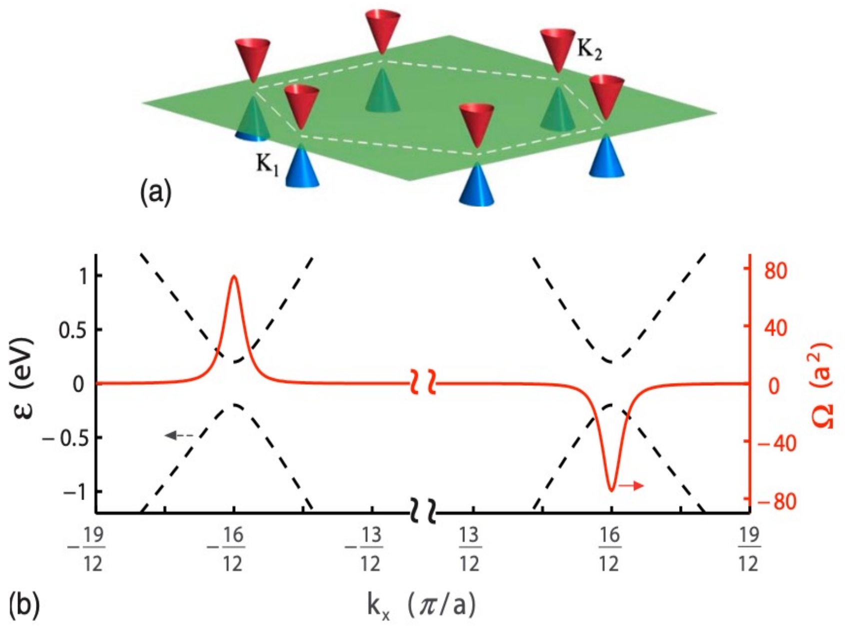
$$\boldsymbol{v}_{\text{anomalous}}(\boldsymbol{k},\boldsymbol{E}) = e\boldsymbol{E}\times\boldsymbol{\Omega}_n(\mathbf{k})$$

Under spatial inversion symmetry

$$\boldsymbol{\Omega}_n(-\boldsymbol{k}) = \boldsymbol{\Omega}_n(\boldsymbol{k}).$$

Under time reversal symmetry

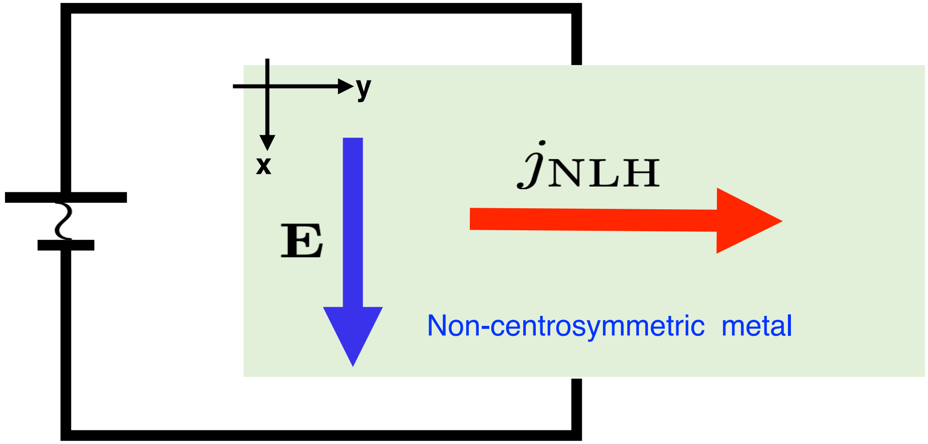
$$\boldsymbol{\Omega}_n(-\boldsymbol{k}) = -\boldsymbol{\Omega}_n(\boldsymbol{k}).$$



Valley Hall effect in gapped graphene

Therefore, for crystals with simultaneous time-reversal and spatial inversion symmetry the Berry curvature vanishes identically throughout the Brillouin zone.

Nonlinear Hall effect



- Time-reversal ✅
- Spatial inversion ❌

$$J_{\mathrm{Hall}}^{(1)} = -\frac{e^2}{\hbar}\sum_{n,\boldsymbol{k}} \boldsymbol{\mathcal{E}}(t)\times\boldsymbol{\Omega}_n(\boldsymbol{k})\rho_{nn}^{(0)} \to 0$$

$$J_{\mathrm{Hall}}^{(2)} = -\frac{e^2}{\hbar}\sum_{n,\boldsymbol{k}} \boldsymbol{\mathcal{E}}(t)\times\boldsymbol{\Omega}_n(\boldsymbol{k})\rho_{nn}^{(1)} \neq 0$$

$$\rho_{nn}^{(0)} = f_n(\boldsymbol{k}) = \frac{1}{1+e^{(\varepsilon_{\boldsymbol{k}}-\mu)/k_B T}}$$

$$\rho_{nn}^{(1)} = \frac{e\tau\,\boldsymbol{\mathcal{E}}(\omega)\cdot\partial_{\boldsymbol{k}} f_n(\boldsymbol{k})}{1+i\omega\tau}e^{i\omega t} + cc.$$

Nonlinear Hall

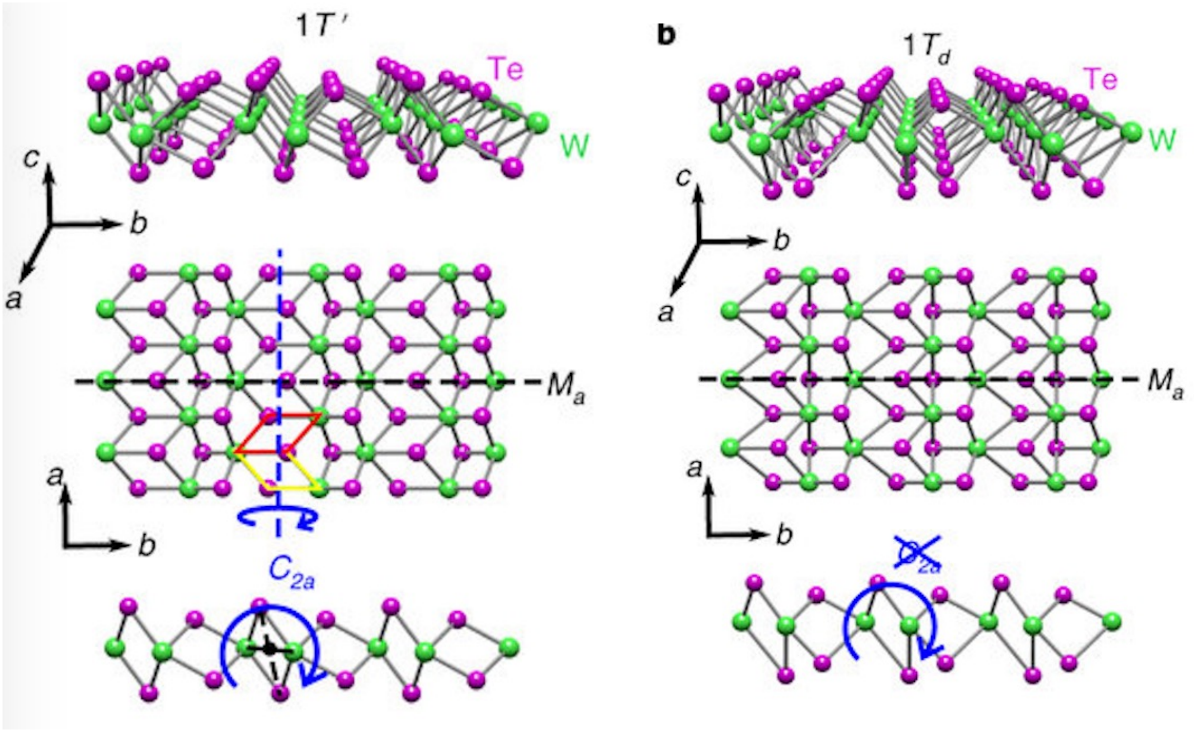
$$J_{\mathrm{Hall}}^{(2)} \propto \mathrm{Re}\left[\frac{\tau}{1+i\omega\tau}\right]|\mathcal{E}(\omega)|^2\boldsymbol{D}$$

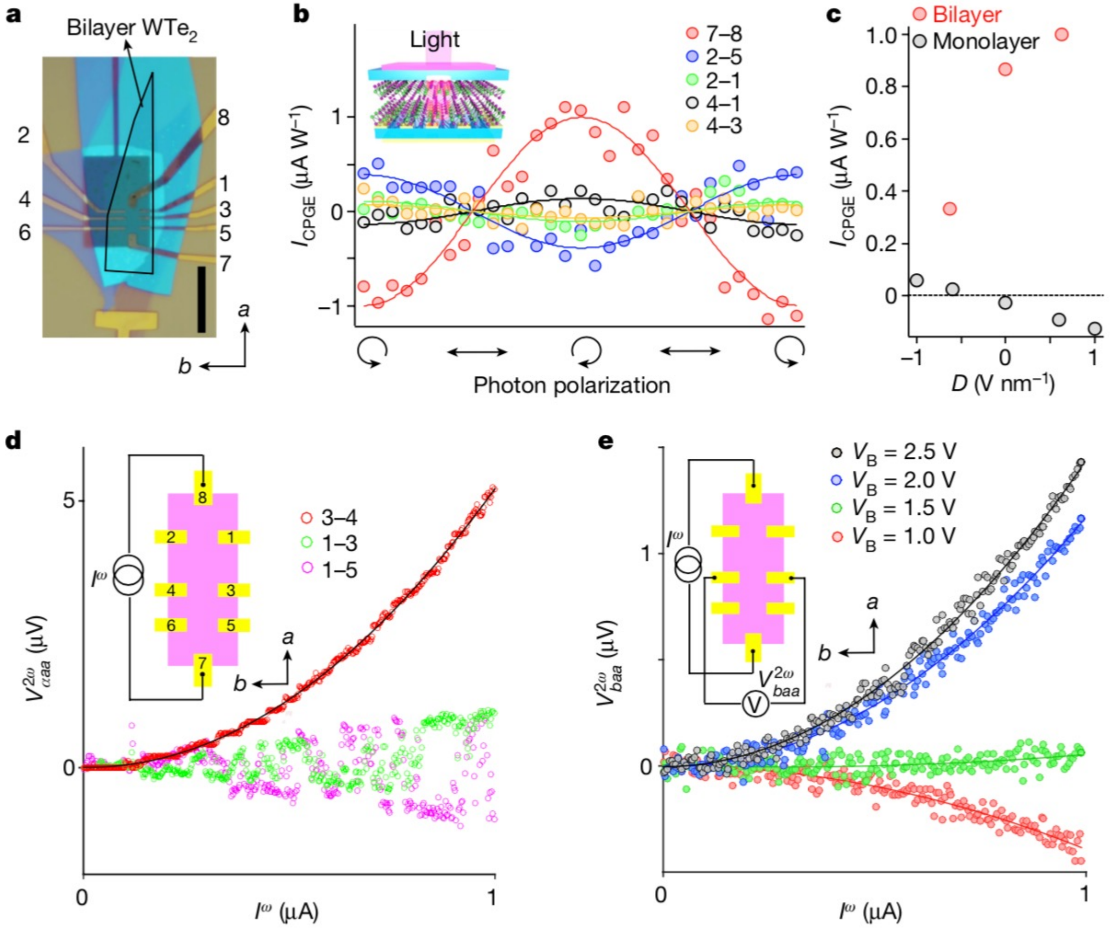
Berry curvature dipole (2D):

$$D_\alpha = \sum_{\boldsymbol{k}} \Omega_{n,z}(\boldsymbol{k})\frac{\partial f_n(\boldsymbol{k})}{\partial k_\alpha} = -\sum_{\boldsymbol{k}} f_n(\boldsymbol{k})\frac{\partial \Omega_n(\boldsymbol{k})}{\partial k_\alpha}$$

I. Sodemann and L. Fu, PRL 115, 216806 (2015)

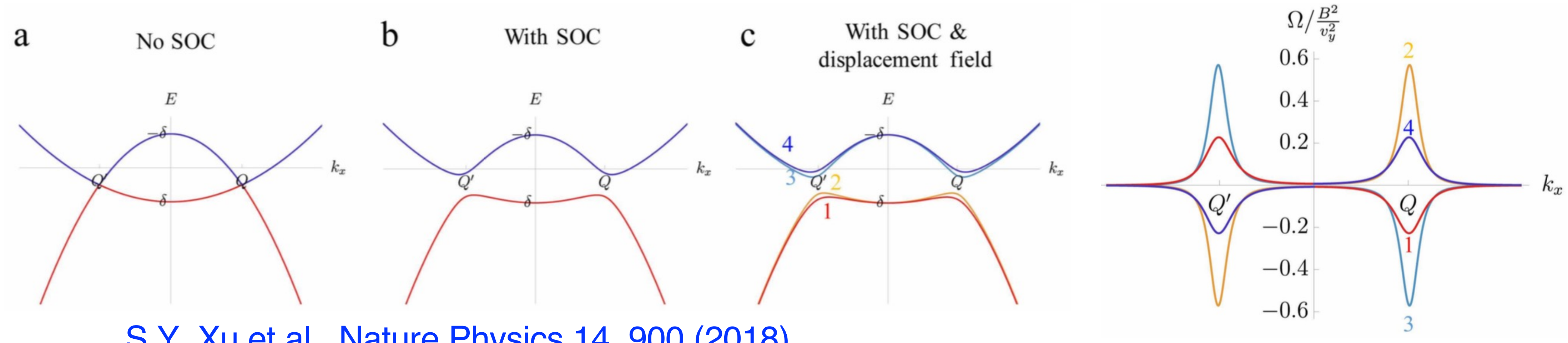
Nonlinear Hall effect in WTe2



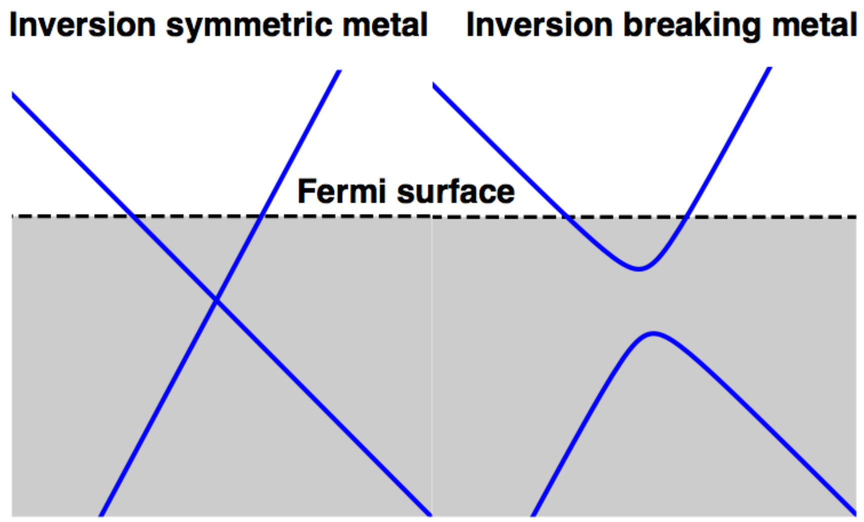


Q. Ma et al., Nature 565, 342 (2019)

Tilted-Dirac fermions model for WTe2



S.Y. Xu et al., Nature Physics 14, 900 (2018).



$$\hat{\mathcal{H}}_{\mathbf{k}} = \alpha\zeta k_x \hat{I} + v(\zeta k_x \hat{\sigma}_x + k_y \hat{\sigma}_y) + m_f \hat{\sigma}_z,$$

Tilting term   Inversion breaking term

Nonlinear Hall conductivity

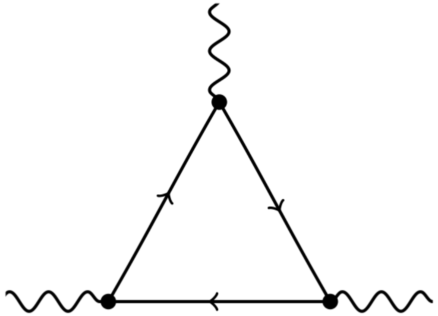
$$\boxed{\sigma^{(2)}_{yxx}(2\omega) = \frac{ie^3}{\omega}\sum_{\mathbf{k},\zeta} n_{\rm F}(\varepsilon^c_{\mathbf{k}})\frac{\partial\Omega_{yx}(k)}{\partial k_x}\; C\left(\frac{\omega}{2\varepsilon_k}\right)}$$

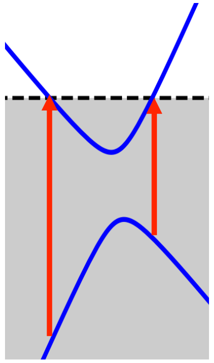
$$C(x) = \frac{1}{(1-4x^2)(1-x^2)}$$

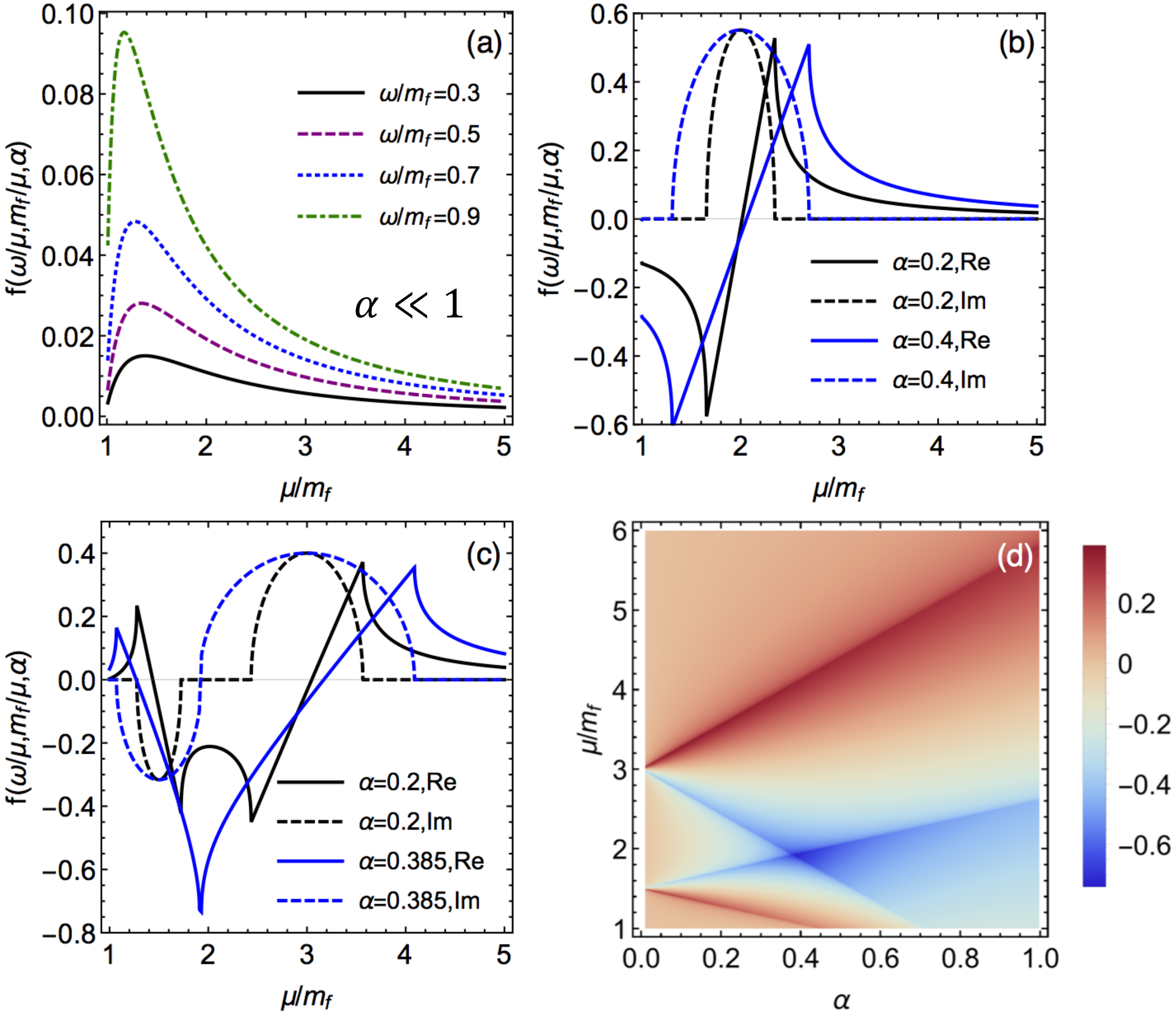
$$\sigma^{(2)}_{yxx}(2\omega) = \frac{ie^3}{\omega^2} f\left(\frac{\omega}{\mu}, \frac{m_f}{\mu}, \alpha\right)$$

for $\alpha \ll 1$:

$$f(x,y,\alpha) \approx \frac{12\alpha}{\pi^2}\frac{xy(1-y^2)\Theta(1-y)}{(x^2-1)(x^2-4)}$$

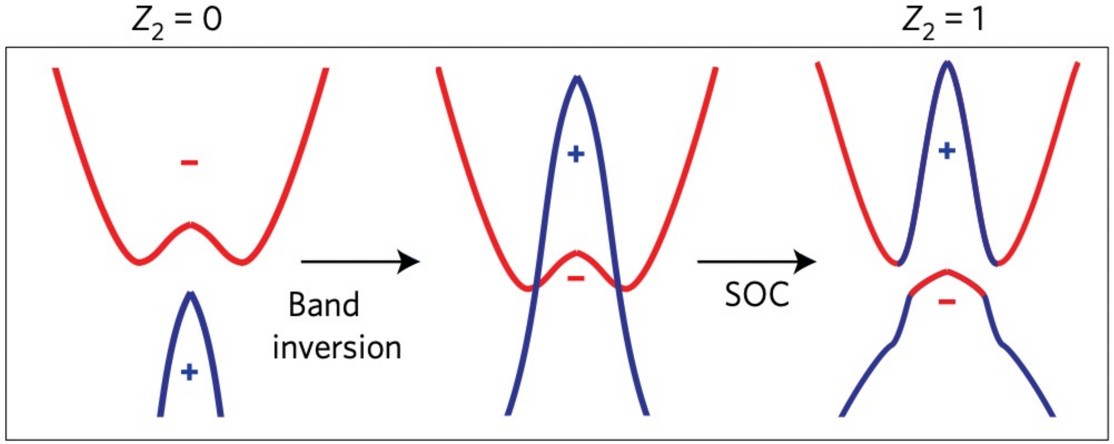


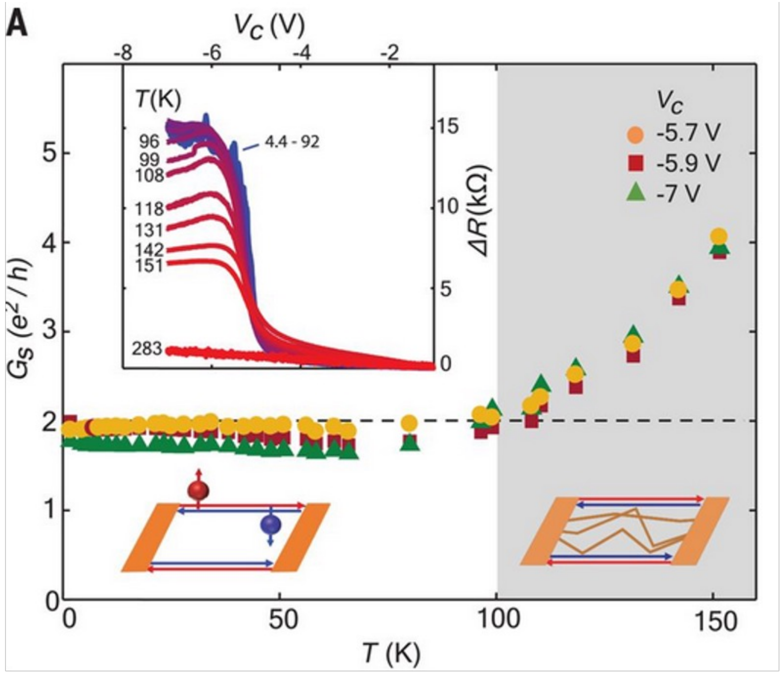




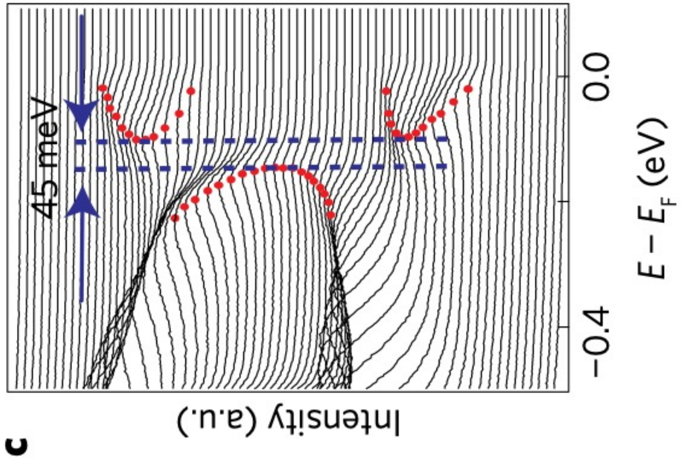
HR, V Juričić, Phys. Rev. Research 2, 013069 (2020)

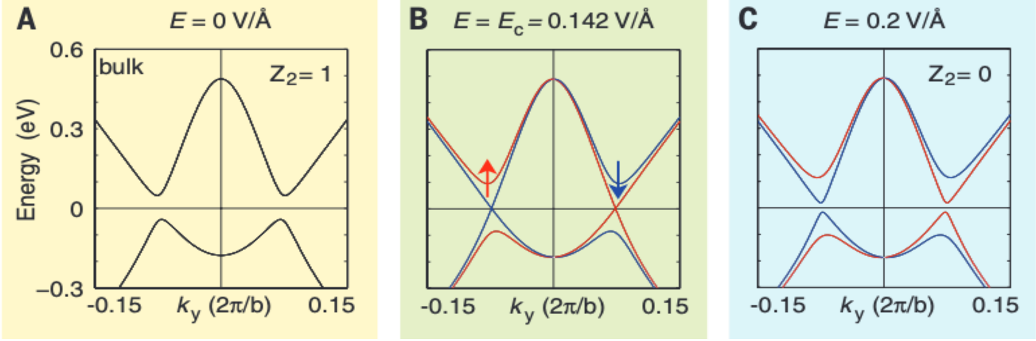
High temperature (~100 K) quantum spin Hall in WTe2

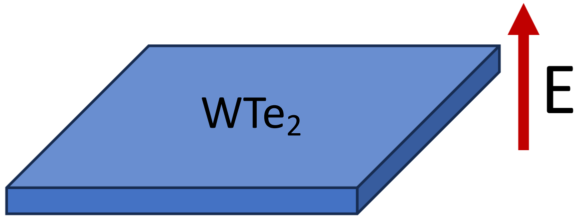




Shujie Tang et al. Nat. Phys. 13 683 (2017)
S. Wu, et al. Science 359, 76 (2018)







Topological | Trivial
$U = 0$ | $U = \delta_{soc}$ | $U > \delta_{soc}$

Second-order Spin Hall current

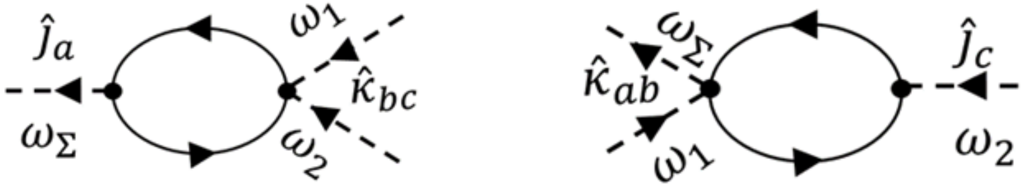
$$J_y^{\text{charge}} = \sigma_{yxx}^{\text{charge}}(0,\omega,-\omega)E_x(\omega)E_x(-\omega) \qquad J_x^{\text{charge}} = \sigma_{xyy}^{\text{charge}}(0,\omega,-\omega)E_y(\omega)E_y(-\omega)$$

Mirror symmetry x→ -x implies

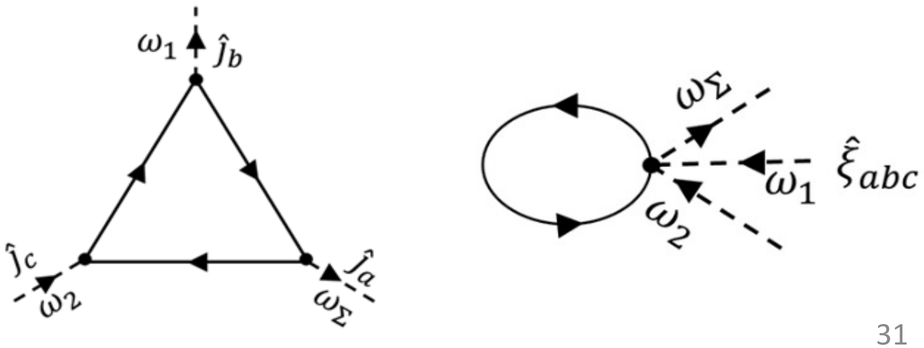
$$\sigma_{yxx}^{\text{charge}} = \sigma_{yxx}^{\uparrow} + \sigma_{yxx}^{\downarrow} = 2\sigma_{yxx}^{\uparrow} \neq 0$$
$$\sigma_{xyy}^{\text{charge}} = \sigma_{xyy}^{\uparrow} + \sigma_{xyy}^{\downarrow} = 0$$

$$\sigma_{yxx}^{\text{spin}} = \sigma_{yxx}^{\uparrow} - \sigma_{yxx}^{\downarrow} = 0$$
$$\sigma_{xyy}^{\text{spin}} = \sigma_{xyy}^{\uparrow} - \sigma_{xyy}^{\downarrow} = 2\sigma_{yxx}^{\uparrow} \neq 0$$

Shift current

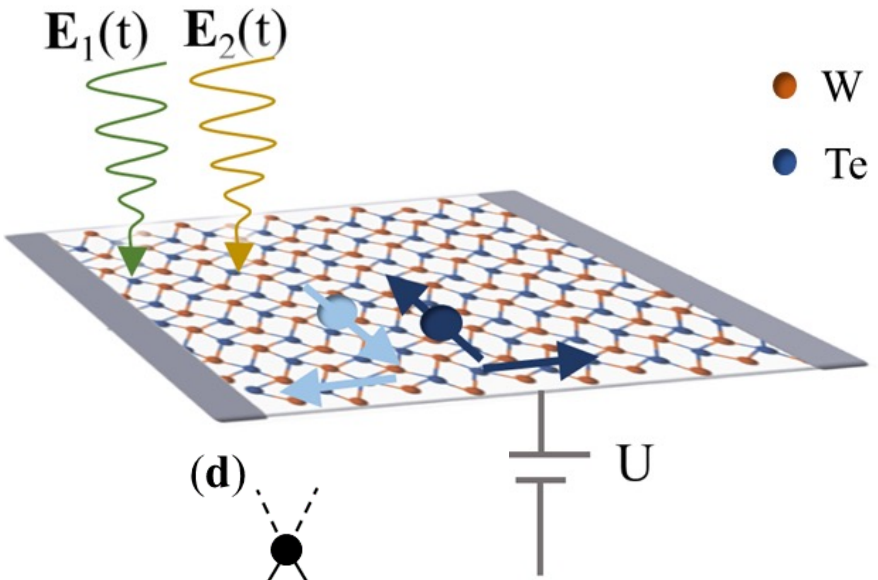


Injection current



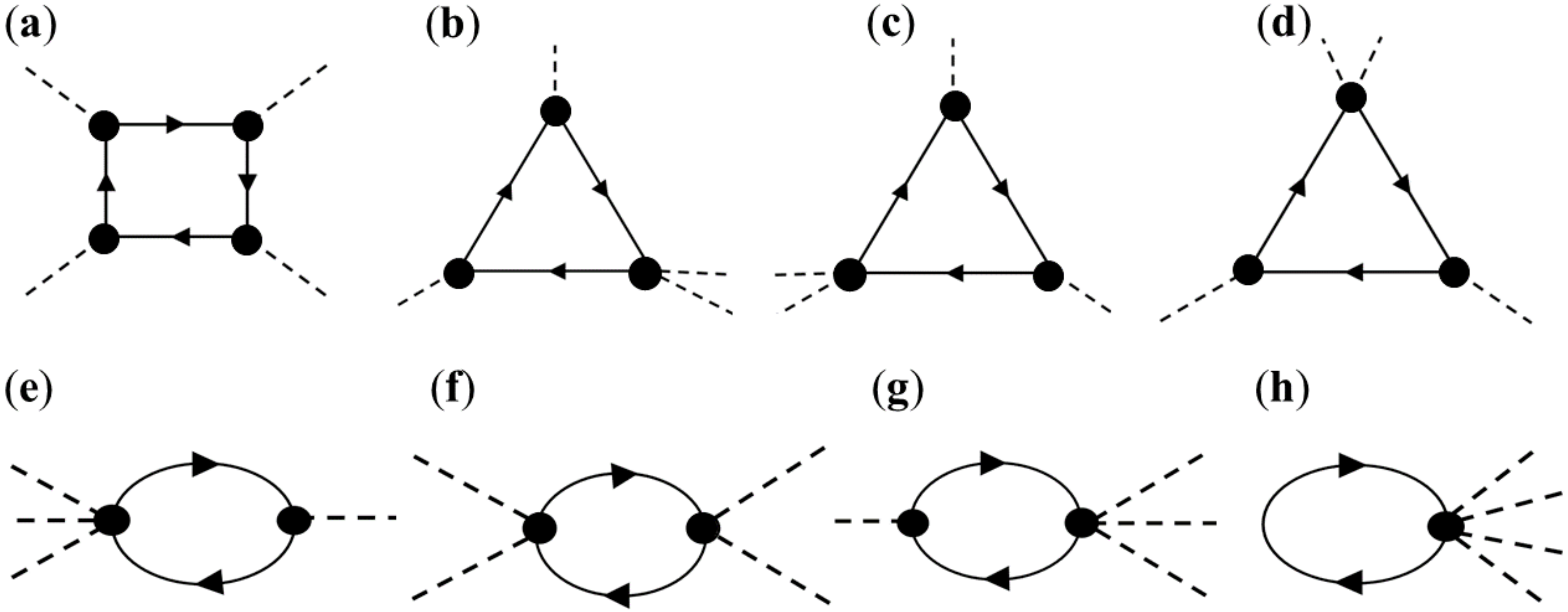
P. Bhalla, HR, arXiv:2303.06238 (2023)

Third order Spin Hall current



$e^{i\omega t}e^{i\omega t}\ e^{-i2\omega t}$ → **two-color rectification current**

$$j_a^{\text{spin}} = \sigma_{abbb}^{(3)\text{spin}}(0,\omega,\omega,-2\omega)E_b^2(\omega)E_b(-2\omega)$$



P. Bhalla, HR, arXiv:2303.06238 (2023)

Nonlinear Spin Hall current

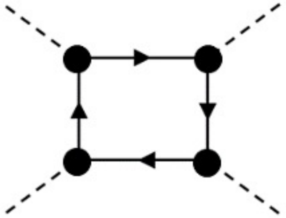
In-gap second-order photocurrent in time-reversal broken systems

$$\sigma^{(2)}_{\text{sub}} = -2C\left(\frac{2\Gamma}{\gamma} - 1\right)\left(\int_{\mathbf{k}} \sum_{nm} f_{nm} |r^a_{nm}|^2 v^a_{nn}\right)$$

D. Kaplan et al. PRL **125**, 227401 (2020)

$$\boldsymbol{v} \to \boldsymbol{E}\times\boldsymbol{\Omega}(\mathbf{k}) \quad \Longrightarrow \quad \sigma^{(2)}_{\text{sub}} \to \sigma^{(3)}_{\text{sub}}$$

$$\sigma^{(3)}_{yxxx} \sim \int_{\mathbf{k}} \Omega^{\text{spin}}_{yx}(\mathbf{k}) \times (\text{even terms in } \boldsymbol{k})$$

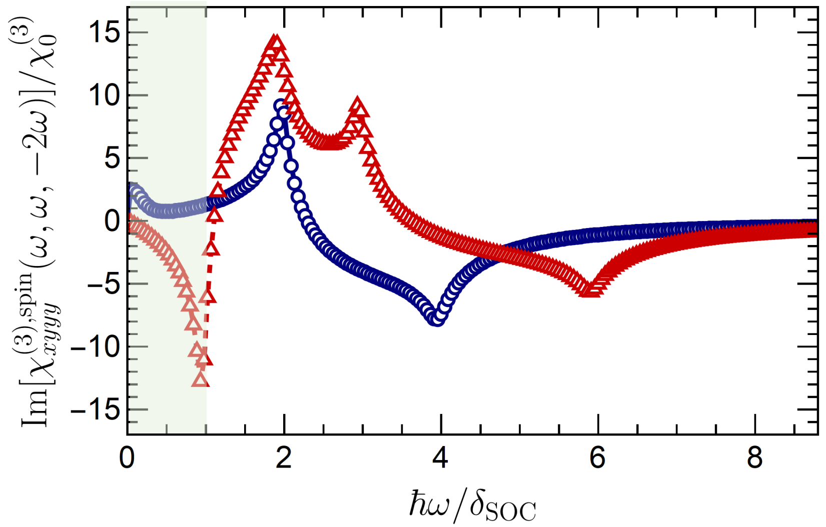


Spin Berry curvature

$$\Omega^{\text{spin}}_{yx} = \Omega_{\uparrow} - \Omega_{\downarrow}$$

Even under time-reversal

$$\Omega^{\text{spin}}_{yx}(-\mathbf{k}) = +\Omega^{\text{spin}}_{yx}(\mathbf{k})$$

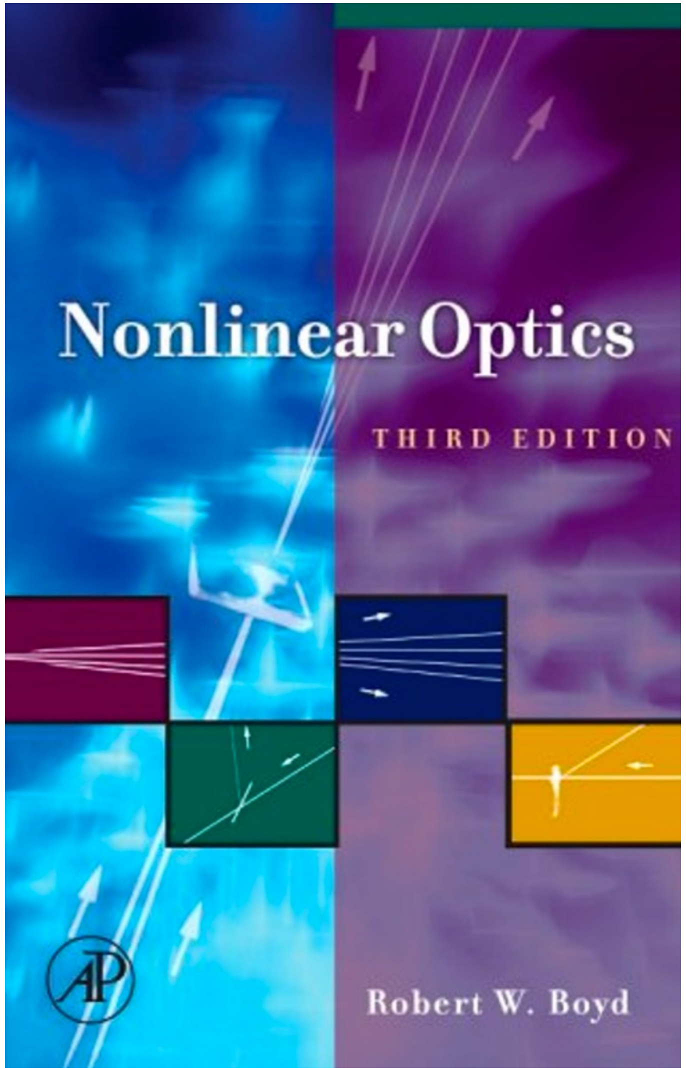


In-gap two-color spin Hall current

P. Bhalla, HR, arXiv:2303.06238 (2023)

Thank you for your attention!

- Reviewed Quantum materials
- Reviewed Nonlinear optics
- Topological nonlinear photocurrents in WTe2



Extra references:

nature photonics
REVIEW ARTICLE
https://doi.org/10.1038/s41566-022-00988-y
Check for updates

High harmonic generation in condensed matter

nature nanotechnology
ARTICLES
https://doi.org/10.1038/s41565-018-0145-8

Broadband, electrically tunable third-harmonic generation in graphene

Giancarlo Soavi[1]*, Gang Wang[1], Habib Rostami[2], David G. Purdie[1], Domenico De Fazio[1], Teng Ma[1], Birong Luo[1], Junjia Wang[1], Anna K. Ott[1], Duhee Yoon[1], Sean A. Bourelle[1], Jakob E. Muench[1], Ilya Goykhman[1], Stefano Dal Conte[3,4], Michele Celebrano[4], Andrea Tomadin[2], Marco Polini[2], Giulio Cerullo[3,4] and Andrea C. Ferrari[1]*

nature physics
REVIEW ARTICLES
PUBLISHED ONLINE: 30 OCTOBER 2017 | DOI: 10.1038/NPHYS4302

The physics of quantum materials

B. Keimer[1]* and J. E. Moore[2,3]*

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]*

REVIEW ARTICLES
PUBLISHED ONLINE: 25 SEPTEMBER 2017 | DOI: 10.1038/NPHYS4274
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Emergent functions of quantum materials

Yoshinori Tokura[1,2]*, Masashi Kawasaki[1,2] and Naoto Nagaosa[1,2]

nature materials
REVIEW ARTICLE
PUBLISHED ONLINE: 25 OCTOBER 2017 | DOI: 10.1038/NMAT5017

Towards properties on demand in quantum materials

D. N. Basov[1]*, R. D. Averitt[2]* and D. Hsieh[3]*