Quantum size effects and optical transitions in topological-insulator nanostructures

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Outline

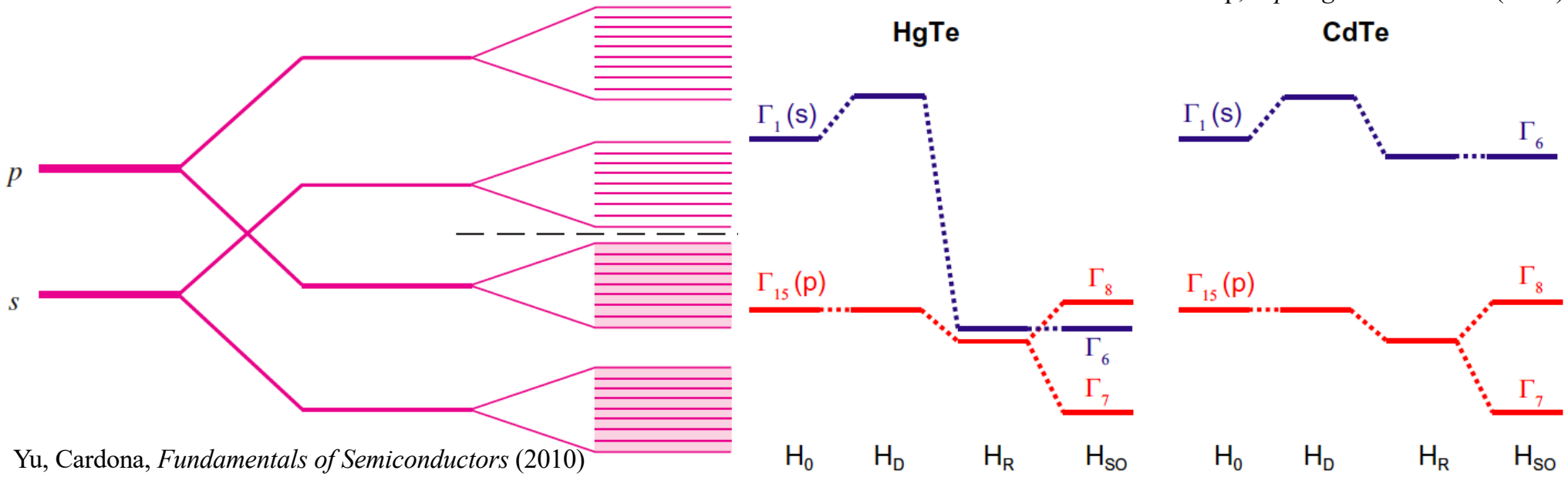
- Introduction & Motivation
  - topological insulators: inverted bulk band structure
  - Dirac-like charge carriers: BHZ model Hamiltonian
- Quantum size effects in topological-insulator nanostructures: quantum wells/rings/nanoparticles
  - fate of topological (sub-)bands & surface states
  - observable consequences: gap oscillations (2D wells), conductance oscillations (1D rings), optical selection rules & transition probabilities (0D nanoparticles)
- Conclusions

Introduction & Motivation

Topological insulators: Bulk band inversion

- atomic levels broaden into bands in solid material
  - (anti-)bonding levels → (conduction) valence bands
- in some materials, relativistic effects reverse order of bonding/anti-bonding bands: band inversion

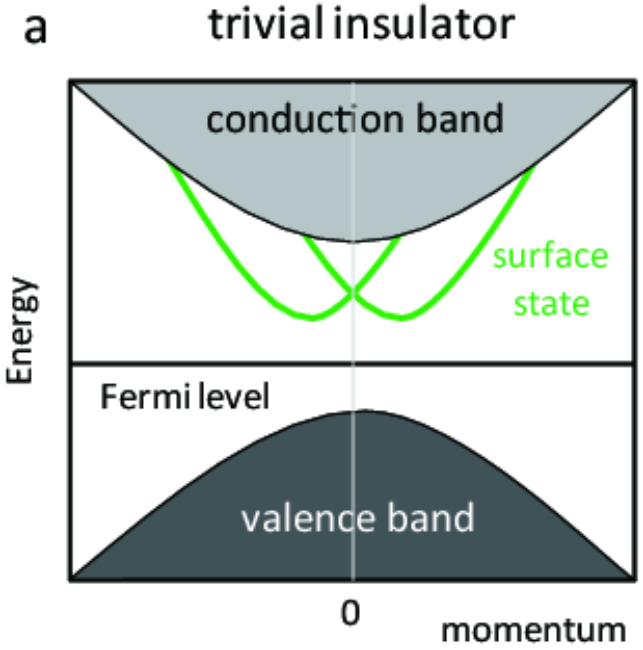
Franz & Molenkamp, *Topological Insulators* (2013)

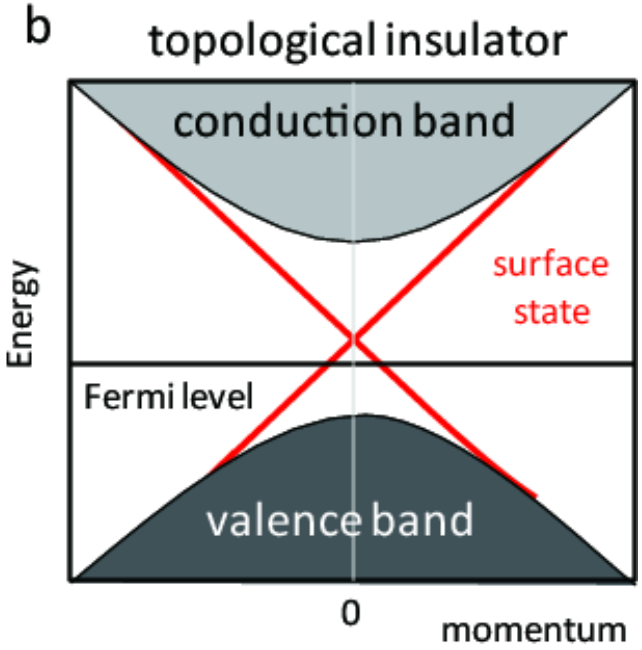


Yu, Cardona, *Fundamentals of Semiconductors* (2010)

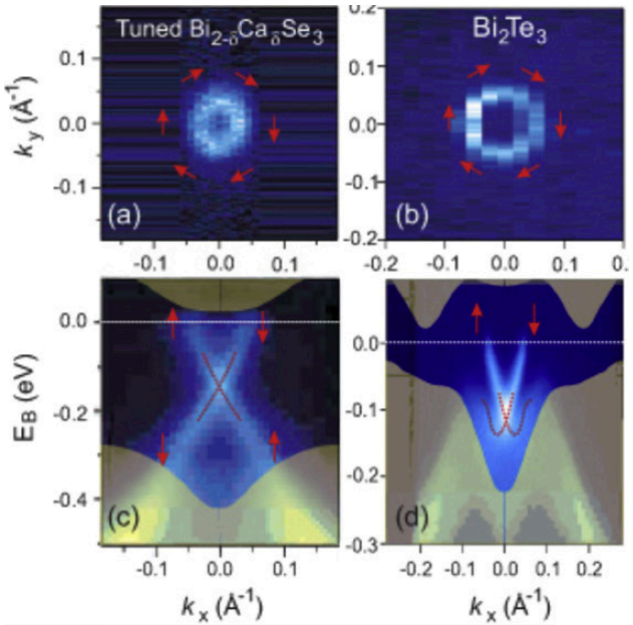
Ordinary vs. topological insulator

- closing of gap required to go from ordinary to the inverted situation: topologically distinct systems!
- gapless states exist at the surface of a topological material (= interface with an ordinary material!)





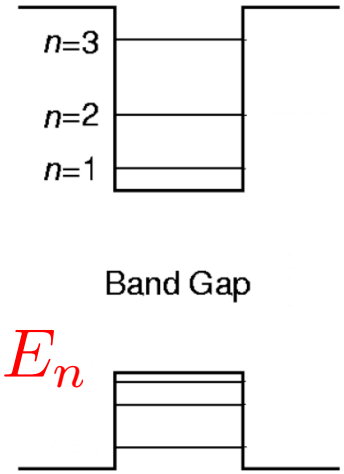
Hasan et al. Phys. Scr. (2015)



www.scholarpedia.org/article/Topological_insulators

Size quantization counteracts band inversion

- quantum bound-state energy adds to bulk band edge: new (quantum-well) sub-bands

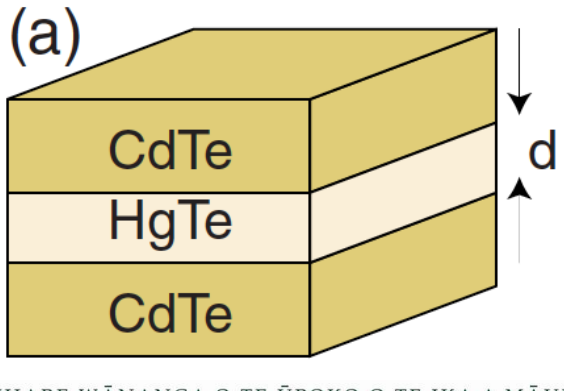


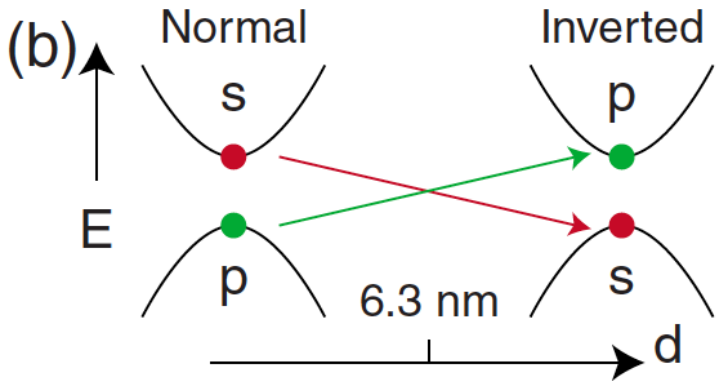
$$\frac{p_x^2+p_y^2}{2m}+\frac{\Delta_0}{2}+\frac{p_z^2}{2m}+V(z)\longrightarrow\frac{p_x^2+p_y^2}{2m}+\frac{\Delta_0}{2}+E_n$$

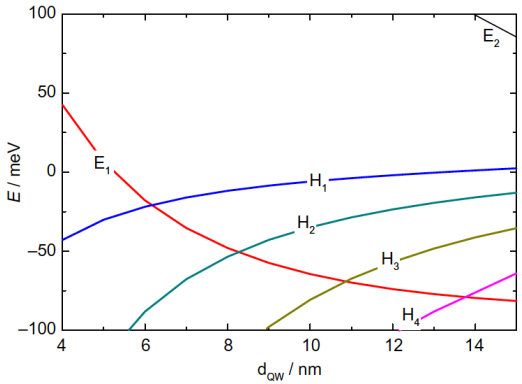
- HgTe quantum well: bulk gap $\Delta_0 < 0$; adjust well width d to tune btw. normal & inverted regimes

Bernevig, Hughes & Zhang, Science (2006); König et al., Science (2007)

Hasan & Kane, RMP (2010)







Franz & Molenkamp (2013)

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$\mathbf{k}\cdot\mathbf{p}$ theory for Dirac-like charge carriers

- topological insulators generally host two-flavour Dirac quasiparticles (pseudospin τ & real spin σ)

$$H = \epsilon(\mathbf{k})\,\mathbb{1}_{4\times4} + \begin{pmatrix} \frac{\Delta(\mathbf{k})}{2} & \gamma\,k_- & 0 & \gamma'\,k_z \\ \gamma\,k_+ & -\frac{\Delta(\mathbf{k})}{2} & -\gamma'\,k_z & 0 \\ 0 & -\gamma'\,k_z & \frac{\Delta(\mathbf{k})}{2} & \gamma\,k_+ \\ \gamma'\,k_z & 0 & \gamma\,k_- & -\frac{\Delta(\mathbf{k})}{2} \end{pmatrix}$$

- includes 2D/3D motion, particle-hole asymmetry

BHZ, Science (2006); Liu et al., Phys. Rev. B (2010); Brems et al., New J. Phys. (2018)

Confining Dirac-like quasiparticles

- two possibilities: use a scalar or a vector potential
  Greiner, *Relativistic Quantum Mechanics* (1990); Alberto et al., Eur. J. Phys (1996)
  - vector potential models electrostatic (e.g. gate-defined) confinement, is not entirely confining (Klein paradox!)
  - scalar potential actually models a finite materials size
- adopt scalar (i.e., mass-confinement) potential!
  - hard-wall, or parabolic, etc. functional form for $V(\mathbf{r})$

$$\mathcal{H} = [\epsilon(\mathbf{k}) + U(\mathbf{r})]\,\mathbb{1}_{4\times4} + \begin{pmatrix} \frac{\Delta(\mathbf{k})}{2} + V(\mathbf{r}) & \gamma\, k_- & 0 & \gamma'\, k_z \\ \gamma\, k_+ & -\frac{\Delta(\mathbf{k})}{2} - V(\mathbf{r}) & -\gamma'\, k_z & 0 \\ 0 & -\gamma'\, k_z & \frac{\Delta(\mathbf{k})}{2} + V(\mathbf{r}) & \gamma\, k_+ \\ \gamma'\, k_z & 0 & \gamma\, k_- & -\frac{\Delta(\mathbf{k})}{2} - V(\mathbf{r}) \end{pmatrix}$$

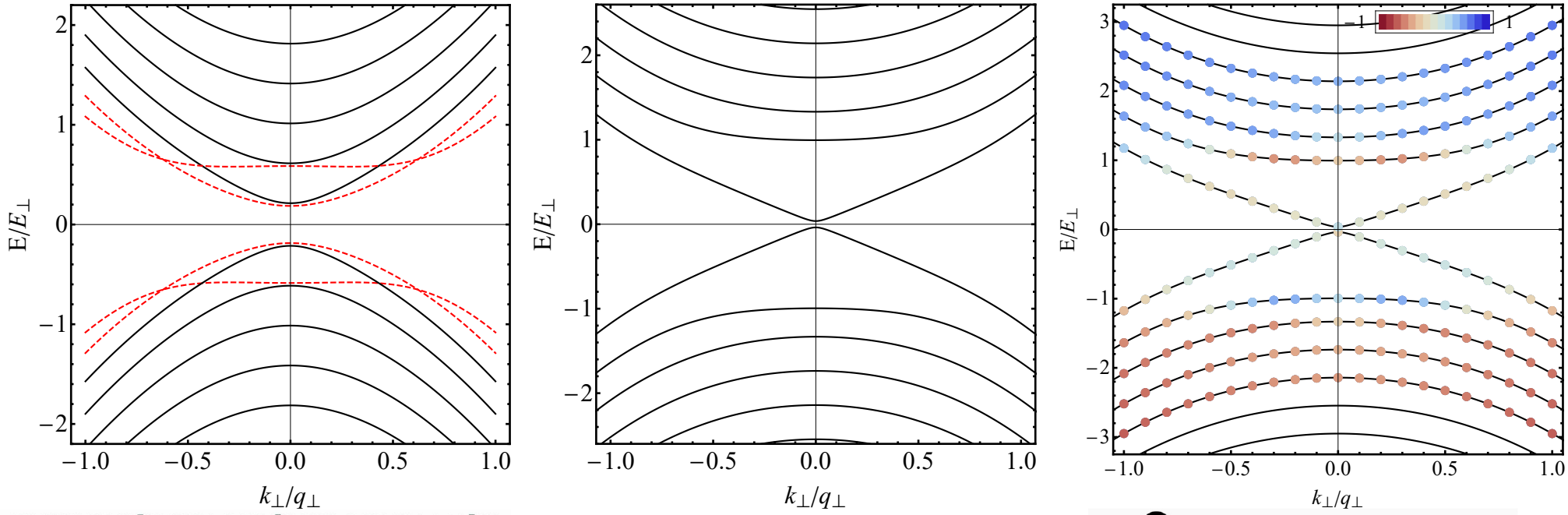
Quasi-2D confinement:

Gap oscillations in Bi_2Se_3-type topological-insulator quantum wells

Size-quantized subbands vs. surface states

- interplay of band-edge renormalisation & mixing

$$\mathcal{H} = \begin{pmatrix} \frac{\Delta(\mathbf{k}_\parallel)}{2} - \frac{\hbar^2}{2M_\perp}\partial_z^2 + V(z) & \gamma\, k_- & 0 & \gamma'(-i\hbar\partial_z) \\ \gamma\, k_+ & -\frac{\Delta(\mathbf{k}_\parallel)}{2} + \frac{\hbar^2}{2M_\perp}\partial_z^2 - V(z) & -\gamma'(-i\hbar\partial_z) & 0 \\ 0 & -\gamma'(-i\hbar\partial_z) & \frac{\Delta(\mathbf{k}_\parallel)}{2} - \frac{\hbar^2}{2M_\perp}\partial_z^2 + V(z) & \gamma\, k_+ \\ \gamma'(-i\hbar\partial_z) & 0 & \gamma\, k_- & -\frac{\Delta(\mathbf{k}_\parallel)}{2} + \frac{\hbar^2}{2M_\perp}\partial_z^2 - V(z) \end{pmatrix}$$

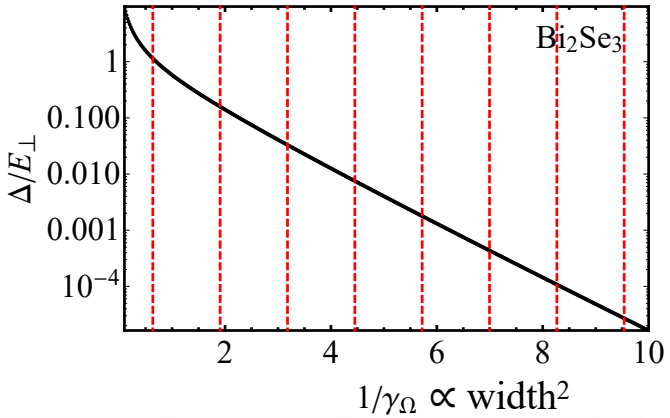


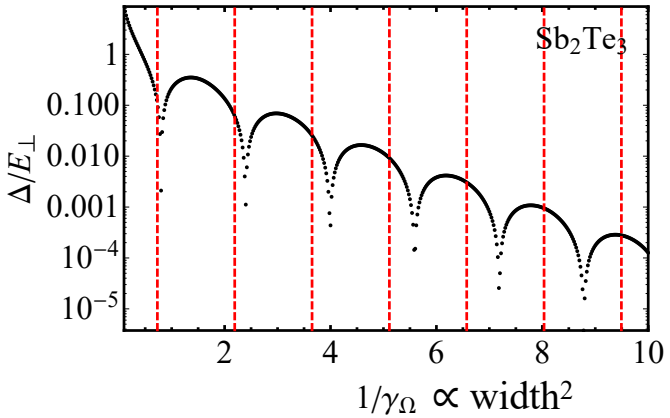
Material-dependent stability of surface states

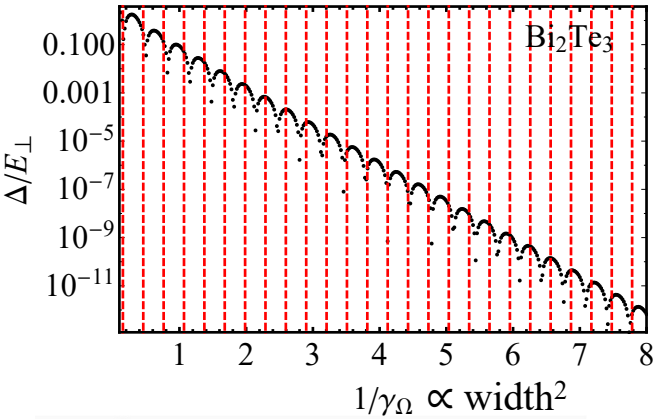
- Bi_2Se_3-type materials show variety of behavior

  Kotulla & UZ, New J. Phys. (2017)

  - Bi_2Se_3 maintains 3D topological-insulator features until band inversion is fully destroyed by confinement
  - Sb_2Te_3 has "clean" 2D topological transition similar to that exhibited by HgTe/CdTe quantum well
  - Bi_2Te_3 remains inverted even at smallest layer width







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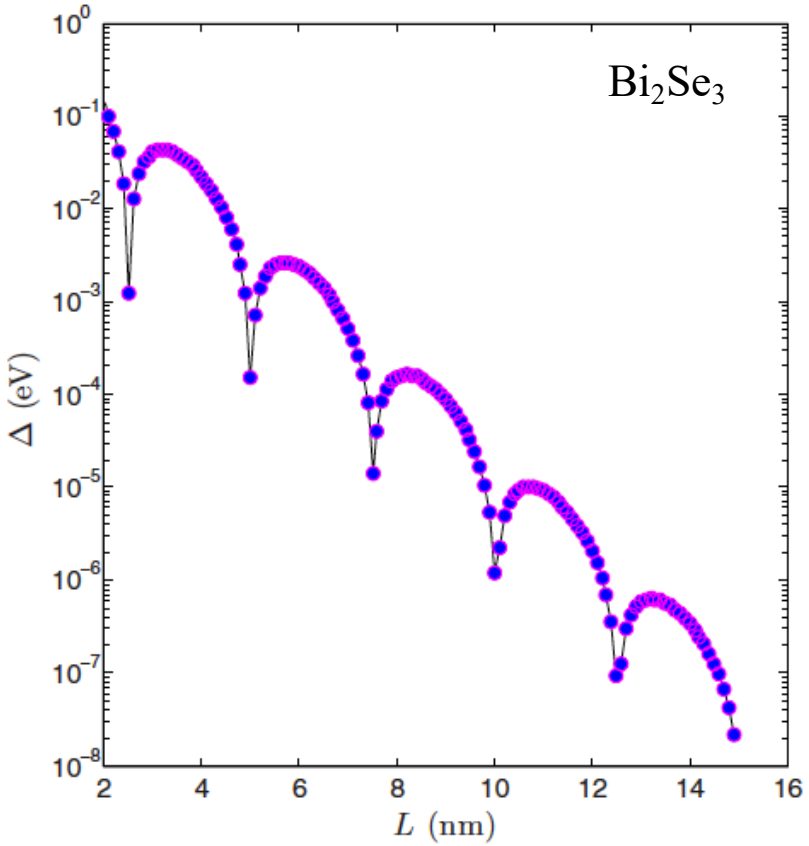
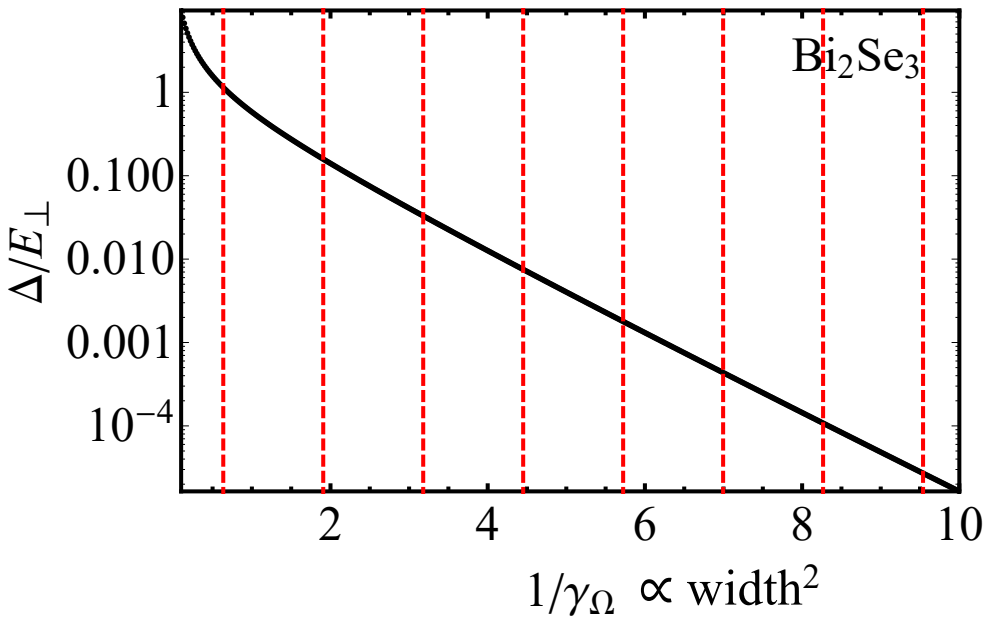
Sensitivity to bulk-bandstructure parameters



Linder et al., Phys. Rev. B (2009)

[band-structure parameters from Zhang et al., Nat. Phys. (2009)]



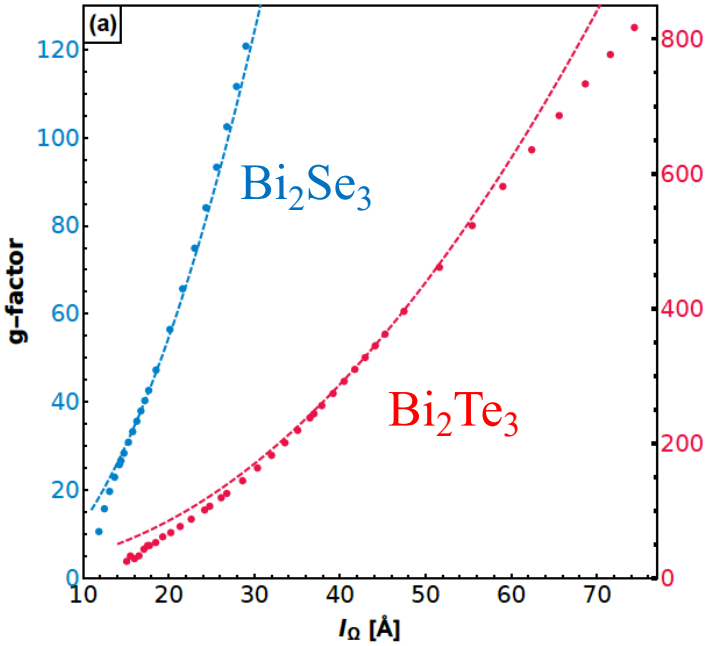
Kotulla & UZ, New J. Phys. (2017)

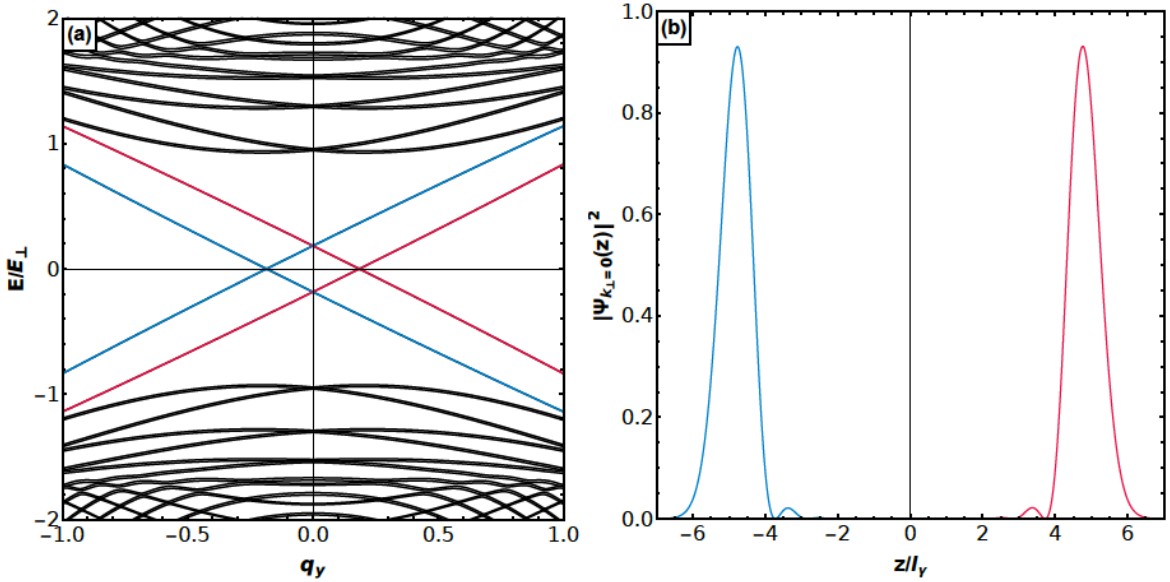
[band-structure parameters from Nechaev & Krasovskii, PRB (2016)]

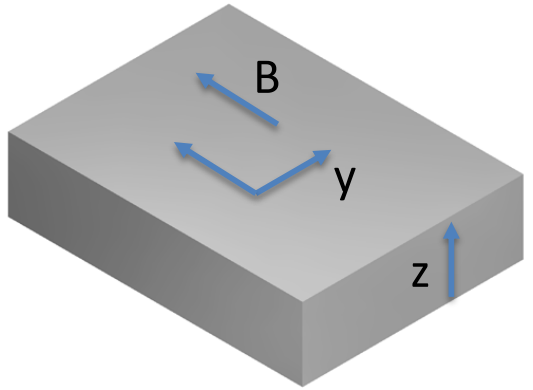
In-plane **B**: Giant surface-state Zeeman splitting

- energy splitting due to in-plane magnetic field much larger for surface states than higher bands
  - large effective *g*-factor

Kotulla, PhD thesis (2019)



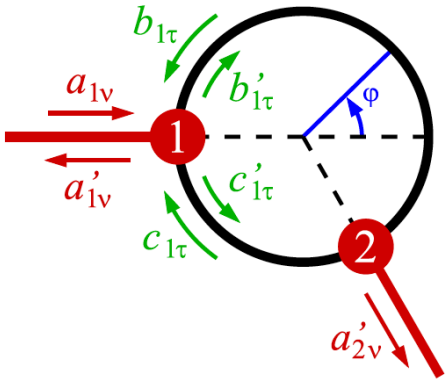




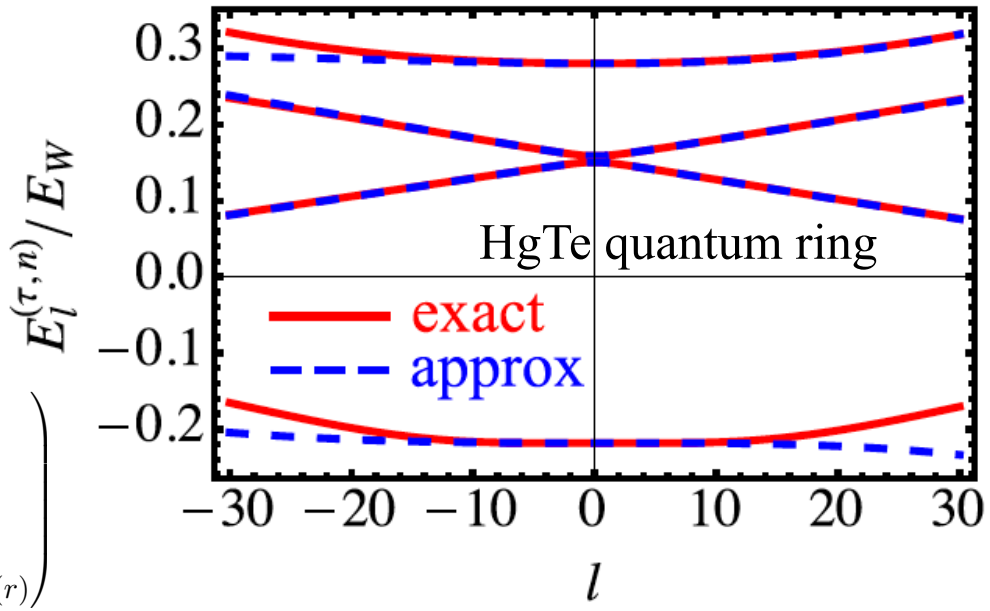
Quasi-1D confinement:
Conductance oscillations in quantum-ring structures

2D Dirac-like electrons in quantum rings

- realizable, e.g., in graphene, HgTe quantum wells
  Recher et al., Phys. Rev. B (2007); Michetti & Recher, Phys. Rev. B (2011)
  - generically broken valley/real spin-reversal symmetry
- can obtain most general effective quasi-1D Dirac Hamiltonian for ring subbands Gioia, UZ, et al., PRB (2018)

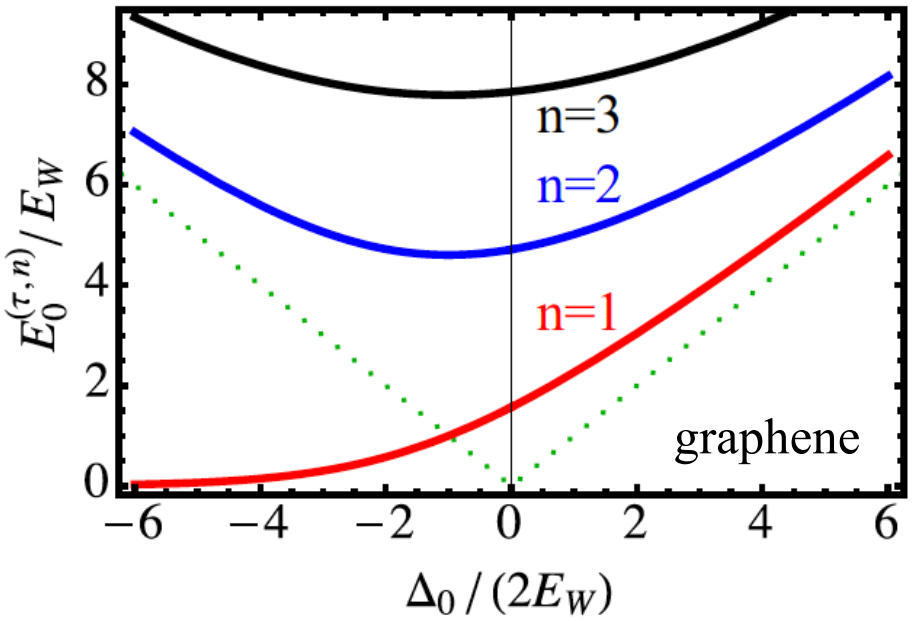


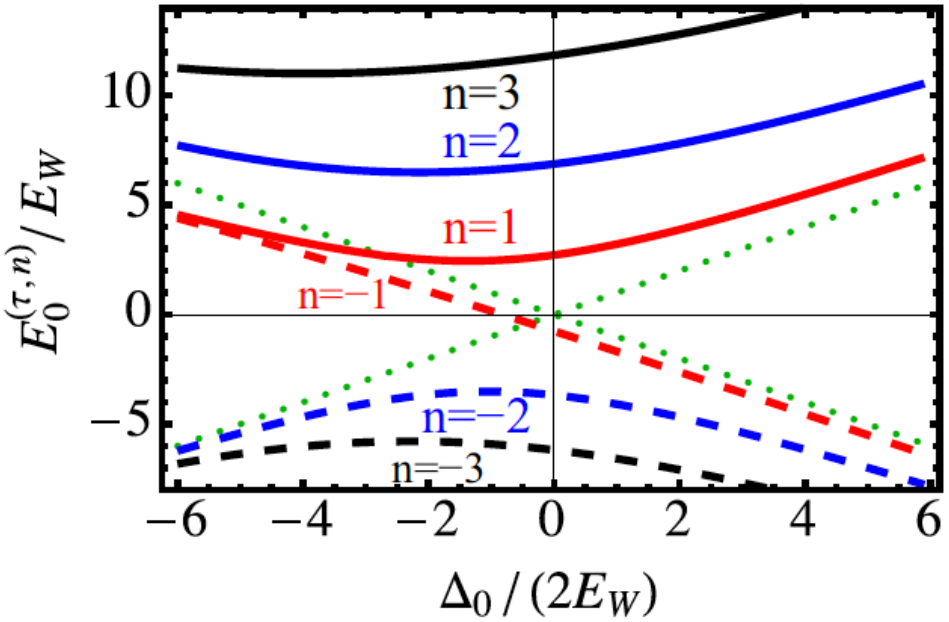
$$\mathcal{H} = \epsilon(\mathbf{k})\,\mathbb{1}_{4\times4} + \begin{pmatrix} \frac{\Delta(\mathbf{k})}{2} + V(r) & \gamma\, k_- & 0 & 0 \\ \gamma\, k_+ & -\frac{\Delta(\mathbf{k})}{2} - V(r) & 0 & 0 \\ 0 & 0 & \frac{\Delta(\mathbf{k})}{2} + V(r) & \gamma\, k_+ \\ 0 & 0 & \gamma\, k_- & -\frac{\Delta(\mathbf{k})}{2} - V(r) \end{pmatrix}$$



Topological regime: Effect of band inversion

- lowest quasi-1D subband energy is below the 2D-bulk band edge if $-\Delta_0/2 \lesssim E_W = \gamma/W$ (W: ring width)





$$\mathcal{H} = \epsilon(\mathbf{k})\,\mathbb{1}_{4\times4} + \begin{pmatrix} \frac{\Delta(\mathbf{k})}{2} + V(r) & \gamma\, k_- & 0 & 0 \\ \gamma\, k_+ & -\frac{\Delta(\mathbf{k})}{2} - V(r) & 0 & 0 \\ 0 & 0 & \frac{\Delta(\mathbf{k})}{2} + V(r) & \gamma\, k_+ \\ 0 & 0 & \gamma\, k_- & -\frac{\Delta(\mathbf{k})}{2} - V(r) \end{pmatrix}$$

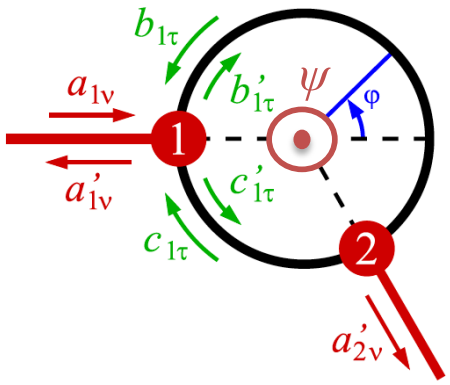
7-nm HgTe quantum well

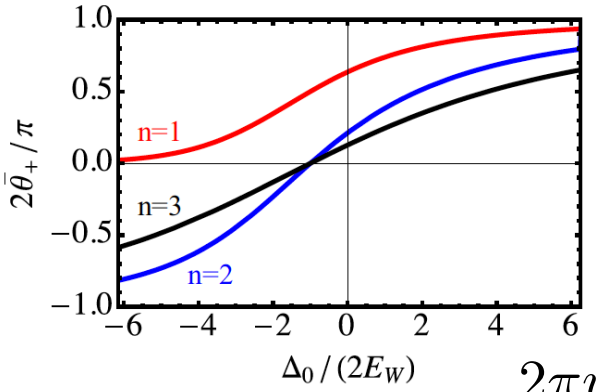
[Rothe et al., NJP (2010)]

Dirac-ring conductance oscillations

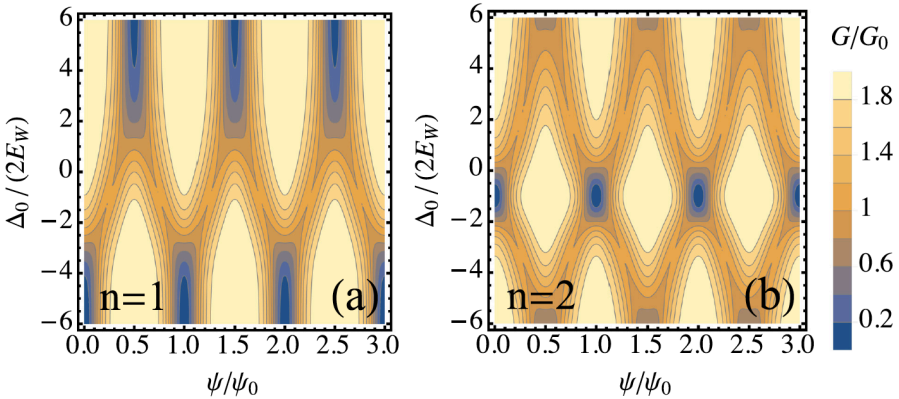
- interference contribution to conductance tuned by enclosed magnetic flux ψ Büttiker et al., Phys. Rev. A (1984)
- geometric (Aharonov-Anandan) phase revealed in ring-conductance oscillations Frustaglia & Richter, PRB (2004)
- Dirac ring: AA phase confinement-dependent and reflects topological property of lowest subband

Gioia, UZ et al., Phys. Rev. B (2018)





$$\theta_{\mathrm{AA}} = 2\theta_+ + \pi - \frac{2\pi\psi}{\psi_0}$$



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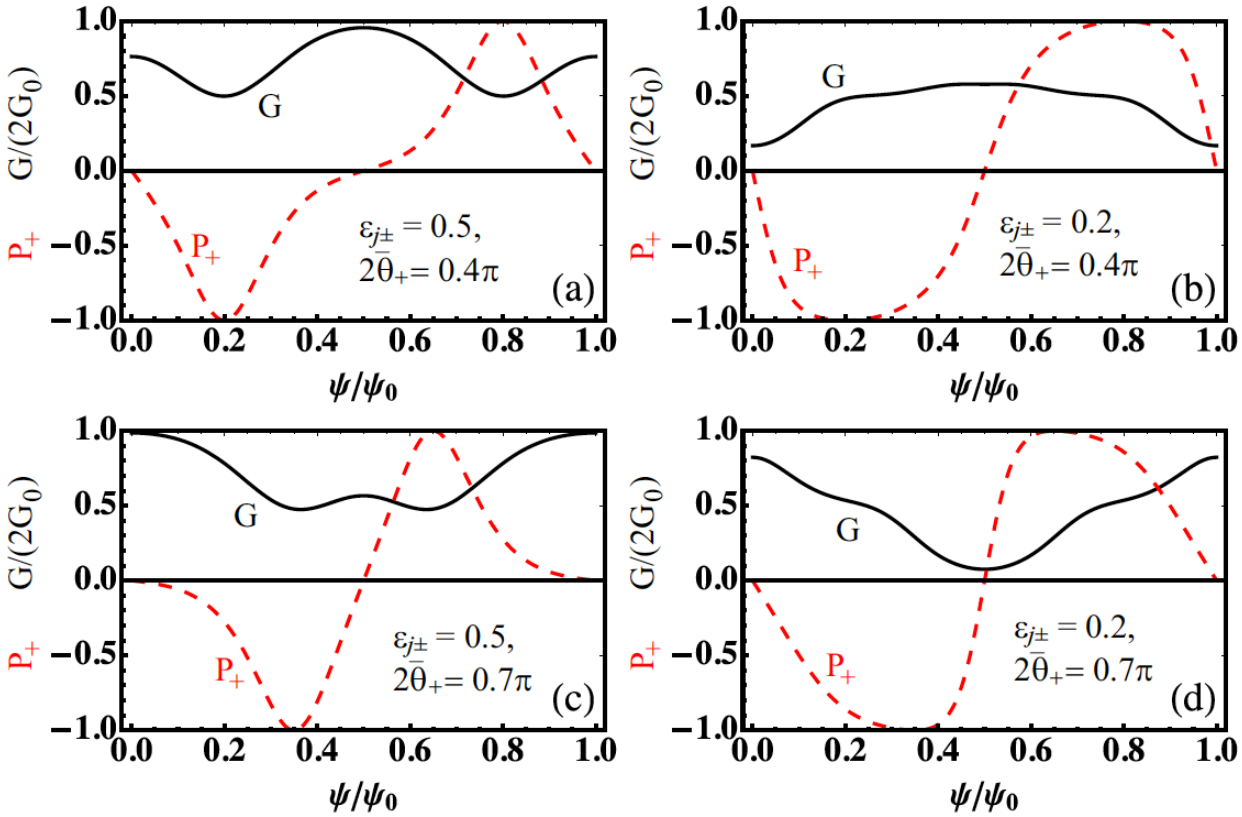
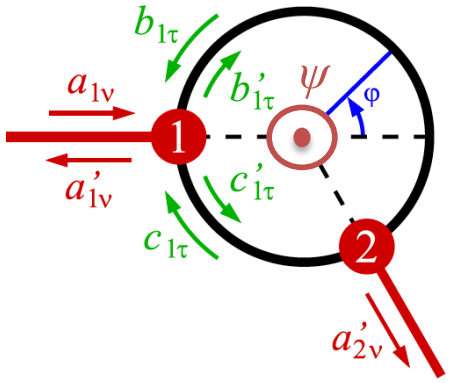
Valley(or spin)-dependent transport

- robust tunable conductance polarization
- engineer based on fully general analytic results!





$$T_\tau(\chi,\tilde{\chi}_\tau,\theta_{\mathrm{AA}}^{(\tau)}) = \frac{4\varepsilon_{1\tau}\varepsilon_{2\tau}\left[\cos^2\chi\,\sin^2\tilde{\chi}_\tau\,\cos^2\left(\theta_{\mathrm{AA}}^{(\tau)}/2\right)+\sin^2\chi\,\cos^2\tilde{\chi}_\tau\,\sin^2\left(\theta_{\mathrm{AA}}^{(\tau)}/2\right)\right]}{\left||\kappa_{1\tau}||\kappa_{2\tau}|e^{i(\varrho_{1\tau}+\varrho_{2\tau})}\cos(2\chi)+|\lambda_{1\tau}||\lambda_{2\tau}|e^{-i(\varrho_{1\tau}+\varrho_{2\tau})}\cos\theta_{\mathrm{AA}}^{(\tau)}-F_\tau(\omega_{1\tau}+\omega_{2\tau},\phi_{1\tau}+\phi_{2\tau},2\tilde{\chi}_\tau)\right|^2}$$

where

$$F_\tau(\omega,\phi,2\tilde{\chi}) = \frac{1}{2}[e^{i\omega}+\sqrt{(1-2\varepsilon_{1\tau})(1-2\varepsilon_{2\tau})}\,e^{-i\phi}]\cos(2\tilde{\chi}) - \frac{i}{2}[e^{i\omega}-\sqrt{(1-2\varepsilon_{1\tau})(1-2\varepsilon_{2\tau})}\,e^{-i\phi}]\sin(2\tilde{\chi}),$$

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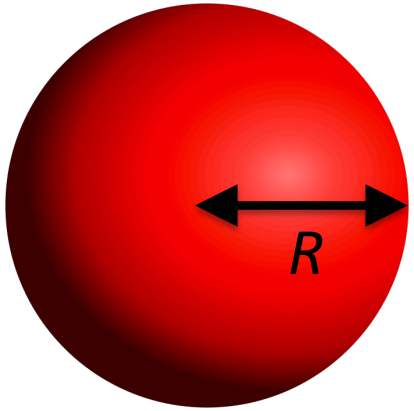
Quasi-0D confinement:

Unconventional optical transitions in topological-insulator nanoparticles

Topological-insulator nanoparticle: Model

- isotropic and particle-hole-symmetric version of 3D-bulk BHZ Hamiltonian + spherical hard-wall mass confinement Imura et al., Phys. Rev. B (2012)
- relevant size scales: nanoparticle radius R, bulk-material Compton length $R_0 = 2\gamma / \Delta_0$
- previous work considered limit $R \gg R_0$



$$\mathcal{H} = \begin{pmatrix} \frac{\Delta(\mathbf{k})}{2} + V(r) & \gamma\, k_z & 0 & \gamma\, k_- \\ \gamma\, k_z & -\frac{\Delta(\mathbf{k})}{2} - V(r) & \gamma\, k_- & 0 \\ 0 & \gamma\, k_+ & \frac{\Delta(\mathbf{k})}{2} + V(r) & -\gamma\, k_z \\ \gamma\, k_+ & 0 & -\gamma\, k_z & -\frac{\Delta(\mathbf{k})}{2} - V(r) \end{pmatrix}$$

General form of TI-nanoparticle states

Gioia, Christie, UZ et al., arXiv:1906.08162

- spherical symmetry: total angular momentum j and its projection m are good quantum numbers
- ramifications of two-flavour Dirac physics
  - half-integer j (spin-1/2 spherical harmonics!)
  - two states with opposite parity exist for fixed j, m
  - intricate structure of angular and radial wave functions

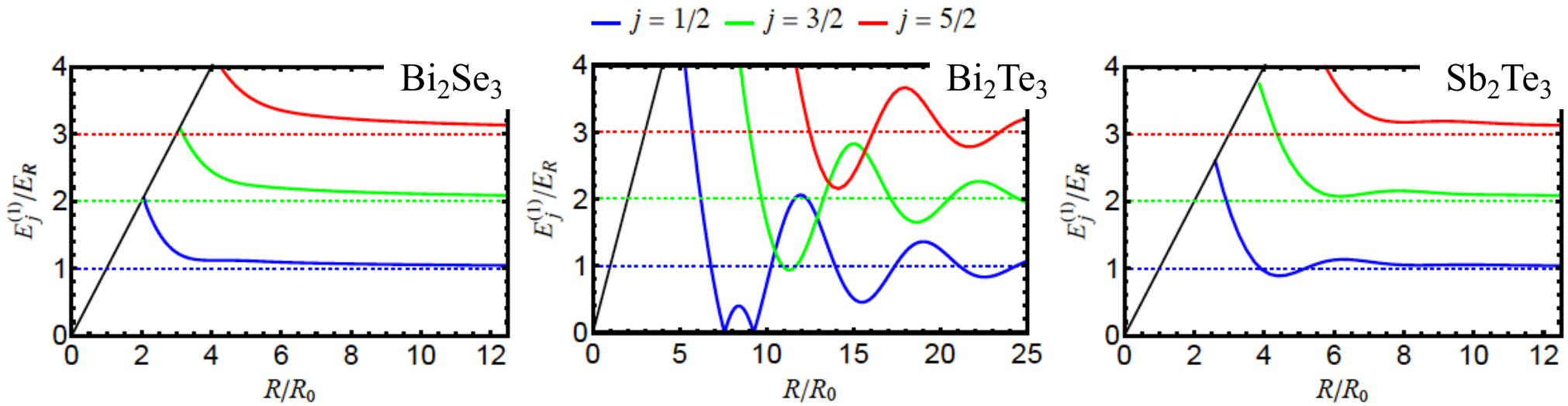
$$\Psi^{(n)}_{jm+}(\mathbf{r}) = \frac{C^{(n)}_{j+}}{2}\begin{pmatrix} \sqrt{\frac{j+m}{j}}\, Y^{m-\frac{1}{2}}_{j-\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j+\uparrow}(r) \\ \sqrt{\frac{j+1-m}{j+1}}\, Y^{m-\frac{1}{2}}_{j+\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j-\uparrow}(r) \\ \sqrt{\frac{j-m}{j}}\, Y^{m+\frac{1}{2}}_{j-\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j+\uparrow}(r) \\ -\sqrt{\frac{j+1+m}{j+1}}\, Y^{m+\frac{1}{2}}_{j+\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j-\uparrow}(r) \end{pmatrix}, \quad \Psi^{(n)}_{jm-}(\mathbf{r}) = \frac{C^{(n)}_{j-}}{2}\begin{pmatrix} \sqrt{\frac{j+1-m}{j+1}}\, Y^{m-\frac{1}{2}}_{j+\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j+\uparrow}(r) \\ \sqrt{\frac{j+m}{j}}\, Y^{m-\frac{1}{2}}_{j-\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j-\uparrow}(r) \\ -\sqrt{\frac{j+1+m}{j+1}}\, Y^{m+\frac{1}{2}}_{j+\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j+\uparrow}(r) \\ \sqrt{\frac{j-m}{j}}\, Y^{m+\frac{1}{2}}_{j-\frac{1}{2}}(\theta,\varphi)\, \phi^{(n)}_{j-\uparrow}(r) \end{pmatrix}$$

Topological-bound-state spectrum

- large-R limit: $E_j^{(1)} \to \left(j + \frac{1}{2}\right) E_R$ with $E_R = \frac{\gamma}{R}$

Imura et al., Phys. Rev. B (2012)

- significant deviations from asymptotic behaviour occur even for not-too-small nanoparticle size
- critical size R_c: no sub-gap state exists for $R < R_c$



Optical transitions: Basic theory

- selection rules and transition rates are governed by the optical-dipole matrix elements

$$\mathbf{d}^{n'j'm'\kappa'}_{n\,j\,m\,\kappa} \equiv \int d^3r\, \left[\Psi^{(n')}_{j'm'\kappa'}(\mathbf{r})\right]^\dagger \mathbf{d}\, \Psi^{(n)}_{jm\kappa}(\mathbf{r})$$

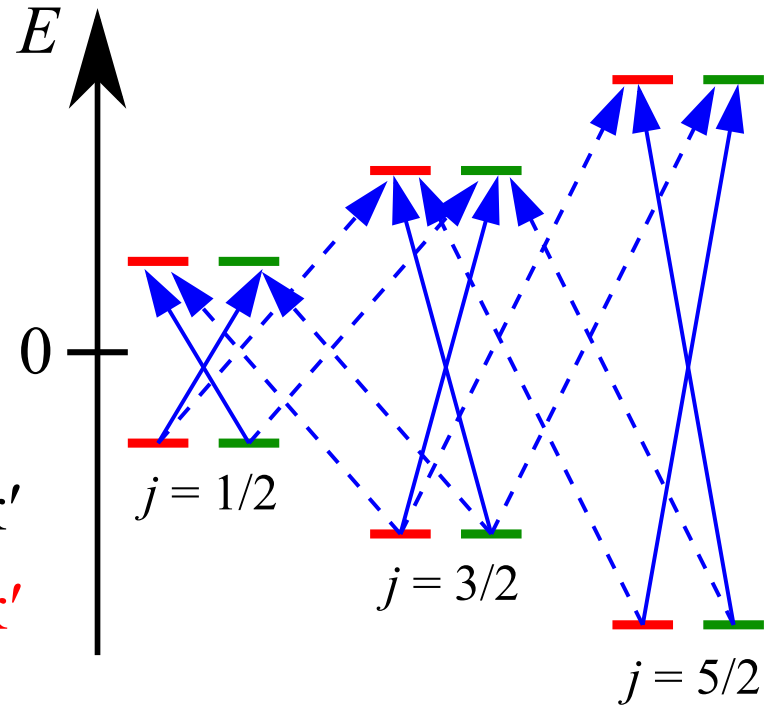
- both intra-band and inter-band transitions possible
  - treat both on the same footing by deriving the general envelope-function-space optical-dipole operator

$$\mathbf{d} = e\,\mathbf{r}\,\tau_0 \otimes \sigma_0 + \frac{e\,R_0}{2}\,\tau_y \otimes \boldsymbol{\sigma}$$

- obtain analytical expressions for matrix elements

Gioia, Christie, UZ, Governale, Sneyd, arXiv:1906.08162

Unconventional optical-dipole transitions



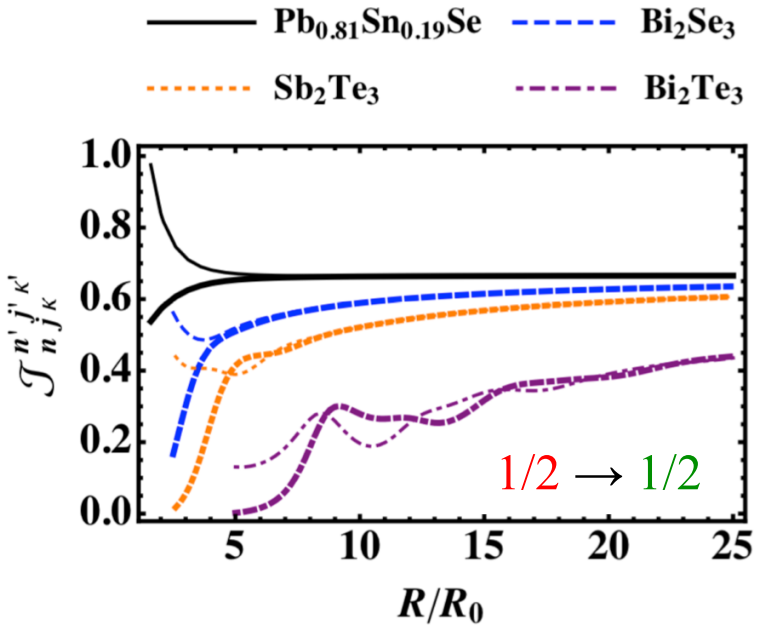
$\Delta m = 0, \pm 1$

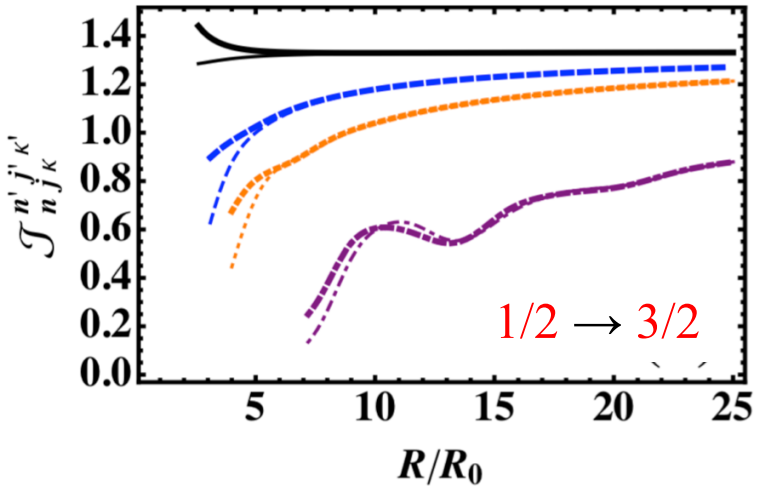
$\Delta j = \pm 1, \kappa = \kappa'$

$\Delta j = 0, \kappa = -\kappa'$

$$\sum_{m=-j}^{j} \left| (d_x \pm i d_y)^{n'j'm'\kappa'}_{n\,j\,m\,\kappa} \right|^2 \equiv \frac{2e^2R^2}{3} \mathcal{T}^{n'j'\kappa'}_{n\,j\,\kappa}$$

$$\sum_{m=-j}^{j} \left| (d_z)^{n'j'm'\kappa'}_{n\,j\,m\,\kappa} \right|^2 \equiv \frac{e^2R^2}{3} \mathcal{T}^{n'j'\kappa'}_{n\,j\,\kappa}$$





Conclusions

- used effective (BHZ) model of bulk topological-insulator bands to study size- quantization effects
  - (parabolic, hard-wall) mass-confinement potential
- quantum wells: materials dependence of gap oscillations, giant Zeeman splitting for in-plane **B**

  Kotulla, UZ, New. J. Phys. **19**, 073025 (2017); Kotulla, PhD Thesis (2019)
- quantum rings: confinement-dependent geometric phase, valley/spin-polarized electric conductance

  Gioia, UZ, Governale, Winkler, Phys. Rev. B **97**, 205421 (2018)
- nanoparticles: unconventional optical transitions

  Gioia, Christie, UZ, Governale, Sneyd, arXiv:1906.08162