Controlling the valley degree of freedom in Graphene systems

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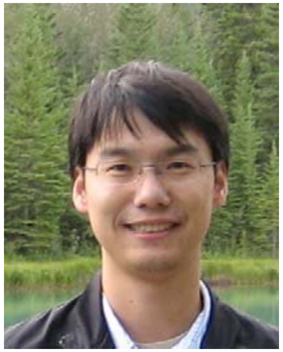


Dr. Di Xiao

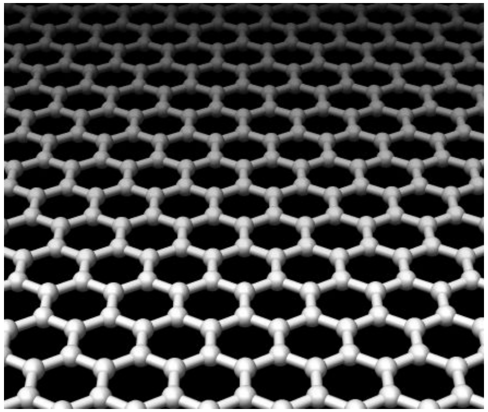
Dr. Tianyi Cai

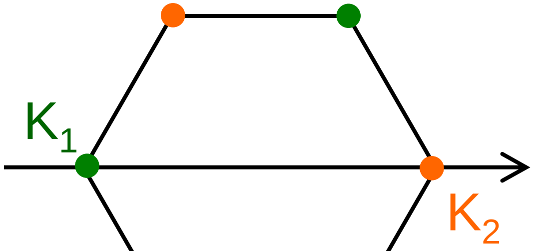
Dr. Wang Yao

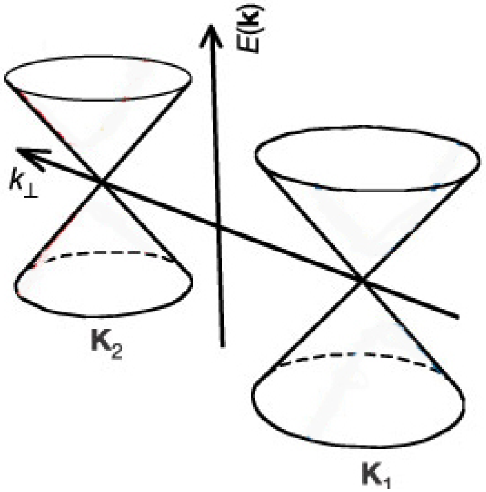
Outline

- The valley index in graphene and inversion symmetry breaking
- Valley contrasting magnetic moment and Berry curvature
- Magnetic control: valley polarization and magnetism
- Electrical control: valley Hall effect and inverse
- Optical control: valley dependent circular dichroism

Valley degree of freedom in Graphene







- **Valley index in graphene**

  Two inequivalent valleys related by time reversal symmetry

- **Long intervalley scattering time**
  - ~ 100 ps observed in bilayers. Gorbachev et al., PRL 07"
  - Valleytronics: analogy to spintronics. Beenakker et al. 07"

- **How to control the valley degree of freedom?**

Learning from spintronics

- Spin has a magnetic moment: magnetic control
- Spin-orbit coupling: electrical control (spin Hall effect)
- Spin-dependent cicular dichroism: optical control

- How to control the valley degree of freedom?

Three basic electronic properties

1. Band energy: particle or hole, effective mass,..
2. Magnetic moment: spin and orbital moment
3. Berry curvature: Berry phase effects

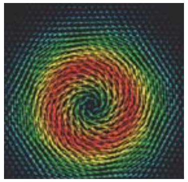
$$\varepsilon_n(\boldsymbol{k}) = \varepsilon_n^0(\boldsymbol{k}) - \boldsymbol{m}(\boldsymbol{k}) \cdot \boldsymbol{B}$$

$$\boxed{\begin{aligned} \dot{\boldsymbol{r}} &= \frac{1}{\hbar}\frac{\partial \varepsilon_n(\boldsymbol{k})}{\partial \boldsymbol{k}} - \dot{\boldsymbol{k}} \times \boldsymbol{\Omega}_n(\boldsymbol{k}) \\ \hbar\dot{\boldsymbol{k}} &= -e\boldsymbol{E} - e\dot{\boldsymbol{r}} \times \boldsymbol{B} \end{aligned}}$$

Berry curvature

$$\boldsymbol{\Omega}_n(\boldsymbol{k}) = i\left\langle \frac{\partial u_{n\boldsymbol{k}}}{\partial \boldsymbol{k}} \right| \times \left| \frac{\partial u_{n\boldsymbol{k}}}{\partial \boldsymbol{k}} \right\rangle$$

Magnetic moment



$$\begin{aligned} \boldsymbol{m}(\boldsymbol{k}) &= -\frac{e}{2}\langle W | (\hat{\boldsymbol{r}} - \boldsymbol{r}_c) \times \boldsymbol{v} | W \rangle \\ &= -i\frac{e}{\hbar}\left\langle \frac{\partial u}{\partial \boldsymbol{k}} \right| \times (\hat{H} - \varepsilon_{\boldsymbol{k}}^0) \left| \frac{\partial u}{\partial \boldsymbol{k}} \right\rangle \end{aligned}$$

Our goal: engineer valley-dependent magnetic moment and Berry curvature.

Symmetry Consideration

Time-reversal symmetry $\Omega(\boldsymbol{k}) = -\Omega(-\boldsymbol{k})$ $m(\boldsymbol{k}) = -m(-\boldsymbol{k})$

Space-inversion symmetry $\Omega(\boldsymbol{k}) = \Omega(-\boldsymbol{k})$ $m(\boldsymbol{k}) = m(-\boldsymbol{k})$

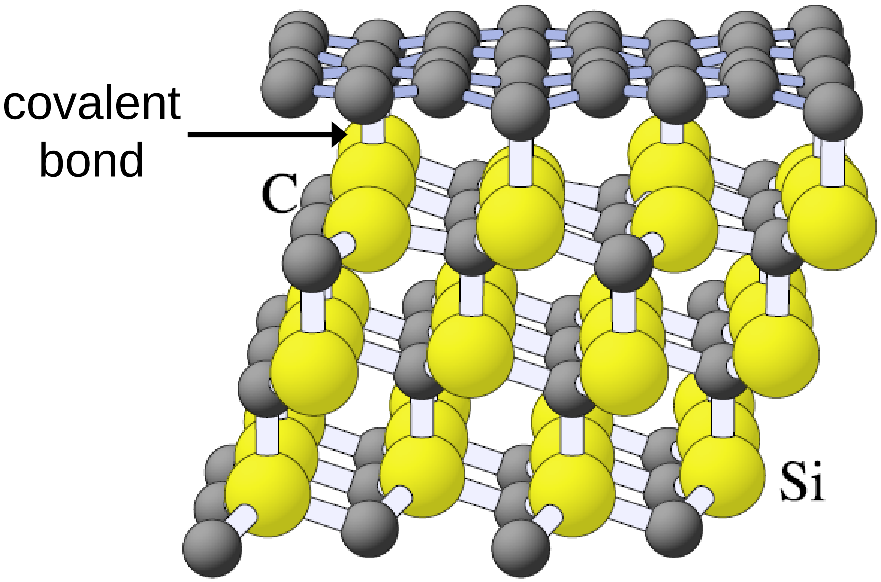
Both symmetries $\Omega(\boldsymbol{k}) = 0$ $m(\boldsymbol{k}) = 0$

Need to break time-reversal (ferromagnet) or spatial inversion symmetries.

Inversion Asymmetry in Eptixially Graphene

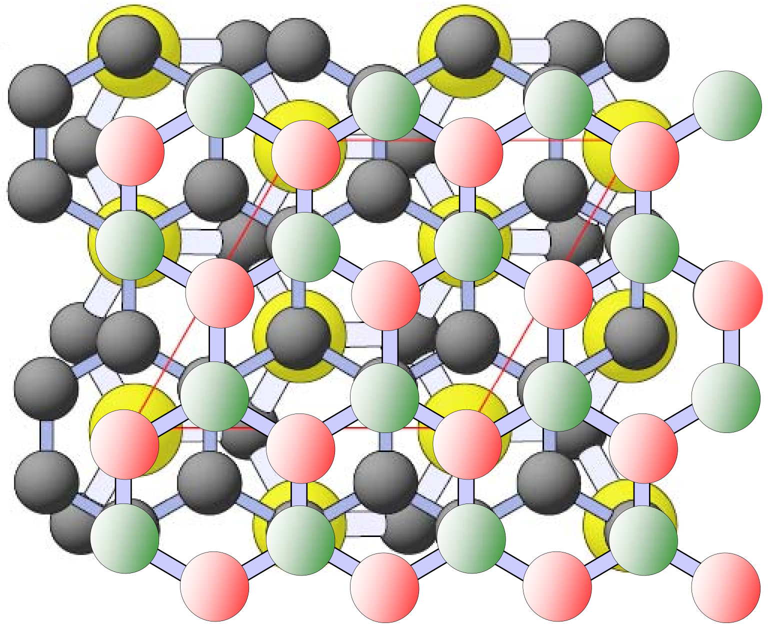
Side view

1st carbon layer: **‘dead’** layer



Top view

2nd carbon layer: **active** layer



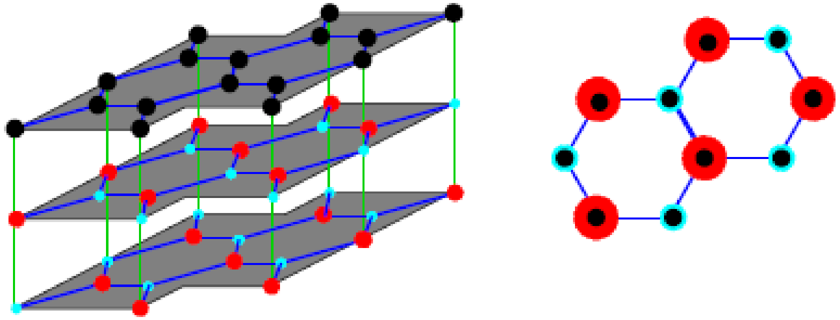
Site energy difference between A and B ⇨ broken inversion symmetry

Mattausch & Pankratov arXiv:0704.0216

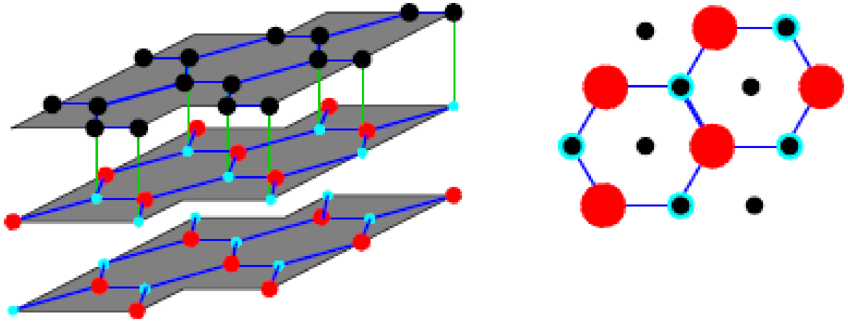
Graphene on Boron Nitride

Giovannetti *et al.* arXiv:0704.1994

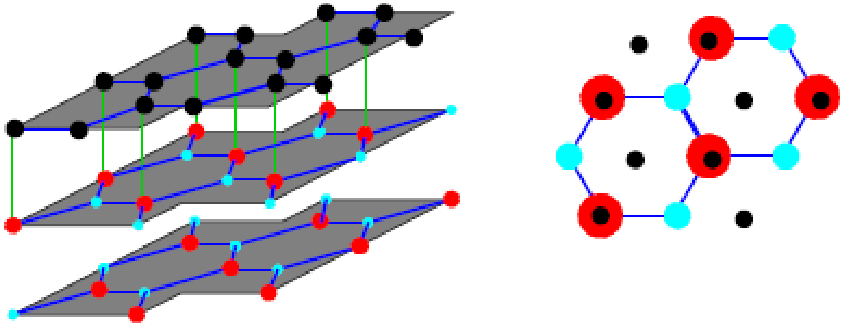
A on Boron / B on Nitride



one sublattice on Boron



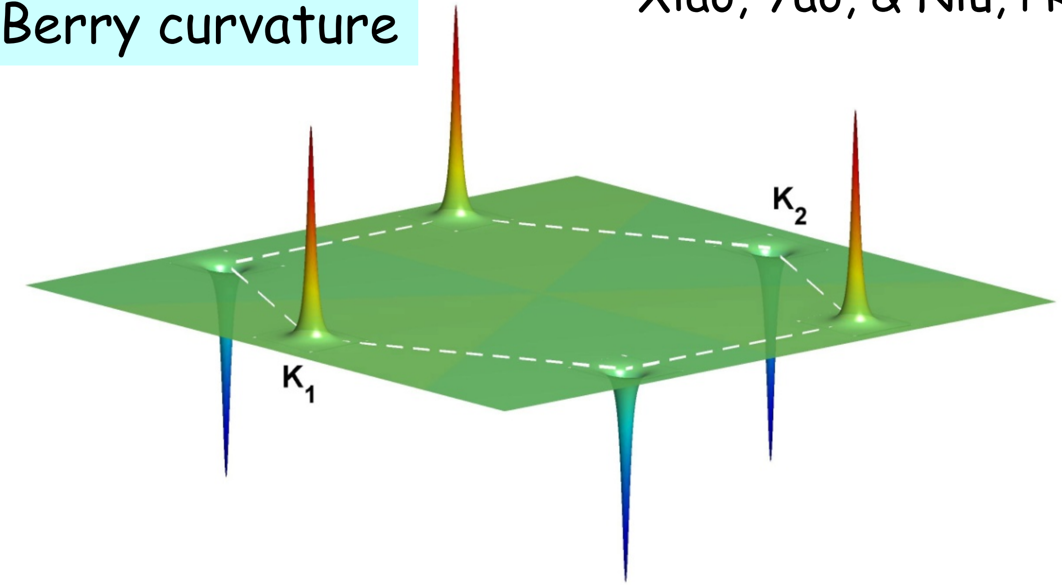
one sublattice on Nitride



Valley Contrasting Berry Curvature

Berry curvature

Xiao, Yao, & Niu, PRL 07"

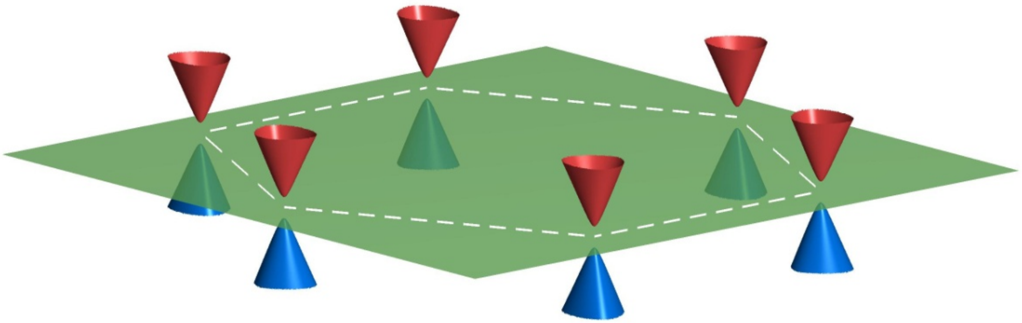


$$\Omega(\boldsymbol{q}) = \tau_z \frac{3\Delta t^2}{2(\Delta^2 + 3q^2a^2t^2)^{3/2}}$$

Valley index

$\tau_z = 1\ (-1)$ for valley K_1 (K_2)

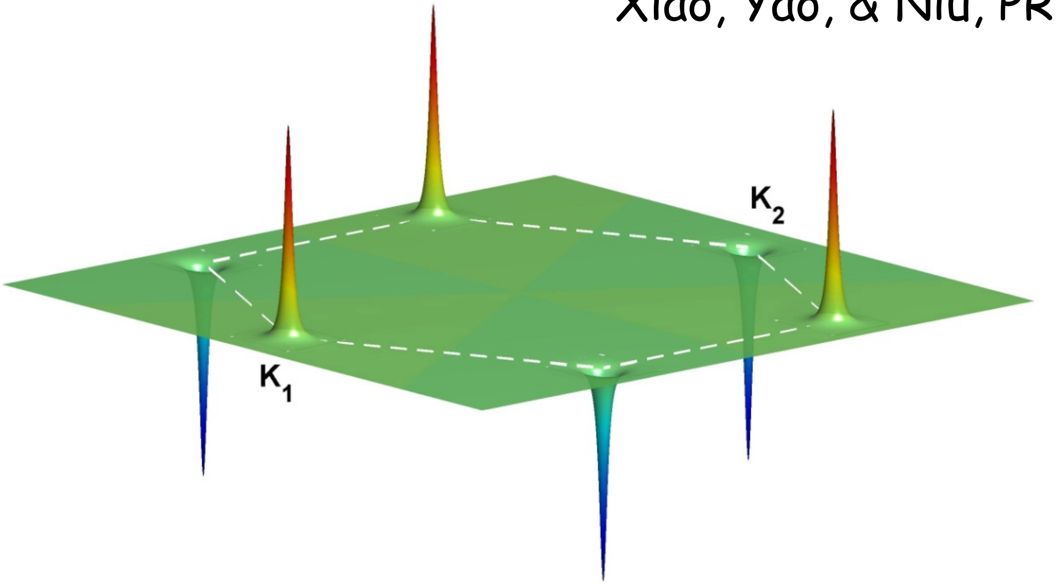
Energy dispersion: massive Dirac bands



$$\varepsilon(q) = \pm\sqrt{\Delta^2 + 3t^2q^2a^2/4}$$

Valley Magnetic Moment

Xiao, Yao, & Niu, PRL 07"



$$\mathrm{m}(\boldsymbol{k}) = \tau_z \frac{3ea^2\Delta t^2}{4\hbar(\Delta^2 + 3q^2a^2t^2)}$$

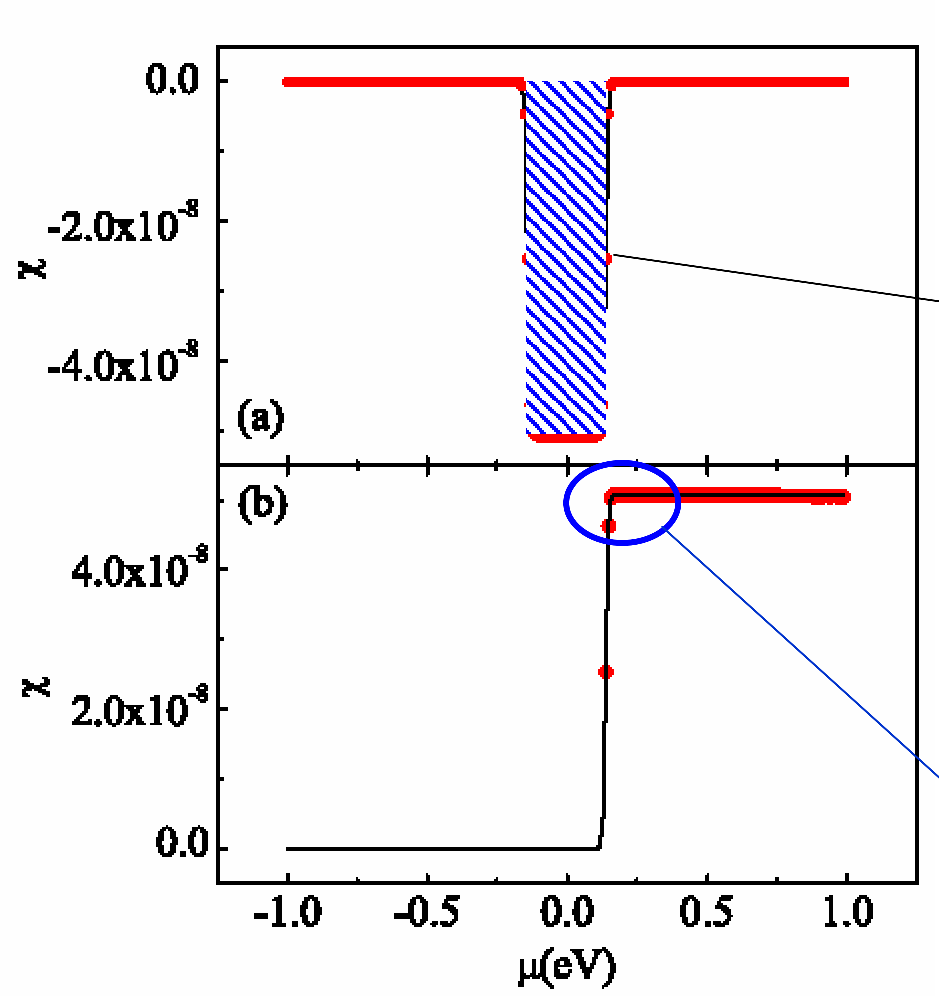
Valley index

$\tau_z = 1\ (-1)$ for valley K$_1$ (K$_2$)

- At band bottom: $\mathrm{m}(\boldsymbol{K}_{1,2}) = \tau_z \mu_B^*$, $\mu_B^* = \frac{e\hbar}{2m_e^*}$,
- Valley index is associated with an intrinsic magnetic moment

$$\mu_B^* \sim 30\mu_B$$

Magnetic Susceptibility



- All bands

$$\chi = \frac{e^2a^2t}{4\pi\hbar^2\Delta}\frac{e^{\beta\mu}\left(e^{\beta\Delta/2}-e^{-\beta\Delta/2}\right)}{1+2e^{\beta\mu}Cosh\left(\beta\Delta/2\right)+e^{2\beta\mu}}$$

- Conduction bands only

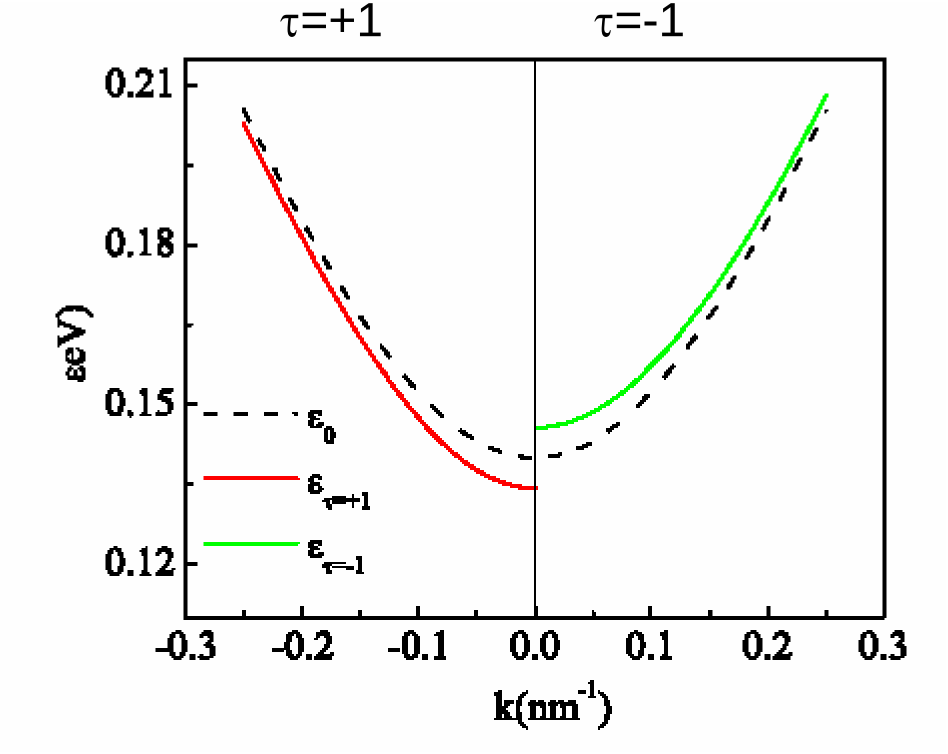
$$\chi_V = \frac{e^2a^2t}{4\pi\hbar^2\Delta}\frac{1}{1+e^{-\beta(\mu-\Delta/2)}}$$

$$\begin{aligned}\chi_V &= \chi_p + \chi_d \\ &= \frac{e^2}{6\pi m^*}\end{aligned} \qquad \begin{cases}\chi_p = \dfrac{3e^2a^2t^2}{8\pi\hbar^2\Delta} \\[2ex] \chi_d = -\dfrac{e^2a^2t^2}{8\pi\hbar^2\Delta}\end{cases}$$

Magnetic control of valley polarization

K1 valley
τ=+1

K2 valley
τ=-1



$$E = E_0 - m \cdot B$$

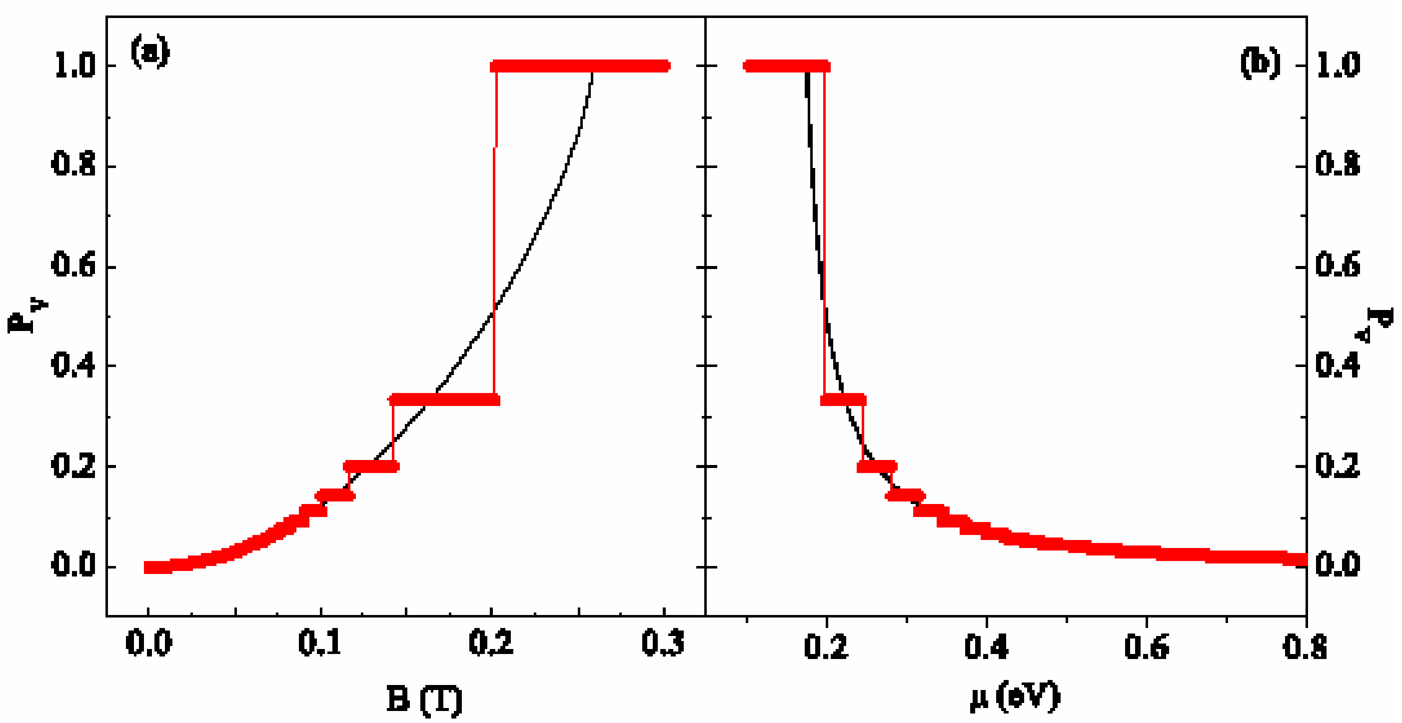
- Valley polarization

$$P_V = \frac{N_{+1} - N_{-1}}{N_{+1} + N_{-1}}$$

- Berry phase correction

$$N_\tau = \int_0^{k_F} \frac{1}{(2\pi)^2} \left(1 + \frac{eB \cdot \Omega_\tau}{\hbar} \right) dk$$

Magnetic control of valley polarization



The variations of valley polarization with magnetic field and chemical potential.

Magnetization from valley polarization

- Valley contrasting orbital magnetization

$$M = 2\int \frac{d\boldsymbol{k}}{(2\pi)^2}[m(\boldsymbol{k}) + (e/\hbar)(\mu - \varepsilon(\boldsymbol{k}))\Omega(\boldsymbol{k})]$$

$$= 2(e/\hbar)\mu \boxed{\int \frac{d\boldsymbol{k}}{(2\pi)^2}\Omega(\boldsymbol{k})} . \longrightarrow \frac{C(\mu)}{2\pi}$$

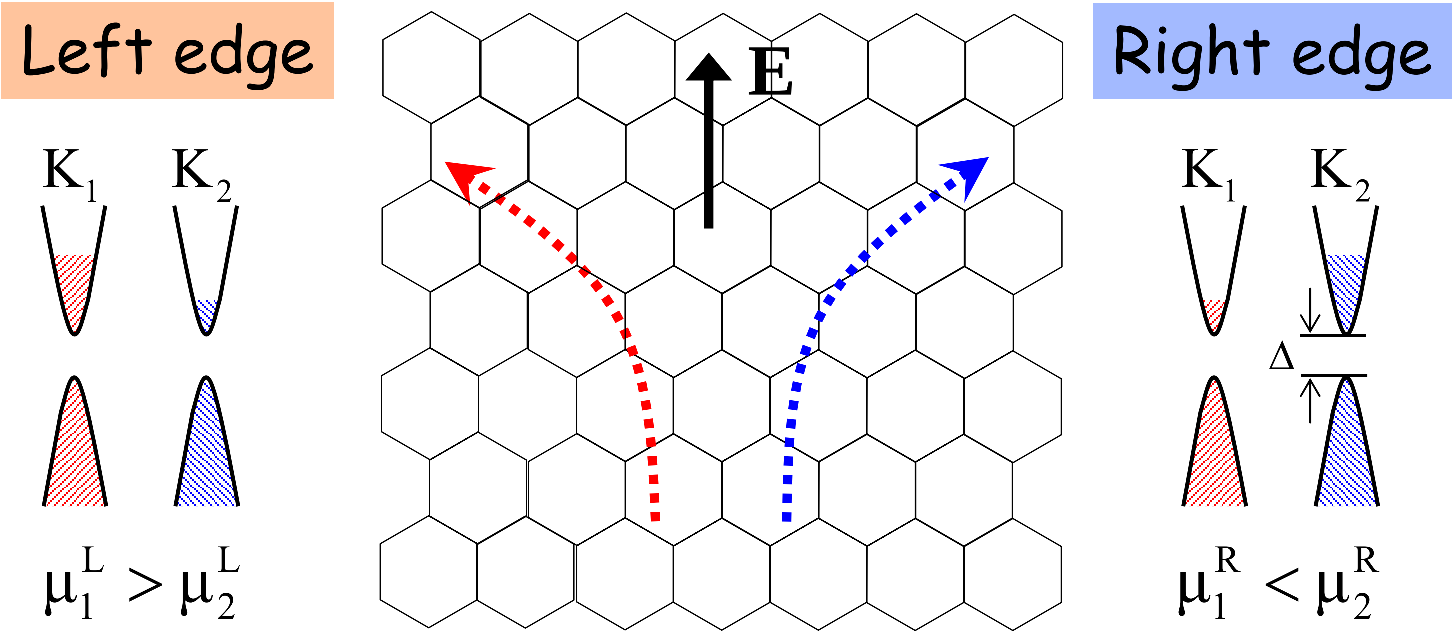
$C(\mu) \rightarrow \frac{\tau_z}{2}$ for $\mu >> \Delta$

Berry phase of π

- Net orbital magnetization by valley polarization

$$\delta M = 2\frac{e}{\hbar}[\mu_1 \frac{C_1(\mu_1)}{2\pi} + \mu_2 \frac{C_2(\mu_2)}{2\pi}] \approx 2\frac{e}{h}C_1(\mu)\delta\mu$$

Valley Hall Effect - Electric Control



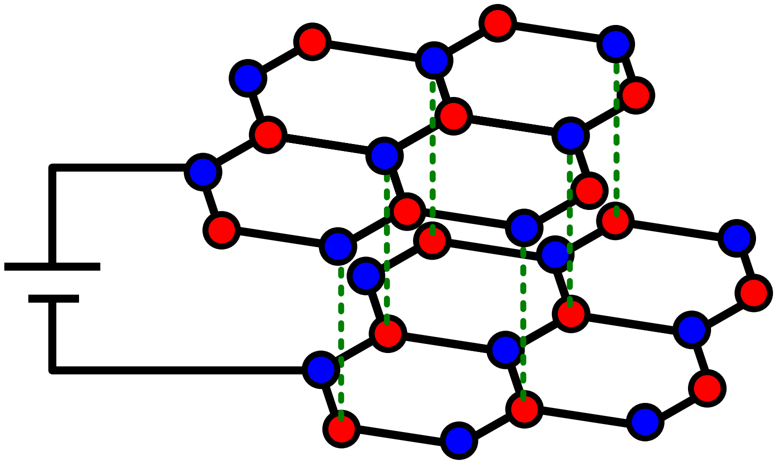
Valley polarization on edge: $\int \delta n_v \, dx = j_x^v \tau_v = 2\frac{\sigma_H \tau_v}{e} E_y$

Edge magnetization: $M_{\text{edge}} = \int \delta M \, dx = \frac{4}{h}\frac{\partial \mu}{\partial n} \mathcal{C}_1(\mu) \sigma_H \tau_v E$

$\sim 100\, \mu_B / \mu\text{m}^2$ for $E \sim \text{mV}/\mu\text{m}$

Xiao, Yao, & Niu, PRL 07"

Biased Graphene Bilayer



$$H(\boldsymbol{k}) = \begin{bmatrix} \frac{\Delta}{2} & V(\boldsymbol{k}) & 0 & 0 \\ V^*(\boldsymbol{k}) & \frac{\Delta}{2} & t_\perp & 0 \\ 0 & t_\perp & -\frac{\Delta}{2} & V(\boldsymbol{k}) \\ 0 & 0 & V^*(\boldsymbol{k}) & -\frac{\Delta}{2} \end{bmatrix}$$

$$V(\boldsymbol{k}) = -t\left(e^{i\boldsymbol{k}\cdot\boldsymbol{d}_1} + e^{i\boldsymbol{k}\cdot\boldsymbol{d}_2} + e^{i\boldsymbol{k}\cdot\boldsymbol{d}_3}\right)$$

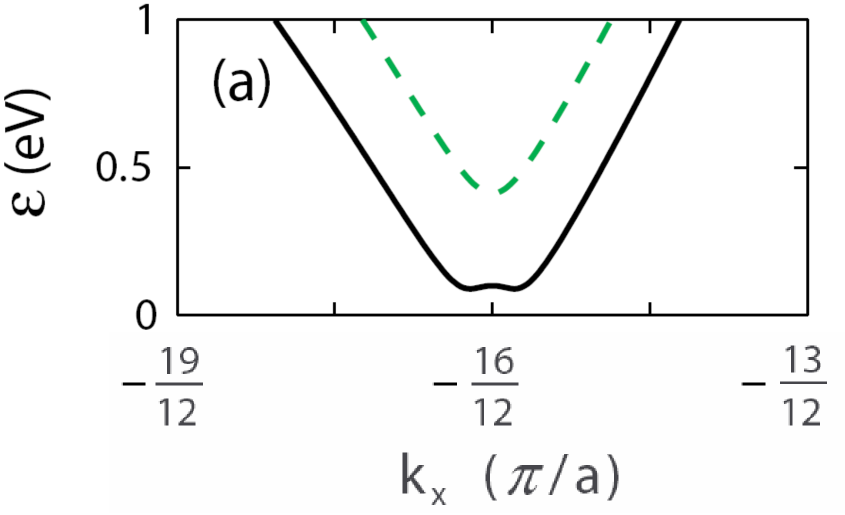
Broken inversion symmetryt bias

● sublattice A1 $\xleftrightarrow{t}$ ● sublattice B1 $-\frac{\Delta}{2}$

$\updownarrow t_\perp$

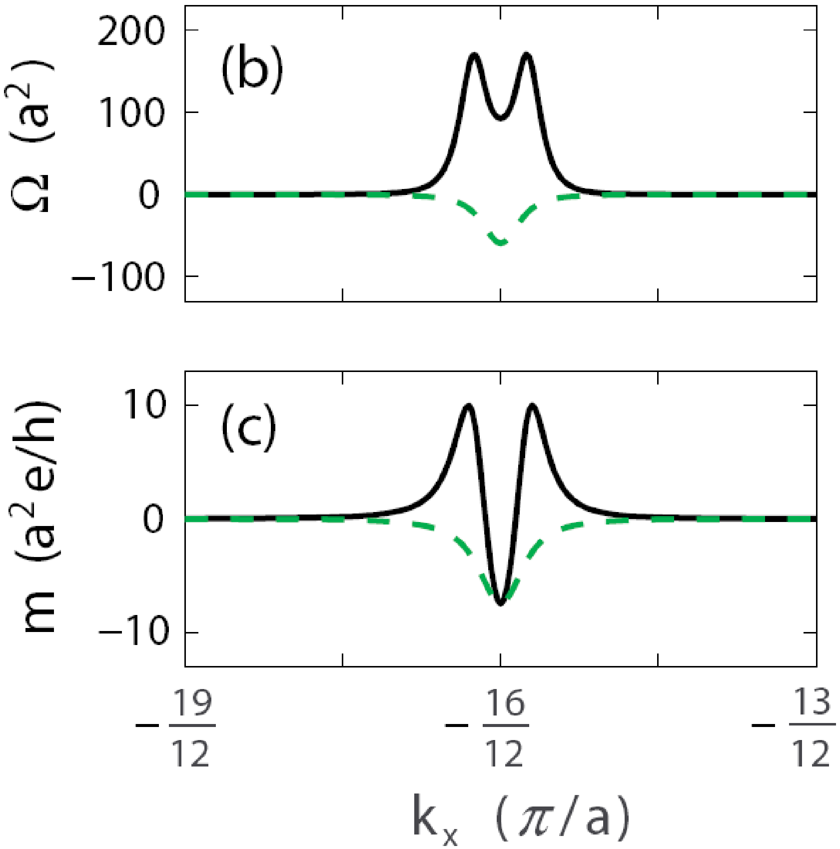
● sublattice A2 $\xleftrightarrow{t}$ ● sublattice B2 $\frac{\Delta}{2}$

Berry Curvature and Orbital Moment in BGB



ARPES, Rotenberger group, 06"

valley K_1, conduction

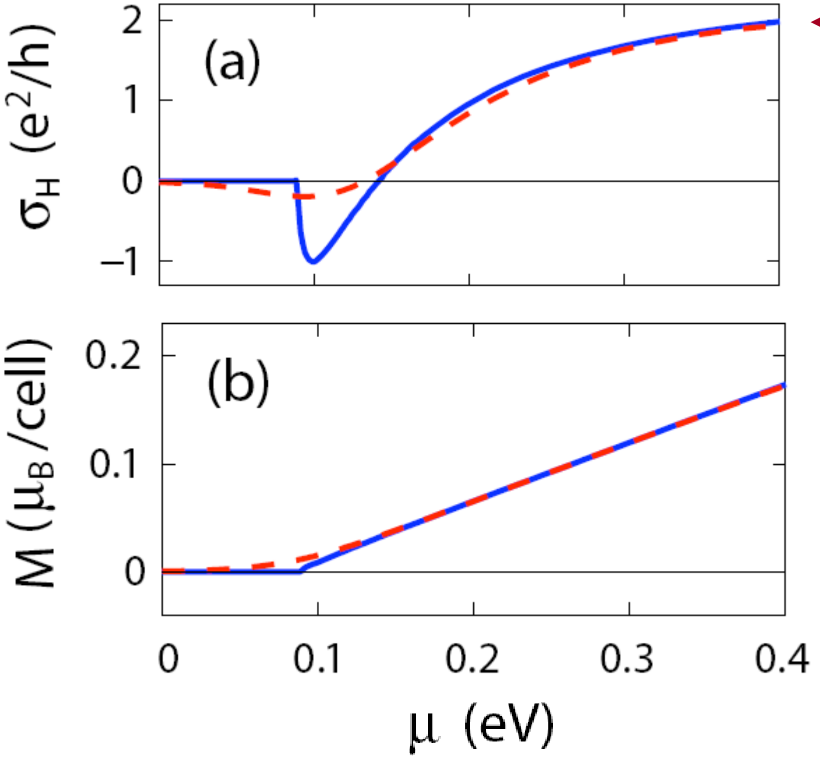


Opposite $\Omega(k)$ and $m(k)$ at valley K_2

Valance band: opposite $\Omega(k)$ but same $m(k)$

Xiao, Yao & Niu, PRL 07"

Valley Hall Conductance in BGB



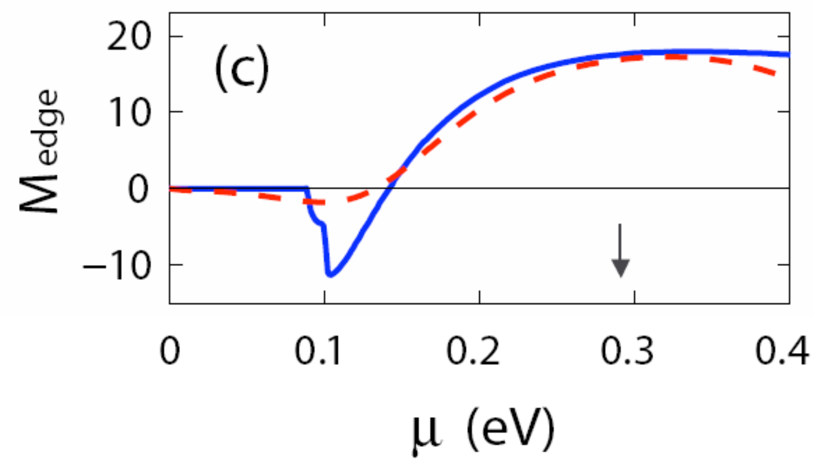
← approaching quantized value

Berry phase of 2π in bilayer

(left up) Hall conductance &
(left down) orbital magnetization
in valley K_1

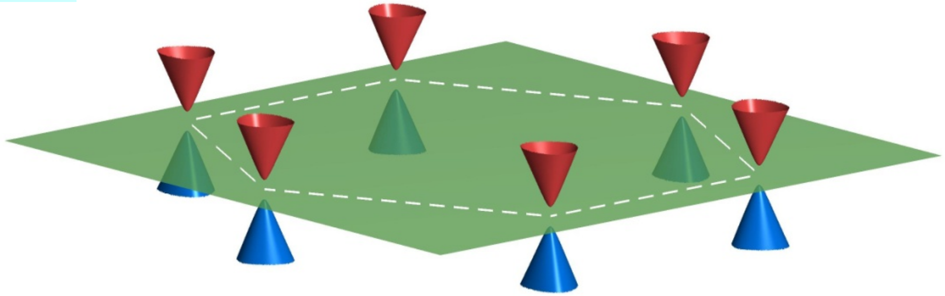
(right) edge magnetization in
Hall geometry

$$M_{edge} = 2\frac{\partial M}{\partial \mu}\frac{\partial \mu}{\partial n}\sigma_H^v \tau_v E_y$$



Optical Interband Transitions?

Energy dispersion



- Valley contrasted magnetic moment and Hall current
- Finite bandgap -> optical interband transitions
- In atoms: selection rule by magnetic moment
- Selection rule in III-V material: inheritance from parent orbitals
- Graphene: c-band and v-band originate from the same orbital

Orbital Moment and Circular Dichroism

- Light-matter coupling in Bloch bands

$$\hat{\mathcal{H}}_{eR} = e/mc\boldsymbol{A}\cdot\hat{\boldsymbol{p}} \qquad \mathcal{P}_{cv}^{\pm} \equiv \langle u_{c,\boldsymbol{k}}|\hat{p}_x \pm i\hat{p}_y|u_{v,\boldsymbol{k}}\rangle = \frac{m_0}{\hbar}\langle u_{c,\boldsymbol{k}}|\frac{\partial H}{\partial k_x} \pm i\frac{\partial H}{\partial k_y}|u_{v,\boldsymbol{k}}\rangle$$

- Oscillator strength (k-resolved):

Difference

$$\frac{|\mathcal{P}_{cv}^{+}(\boldsymbol{k})|^2 - |\mathcal{P}_{cv}^{-}(\boldsymbol{k})|^2}{m_0\left(\varepsilon_c(\boldsymbol{k}) - \varepsilon_v(\boldsymbol{k})\right)} = -2\frac{m(\boldsymbol{k})}{\mu_B}$$

Sum

$$\frac{|\mathcal{P}_{cv}^{+}(\boldsymbol{k})|^2 + |\mathcal{P}_{cv}^{-}(\boldsymbol{k})|^2}{2m_0\left(\varepsilon_c(\boldsymbol{k}) - \varepsilon_v(\boldsymbol{k})\right)} = m_0\mathrm{Tr}\left[\frac{1}{2}\frac{\partial^2\varepsilon_c(\boldsymbol{k})}{\hbar^2\partial k_i\partial k_j}\right]$$

- Degrees of circular polarization:

$$\eta(\boldsymbol{k}) \equiv \frac{|\mathcal{P}_{cv}^{+}(\boldsymbol{k})|^2 - |\mathcal{P}_{cv}^{-}(\boldsymbol{k})|^2}{|\mathcal{P}_{cv}^{+}(\boldsymbol{k})|^2 + |\mathcal{P}_{cv}^{-}(\boldsymbol{k})|^2} = -\frac{m(\boldsymbol{k})}{\mu_B^*(\boldsymbol{k})}$$

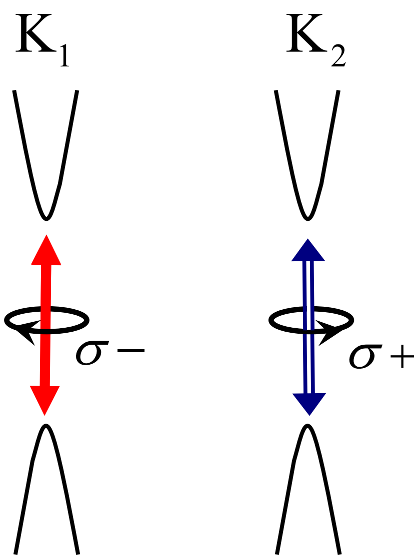
Valley Contrasting Optical Selection Rules

- Matrix element of interband transition

$$|\mathcal{P}_{cv}^{\pm}(\boldsymbol{k})|^2 = m_0^2 v_0^2 (1 \mp \tau_z \cos\theta)^2$$

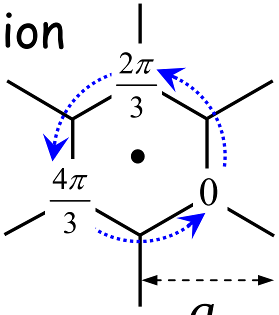
$\cos\theta \simeq 1$ near Dirac points

Valley optical selection rule



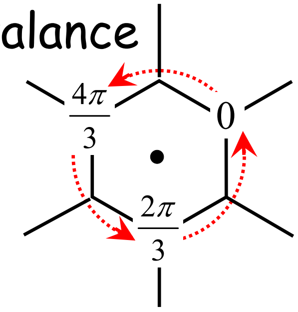
Phase winding of Bloch function at K_1

conduction



$l_c = 1$

valance



$l_v = -1$

Effective azimuthal rule

$$l_v + j = l_c + 3N$$

WY, Xiao & Niu, arXiv:0705.4683

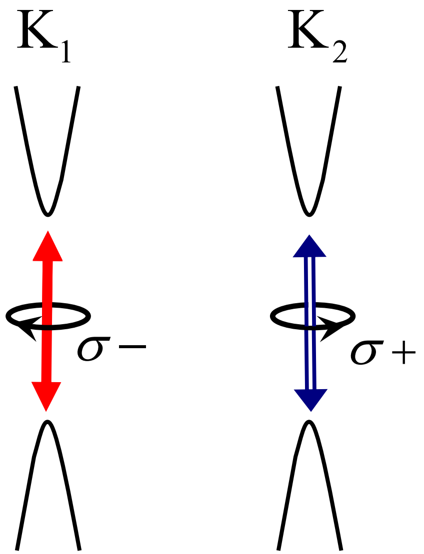
Valley Contrasting Optical Selection Rules

- Matrix element of interband transition

$$|\mathcal{P}^{\pm}_{cv}(\boldsymbol{k})|^2 = m_0^2 v_0^2 (1 \mp \tau_z \cos\theta)^2$$

$\cos\theta \simeq 1$ near Dirac points

Valley optical selection rule



- Optical strength

$|\mathcal{P}_{cv}|^2 / m_0 \sim 5\ \text{eV}$ (~ 21.5 eV in GaAs)

- Far away from Dirac points

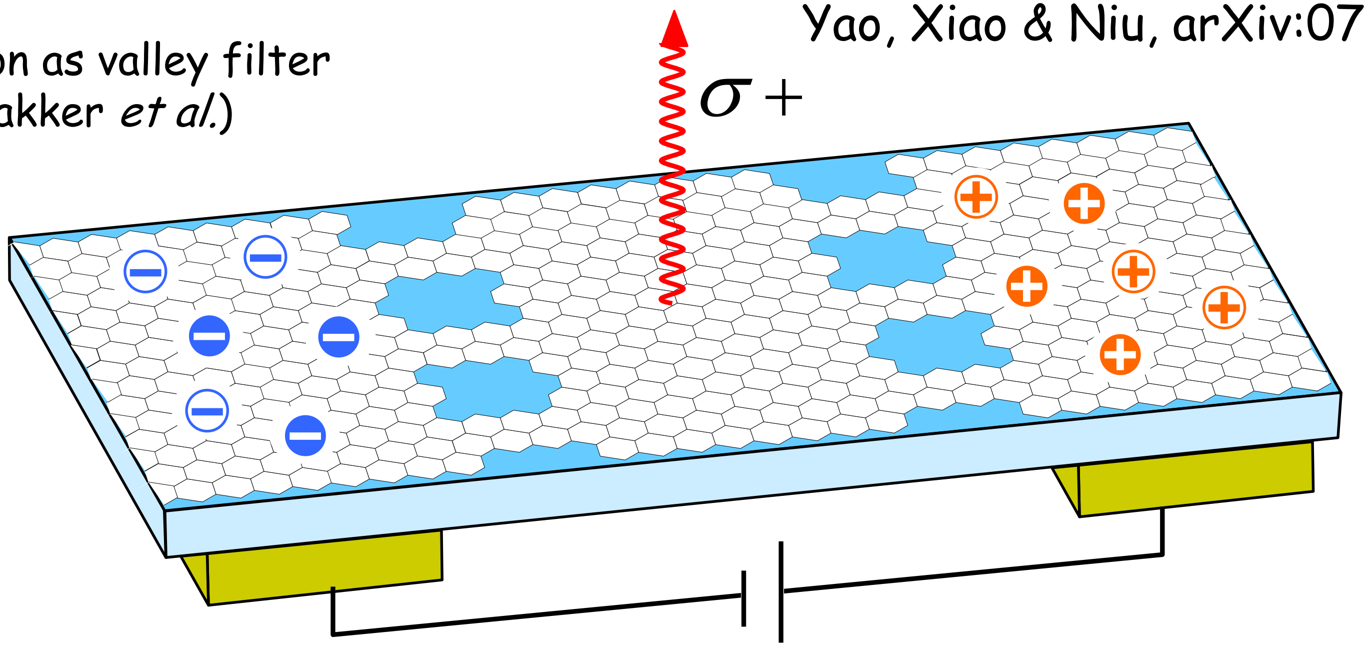
No circular dichroism, constant high frequency optical conductivity

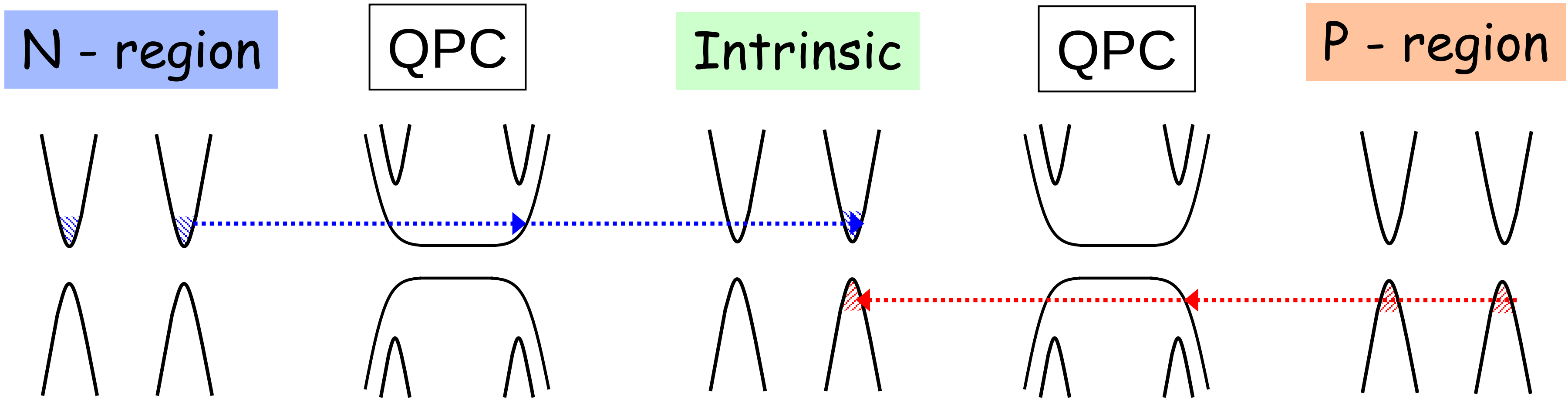
WY, Xiao & Niu, arXiv:0705.4683

Valley LED

Nano-ribbon as valley filter
(Beenakker *et al.*)

Yao, Xiao & Niu, arXiv:0705.4683





N - region

QPC

Intrinsic

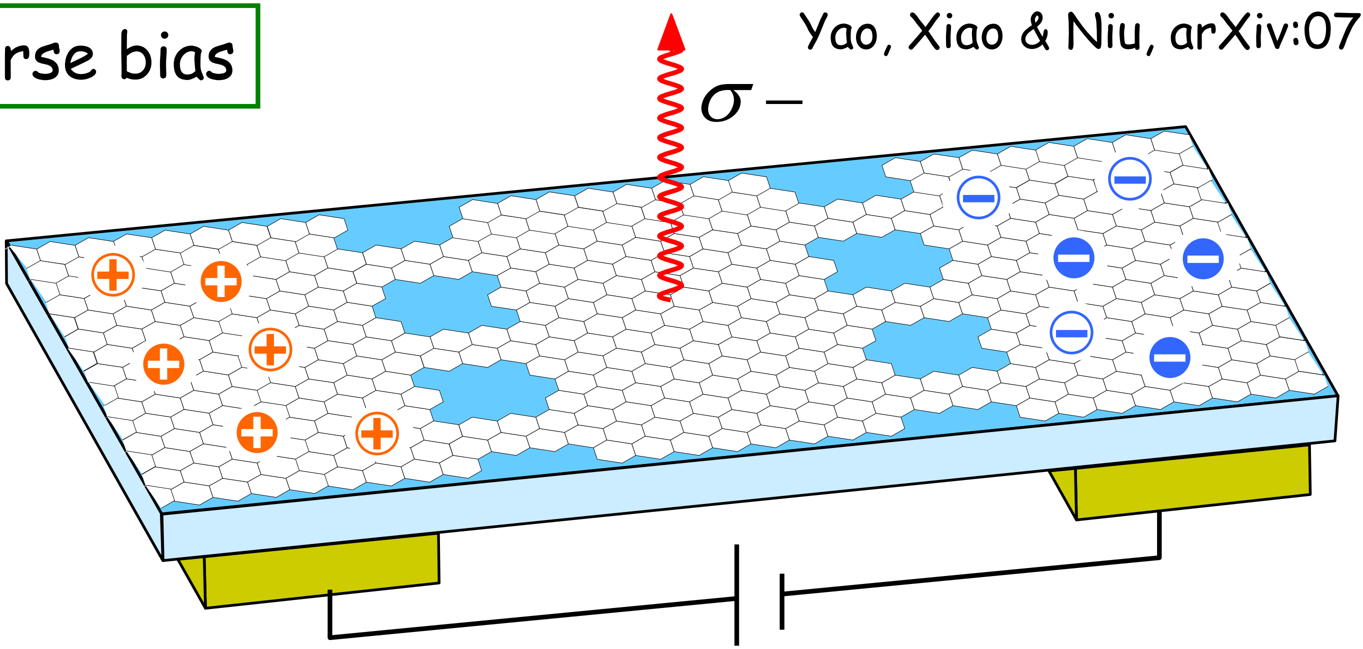
QPC

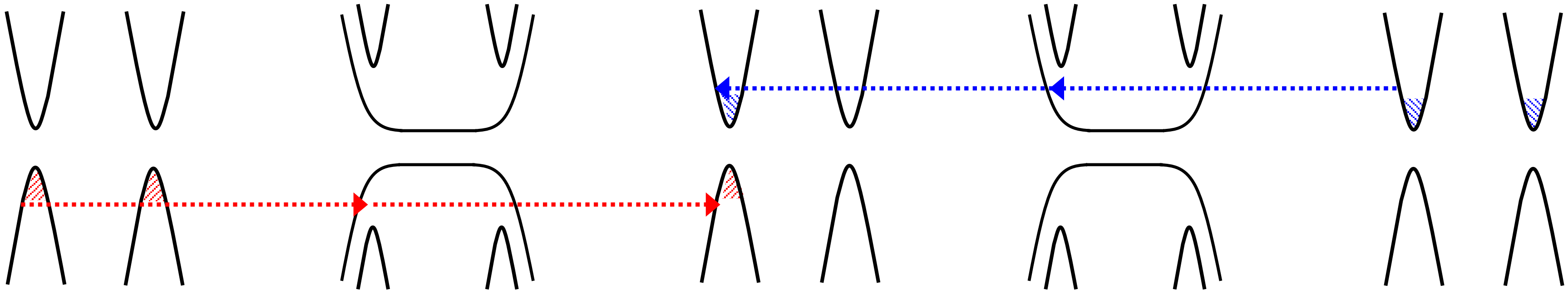
P - region

Valley LED

Reverse bias

Yao, Xiao & Niu, arXiv:0705.4683





P - region

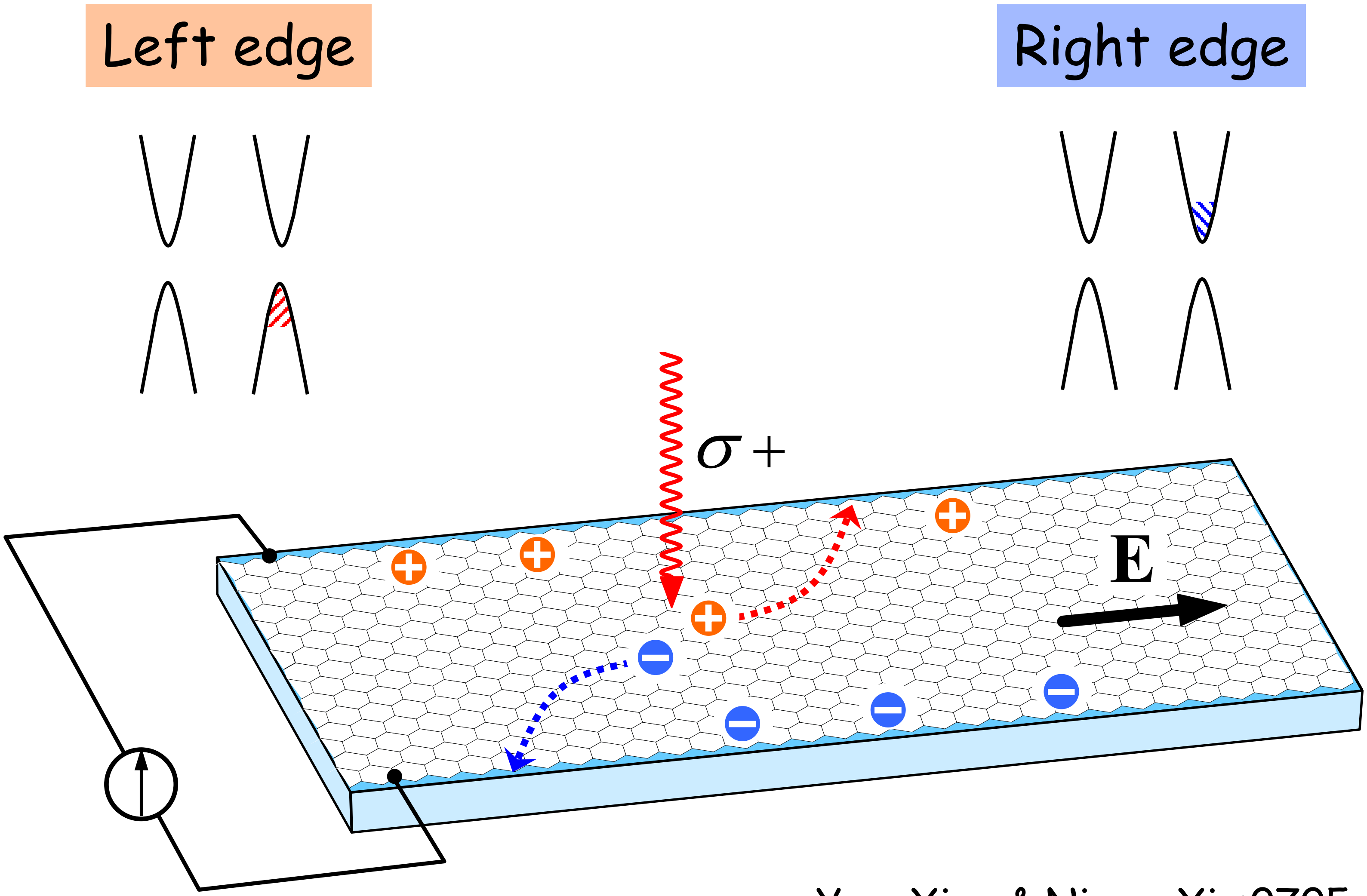
QPC

Intrinsic

QPC

N - region

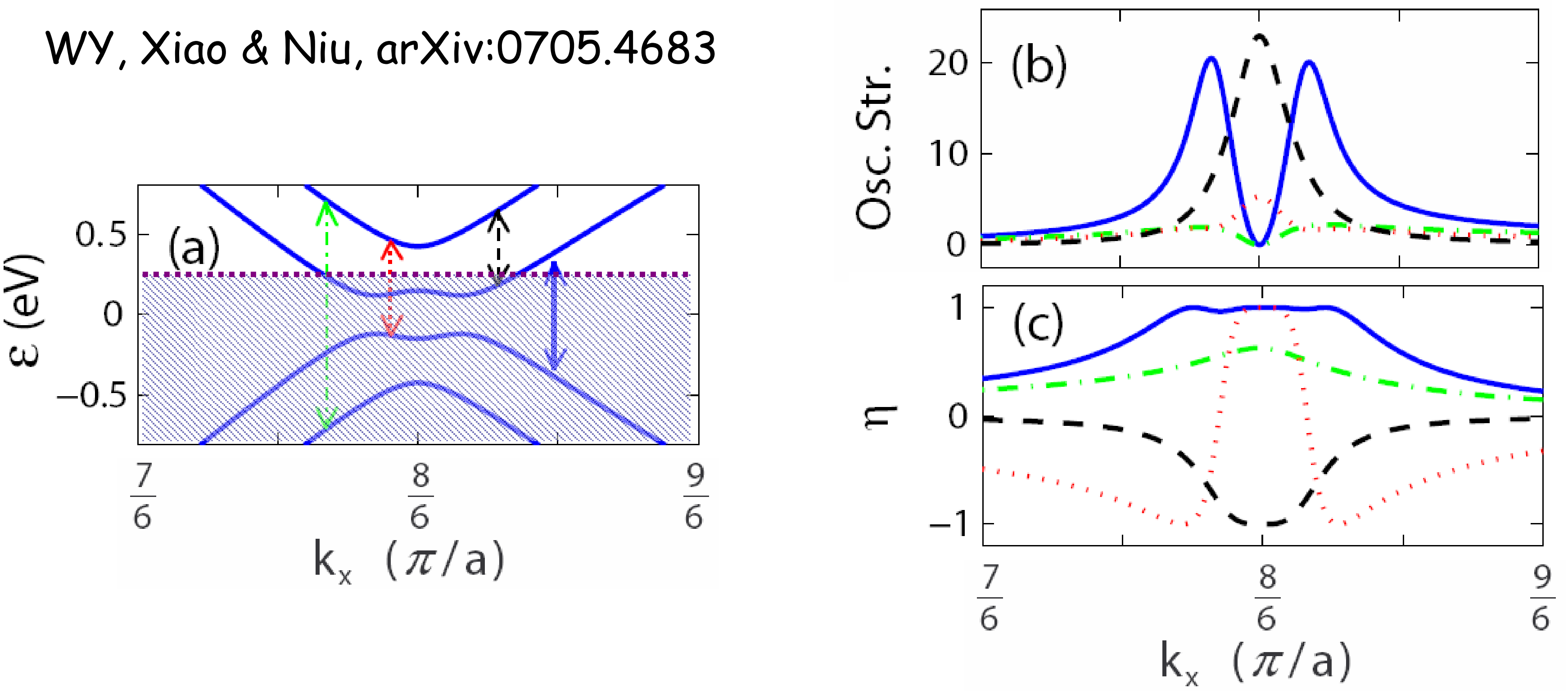
Photo-induced Anomalous Hall Effect



Yao, Xiao & Niu, arXiv:0705.4683

Bilayer with Interlayer Gate Voltage

WY, Xiao & Niu, arXiv:0705.4683



- Selection rule for transition between conduction bands
- Valley optoelectronics in metallic system

Dichroic Sum Rules for Ferromagnets

- Interband optical transition, magnetic moment & Berry curvature

$$\eta(\boldsymbol{k}) = -\frac{\boldsymbol{m}(\boldsymbol{k})\cdot\hat{z}}{\mu_B^*(\boldsymbol{k})} = -\frac{\boldsymbol{\Omega}(\boldsymbol{k})\cdot\hat{z}}{\mu_B^*(\boldsymbol{k})}(\varepsilon_c(\boldsymbol{k})-\varepsilon_i(\boldsymbol{k}))\frac{e}{2\hbar}$$

- Dichroism and orbital magnetization

$$\frac{\mu_B}{2}(\langle f_-\rangle - \langle f_+\rangle) = \hat{z}\cdot\int_{BZ}\frac{d\boldsymbol{k}}{(2\pi)^d}g(\boldsymbol{k})\boldsymbol{m}(\boldsymbol{k}),$$

Total oscillator strength: $$\langle f_\pm\rangle \equiv \sum_i \int_{BZ}\frac{d\boldsymbol{k}}{(2\pi)^d}g(\boldsymbol{k})\frac{\left|\mathcal{P}_x^{ci}(\boldsymbol{k}) \pm \mathcal{P}_y^{ci}(\boldsymbol{k})\right|^2}{m_e\left(\varepsilon_c(\boldsymbol{k})-\varepsilon_i(\boldsymbol{k})\right)}.$$

- Dichroism and anomalous Hall conductivity

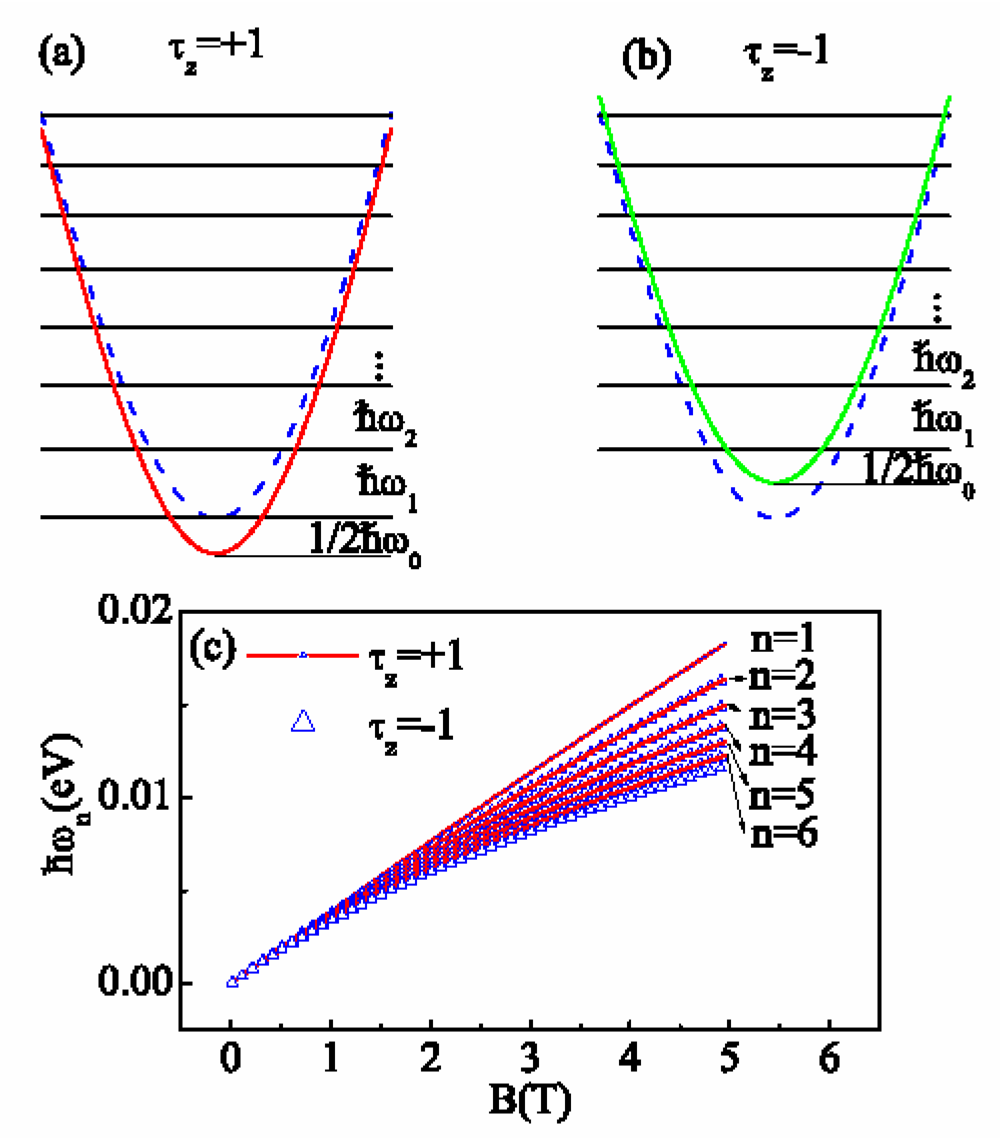
$$\sigma_H = \frac{\epsilon_0}{\pi}\int d\omega(\epsilon_-^i(\omega) - \epsilon_+^i(\omega))$$

Interband absorptions: $$\epsilon_\pm^i(\omega) = \frac{\pi e^2}{\epsilon_0 m_e^2\omega^2}\sum_i\int_{BZ}\frac{d\boldsymbol{k}}{(2\pi)^2}g(\boldsymbol{k})\left|\mathcal{P}_x^{ci}(\boldsymbol{k})\pm i\mathcal{P}_y^{ci}(\boldsymbol{k})\right|^2\delta(\varepsilon_c(\boldsymbol{k})-\varepsilon_i(\boldsymbol{k})-\hbar\omega)$$

Summary

- Electrons classified by valley index in graphene
- Valley contrasting topological properties from inversion symmetry breaking
- Valley analog of spin electronics and spin optoelectronics
- Generalization to other non-central valley semiconductors, Si or AlAs

Quantum calculations of Landau Levels



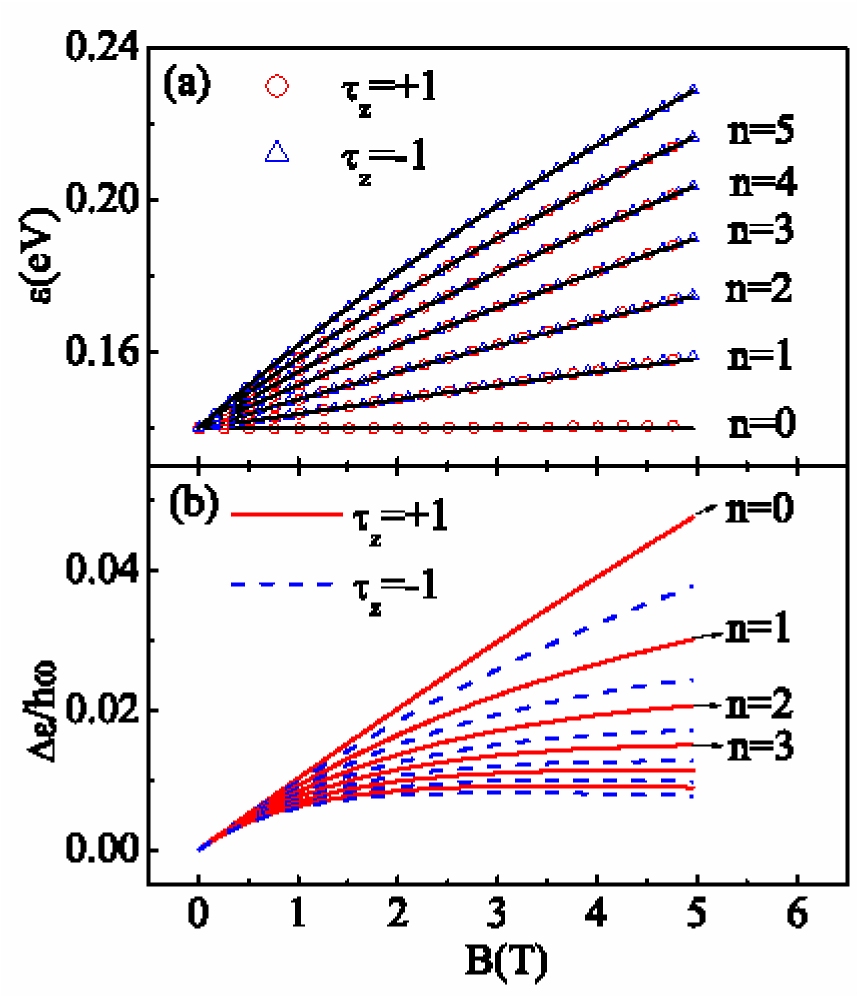
$$E_n = \pm\sqrt{\left(\frac{\Delta}{2}\right)^2 + \frac{3a^2t^2eB}{4\hbar}\left(2n+1-\tau_z\right)}$$

- The lowest LLs are located at $\frac{1}{2}\hbar\omega^*$

$$\omega^* = \frac{2\hbar\Delta}{3a^2t^2}$$

- E_{n+1}-E_n is not a constant, which is related to the Berry curvature.

The comparison in Landau levels



- The field dependence of LLs, E_{semi} (red circle and blue triangle) is obtained from the semiclassical quantum condition.

$$A = \frac{2\pi e B}{\hbar}\left(n + \frac{1}{2} - \frac{\Gamma}{2\pi}\right)$$

- For comparison, the quantum results are also shown (solid line).