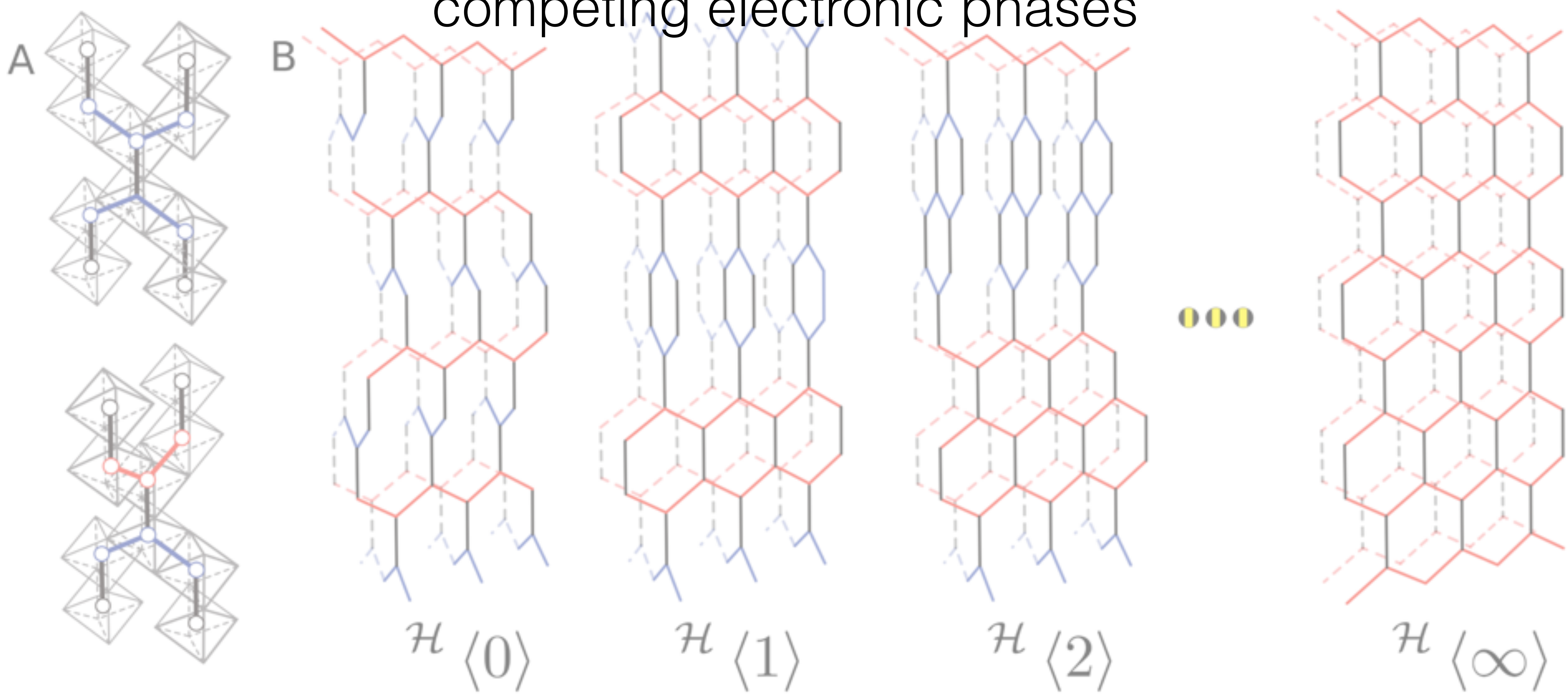
From synthesis to thermodynamics

The interplay of competing crystalline phases and competing electronic phases



Quantum oscillations in Weyl semi-metals

Quantum oscillatory phenomena from Fermi Arcs

Philip J. M. W. Moll, Nityan Nair, James G. Analytis
University of California, Berkeley

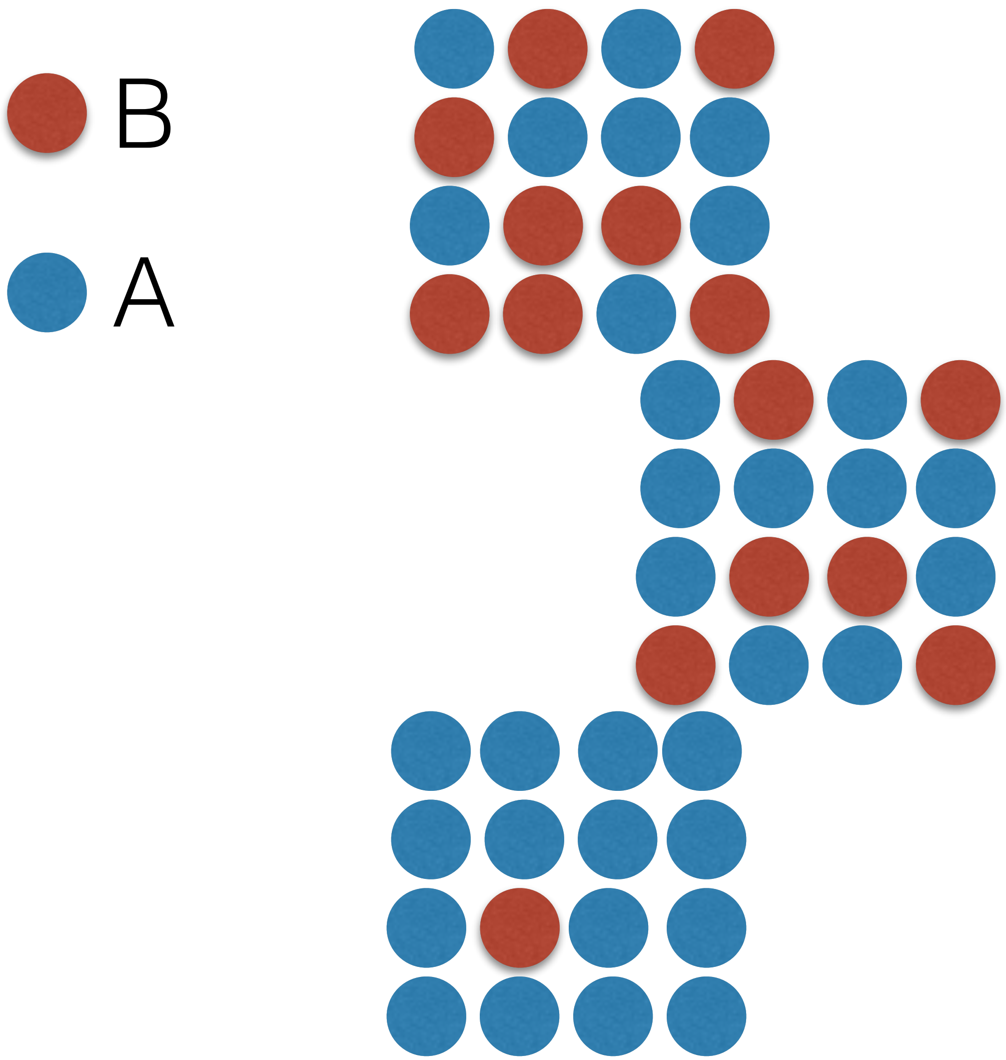
GORDON AND BETTY MOORE FOUNDATION

THE UNIVERSITY OF CALIFORNIA BERKELEY · LET THERE BE LIGHT · 1868

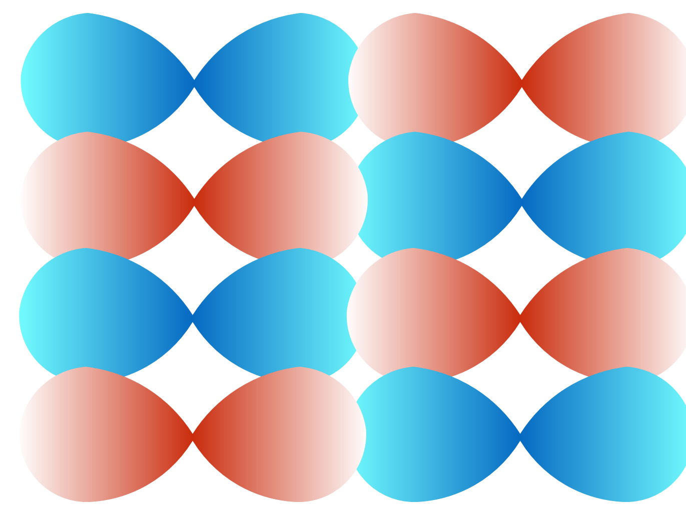
Synthesis

- What techniques do we use to make these materials?
- Why are some materials difficult to make and other easy?

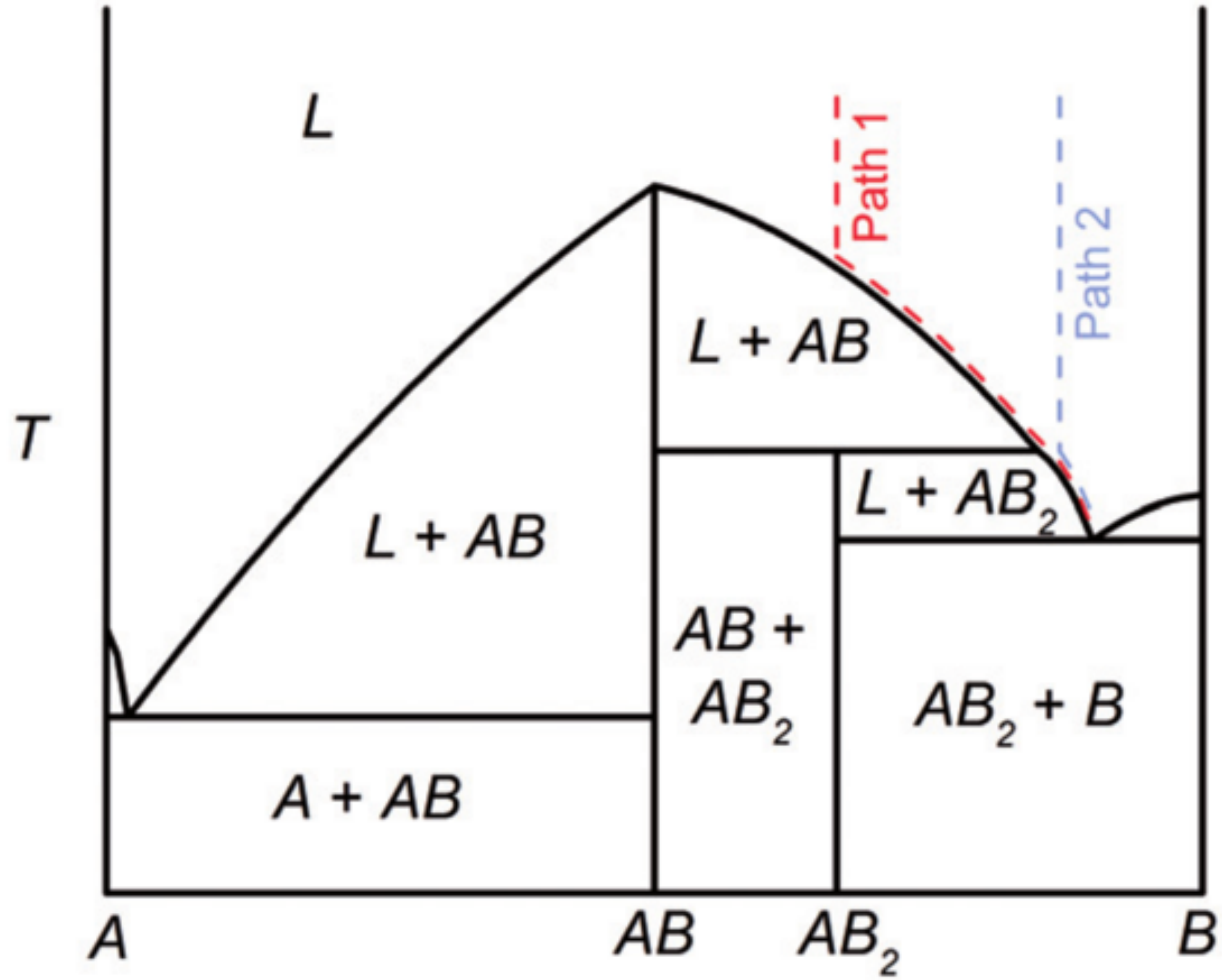
Making compounds



vs



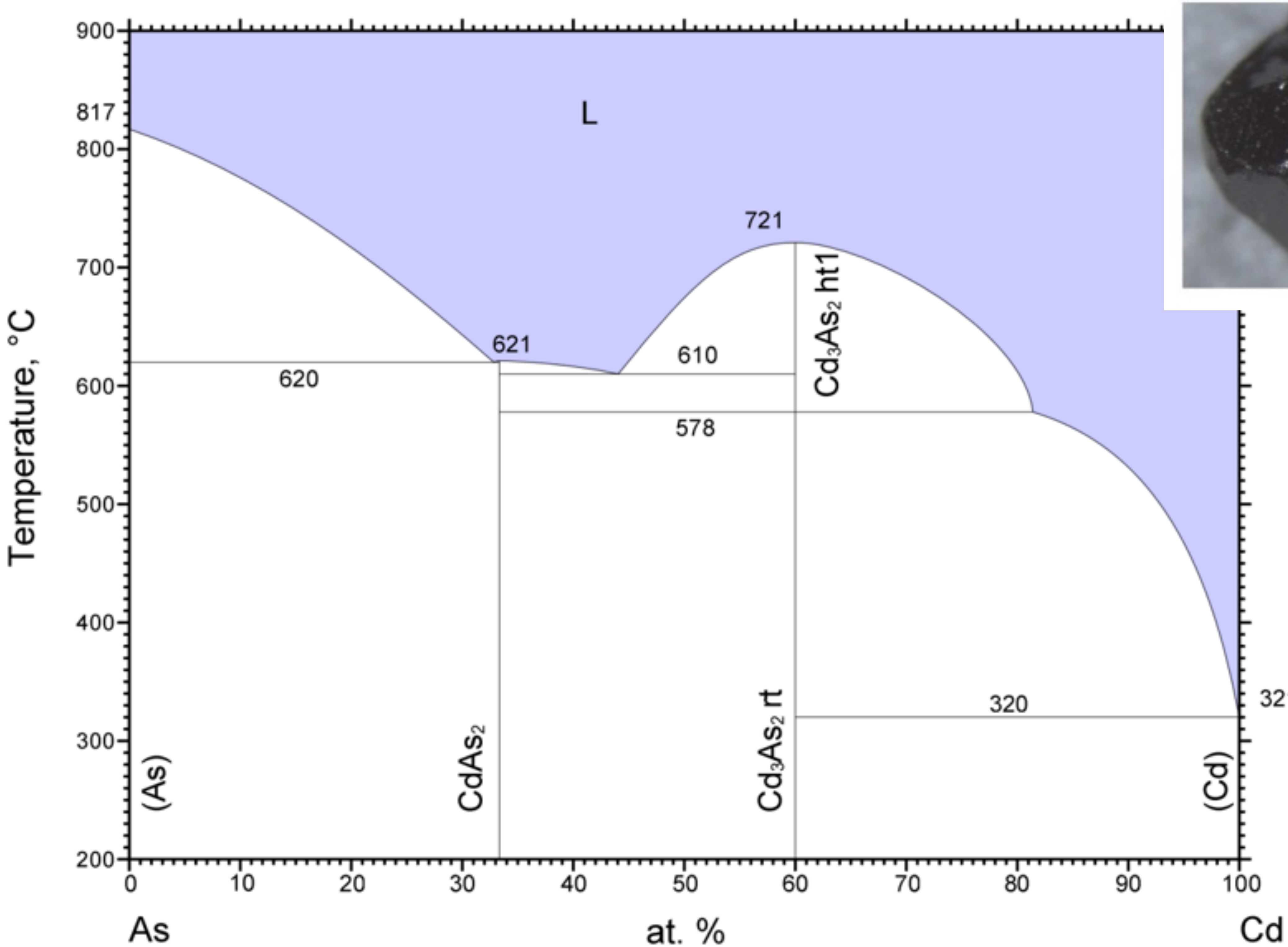
- The compounds that one can make are generally determined by where the minimum in the free energy occurs, which in turn depends on a balance of the entropy (generally the change in configurational entropy) and the enthalpy (mostly the gain in energy in bonding).

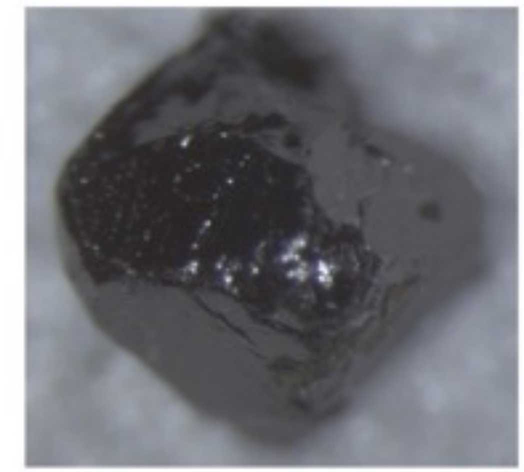


Binary phase diagrams

Fisher/Shapiro/Analytis Philosophical Magazine 2012

Phase diagram of Cd_3As_2





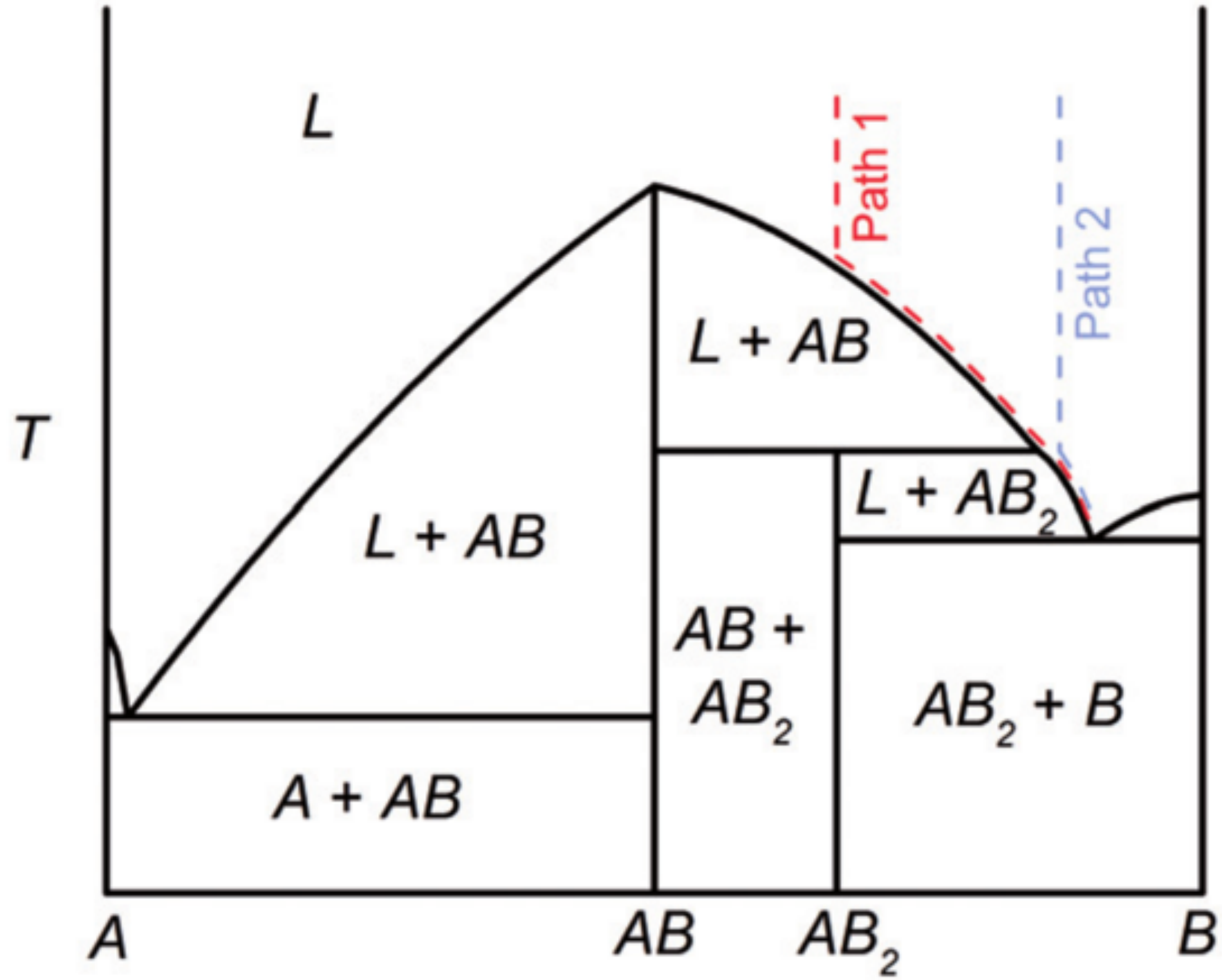
Congruent and incongruent melts

- In many binary phase diagrams more than one solid phase can be made.

- Intermediate phases can be classed as either

  - *congruently melting* (melt from a homogeneous solid to a homogeneous liquid)

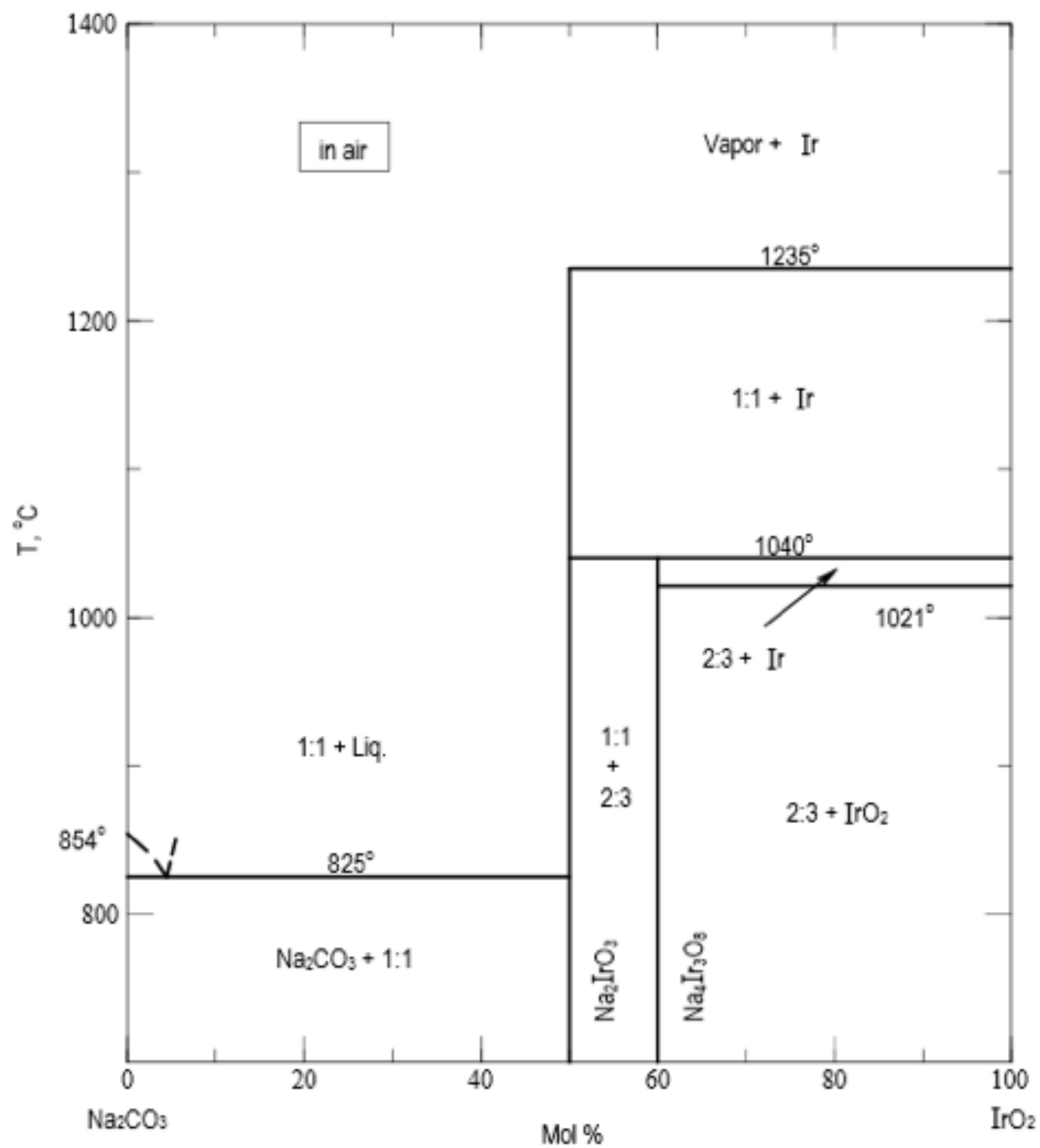
  - *incongruently melting* (the solid decomposes on heating to a two-phase mixture of solid and liquid where multiple solid phases may be in equilibrium)

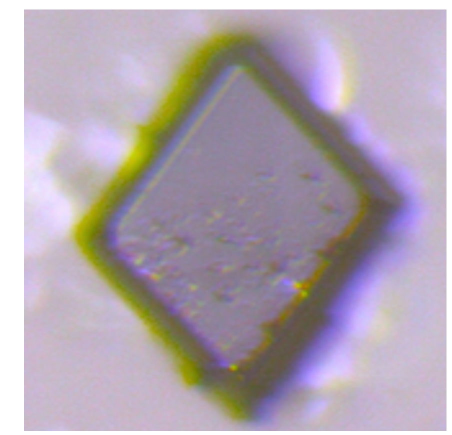


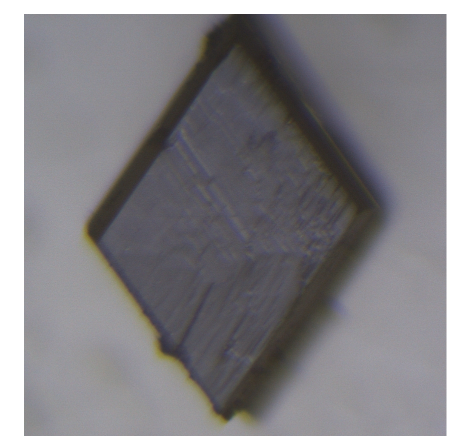
Binary phase diagrams

Fisher/Shapiro/Analytis Philosophical Magazine 2012

Phase diagram of Na_2CO_3-IrO_2







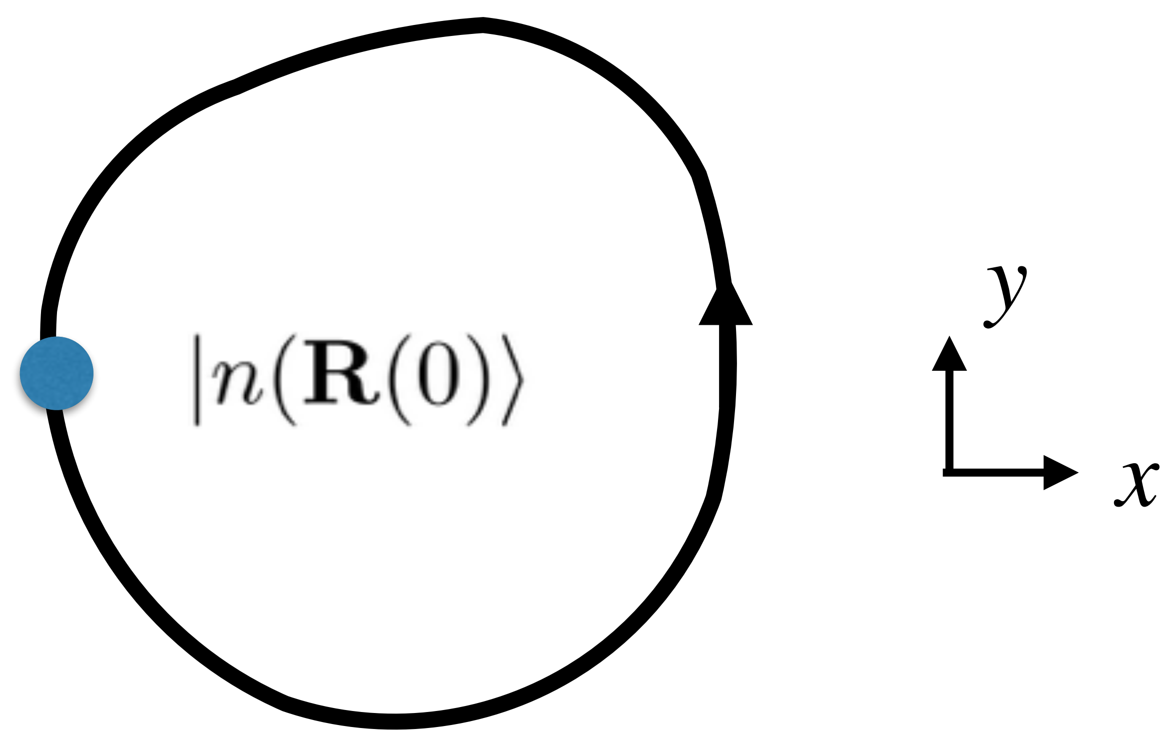
See Ruiz (Sunday afternoon)

PARTII:

Adiabaticity, Quantum Oscillations and the Berry's phase.

Adiabatic theorem

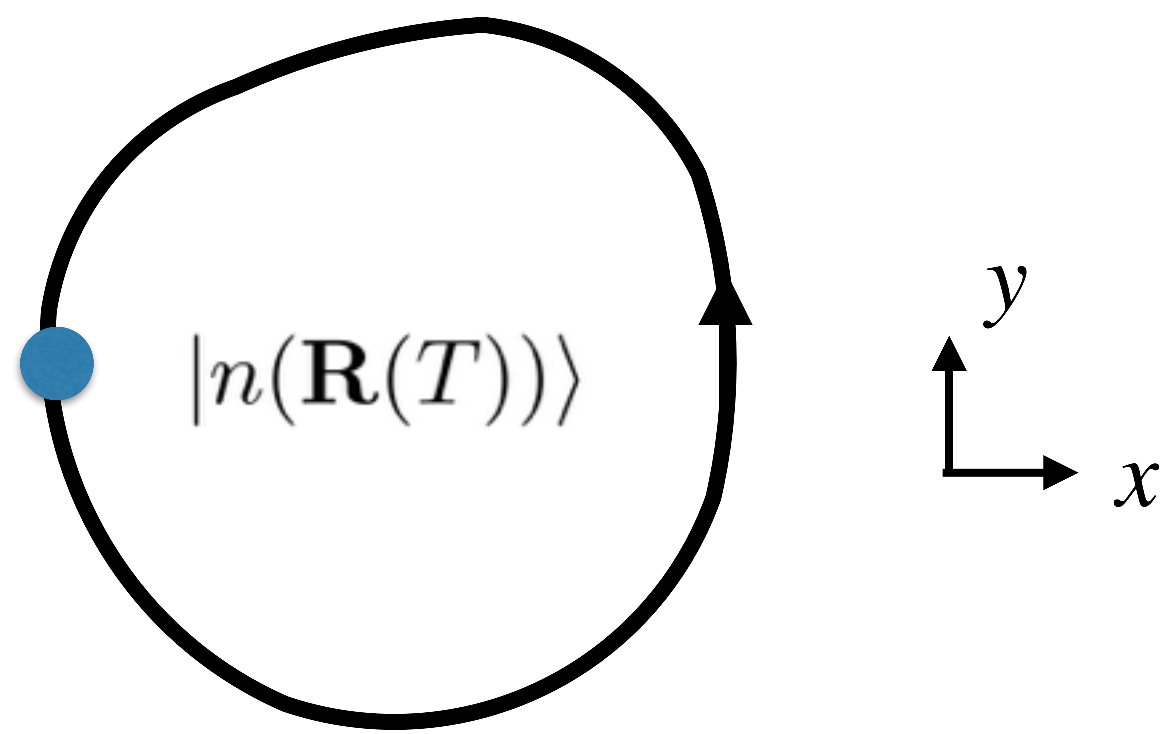
$$|n(\mathbf{R}(0)\rangle \to e^{i\gamma_B} e^{i\phi} |n(\mathbf{R}(t))\rangle$$



$$\phi = -\frac{1}{\hbar}\int_0^T E_n(\mathbf{R}(t))dt \qquad \gamma_B(C) = i\oint_C \langle n(\mathbf{R})|\nabla_R|n(\mathbf{R})\rangle$$

Adiabatic theorem

$$|n(\mathbf{R}(0)\rangle \rightarrow e^{i\gamma_B} e^{i\phi} |n(\mathbf{R}(t))\rangle$$

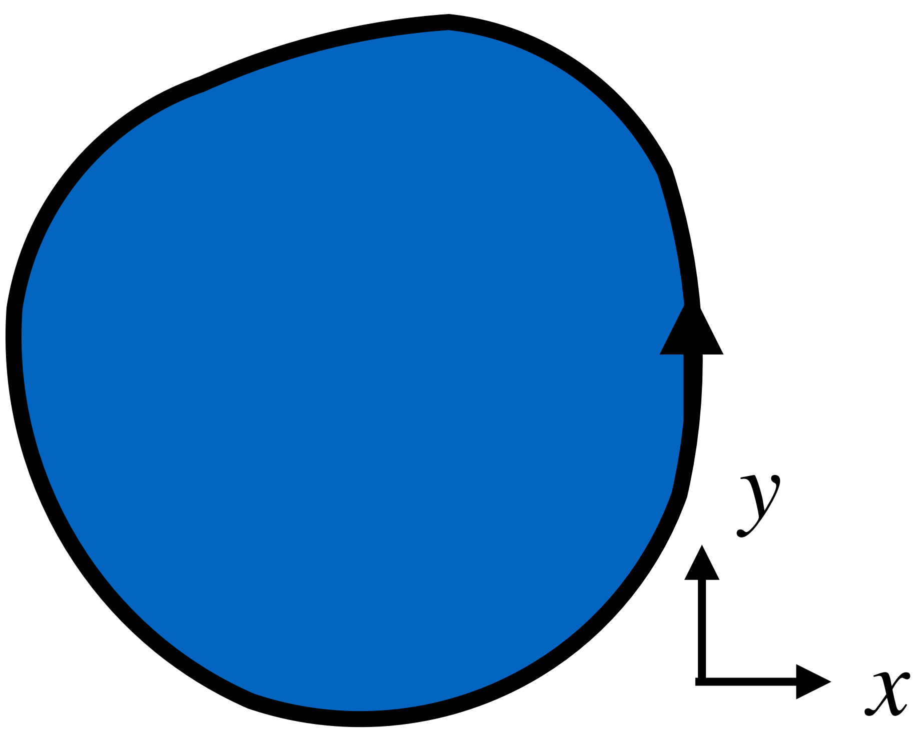


$$\phi = -\frac{1}{\hbar}\int_0^T E_n(\mathbf{R}(t))dt \qquad \gamma_B(C) = i\oint_C \langle n(\mathbf{R})|\nabla_R|n(\mathbf{R})\rangle$$

Onsager's argument

$$\oint_C \mathbf{p} \cdot d\mathbf{q} = (n + \gamma) 2\pi\hbar$$

Adiabatic invariant

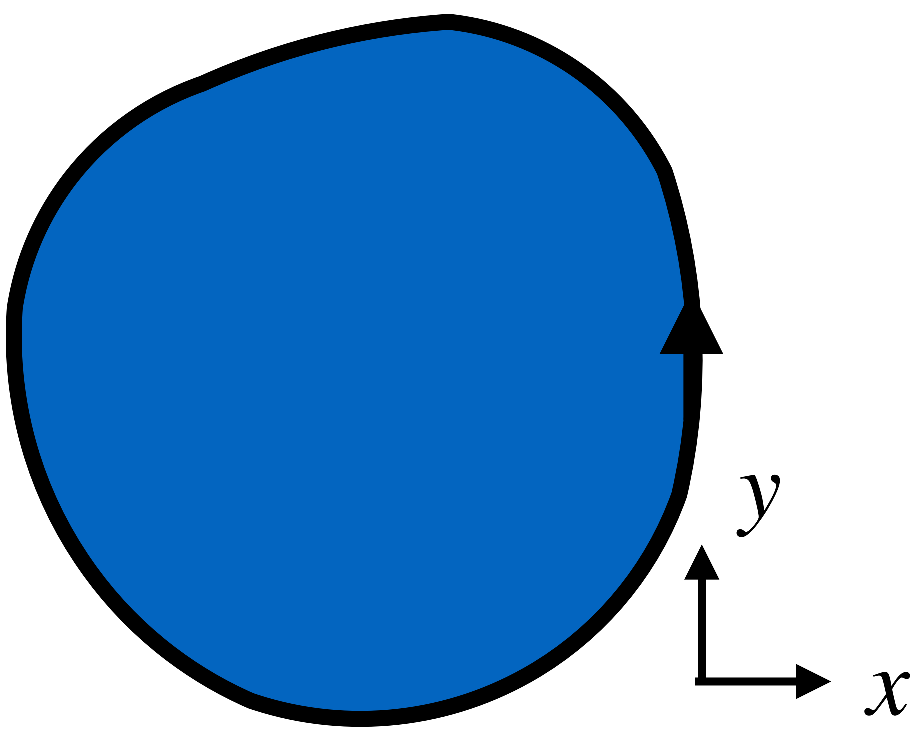


Onsager's argument

$$\oint_C \mathbf{p} \cdot d\mathbf{q} = (n + \gamma) 2\pi\hbar$$

Adiabatic invariant

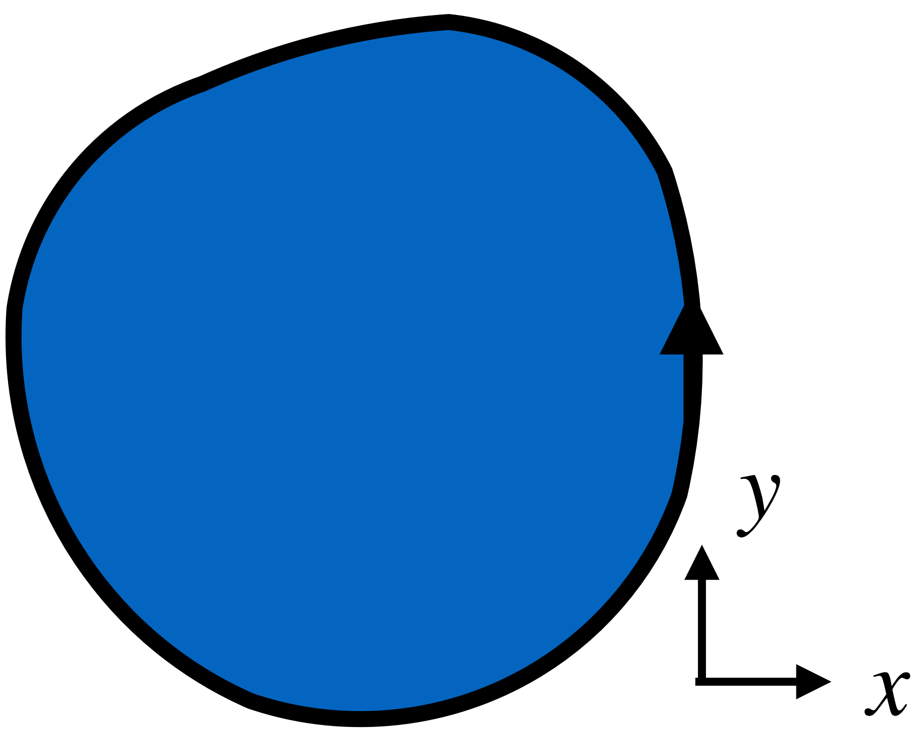
$$\mathbf{p} = \hbar\mathbf{k} - e\mathbf{A}$$



Onsager's argument

$$\oint_C \mathbf{p} \cdot d\mathbf{q} = (n + \gamma) 2\pi\hbar$$

Adiabatic invariant



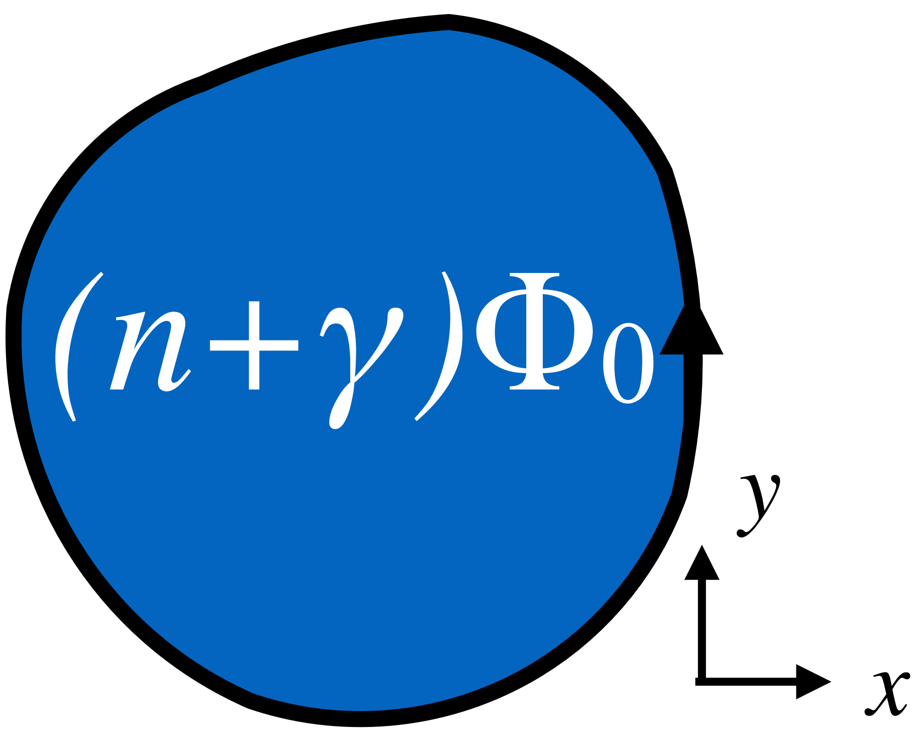
$$\mathbf{p} = \hbar\mathbf{k} - e\mathbf{A}$$

$$\Phi \equiv BA = (n + \gamma) 2\pi \frac{\hbar}{e}$$

Onsager's argument

$$\oint_C \mathbf{p} \cdot d\mathbf{q} = (n+\gamma)2\pi\hbar$$

Adiabatic invariant

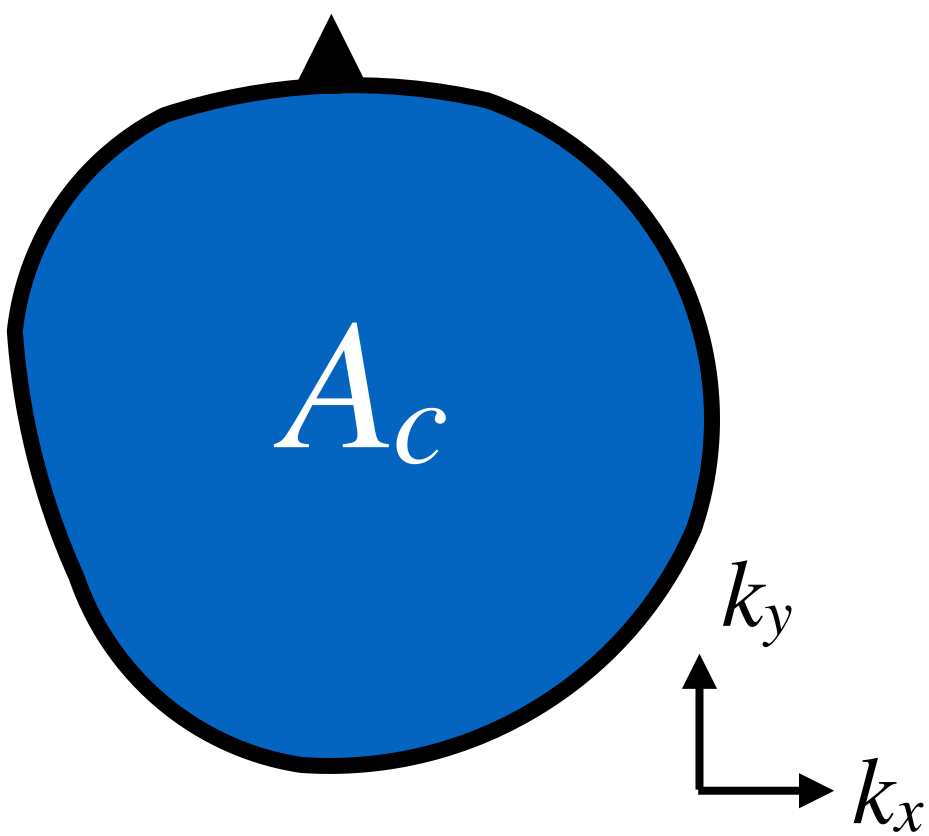


$$\mathbf{p} = \hbar\mathbf{k} - e\mathbf{A}$$

$$\Phi \equiv BA = (n+\gamma)2\pi\frac{\hbar}{e}$$
$$= (n+\gamma)\Phi_0$$

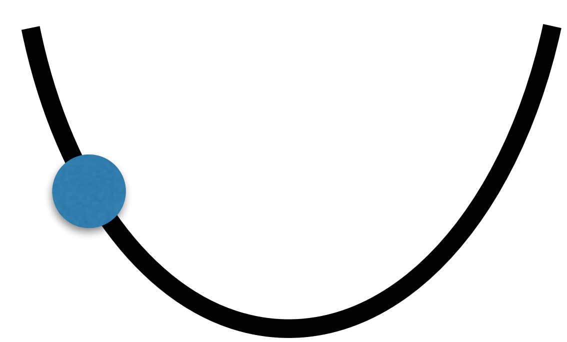
From flux quantization to energy quantization

$$A_c = (n+\gamma)4\pi^2 B/\Phi_0$$



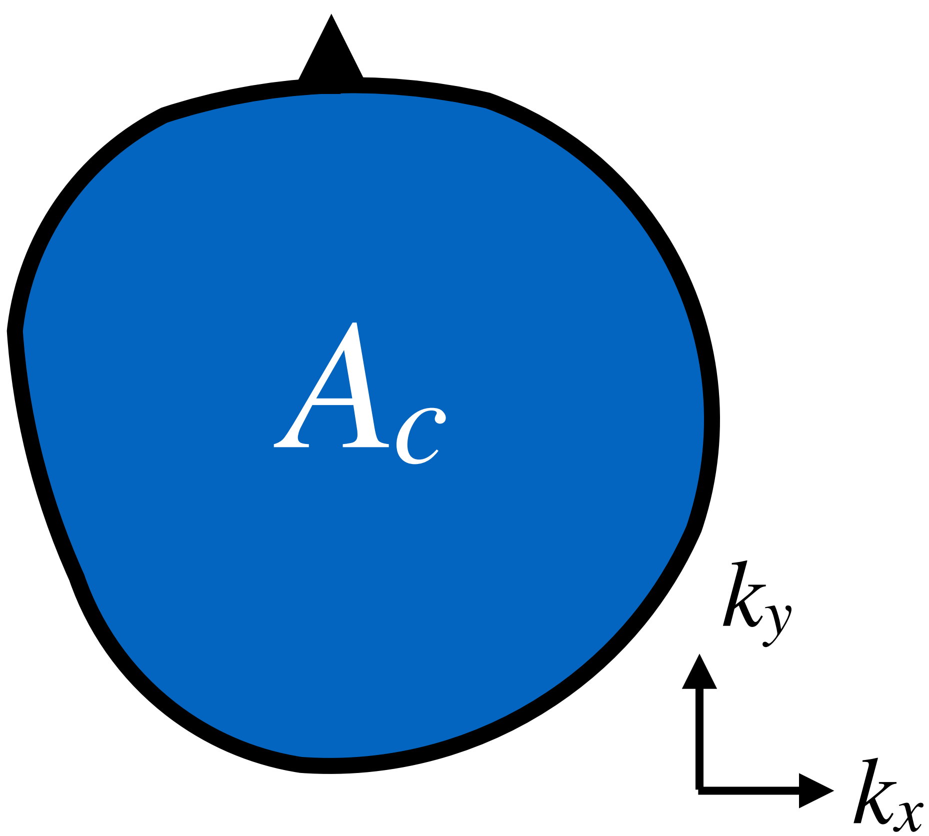
$$\omega_c = \frac{eB}{m^*}$$

$$\varepsilon_c = (n+\gamma)\hbar\omega_c$$



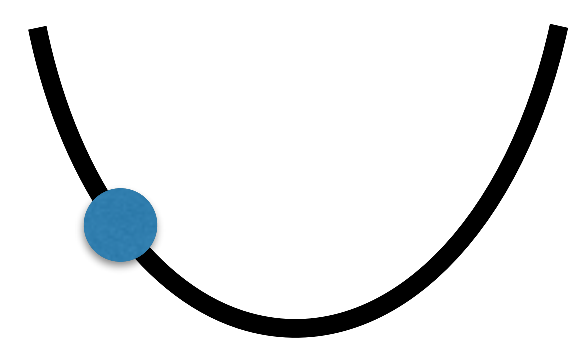
From flux quantization to energy quantization

$$A_c = (n+\gamma)4\pi^2 B/\Phi_0$$



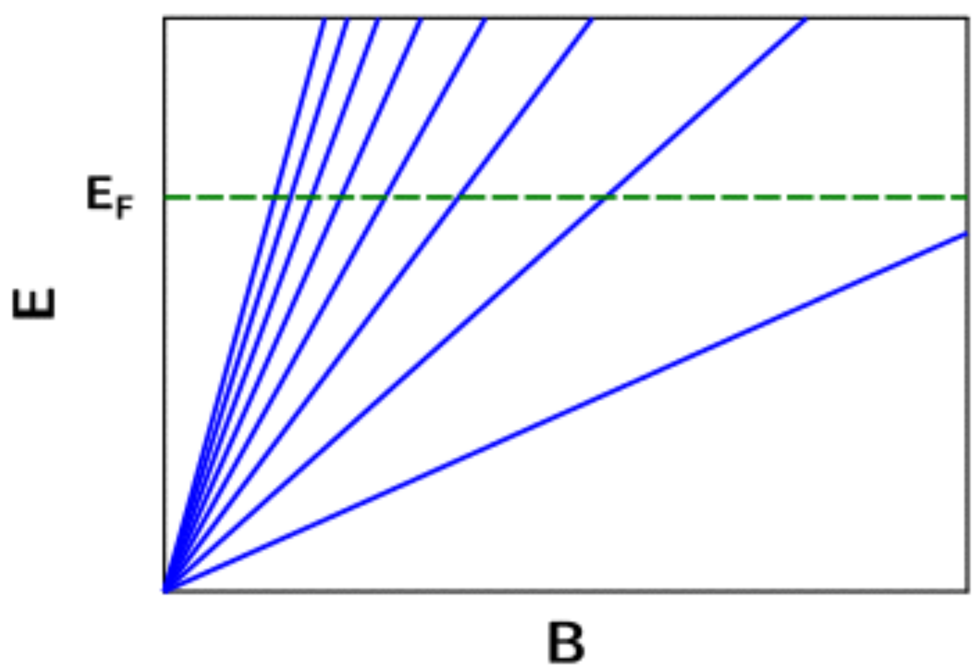
$$\omega_c = \frac{eB}{m^*}$$

$$\varepsilon_c = (n+\gamma)\hbar\omega_c$$



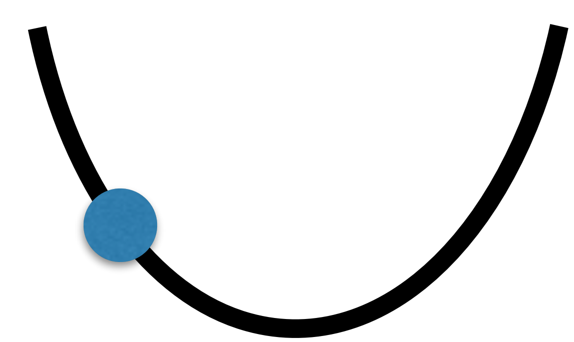
From flux quantization to energy quantization

$$A_c = (n+\gamma)4\pi^2 B/\Phi_0$$



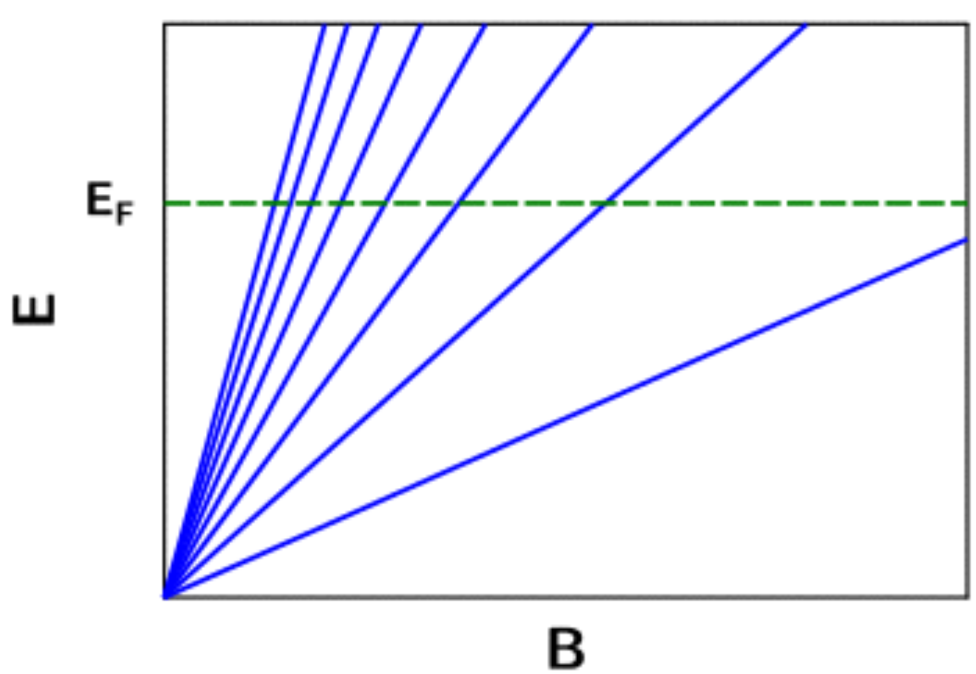
$$\omega_c = \frac{eB}{m^*}$$

$$\varepsilon_c = (n+\gamma)\hbar\omega_c$$



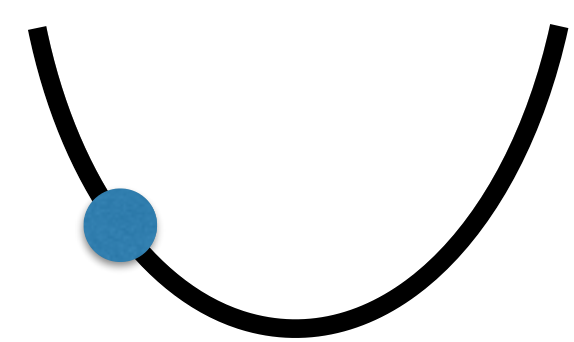
From flux quantization to energy quantization

$$A_c = (n + \gamma) 4\pi^2 B / \Phi_0$$

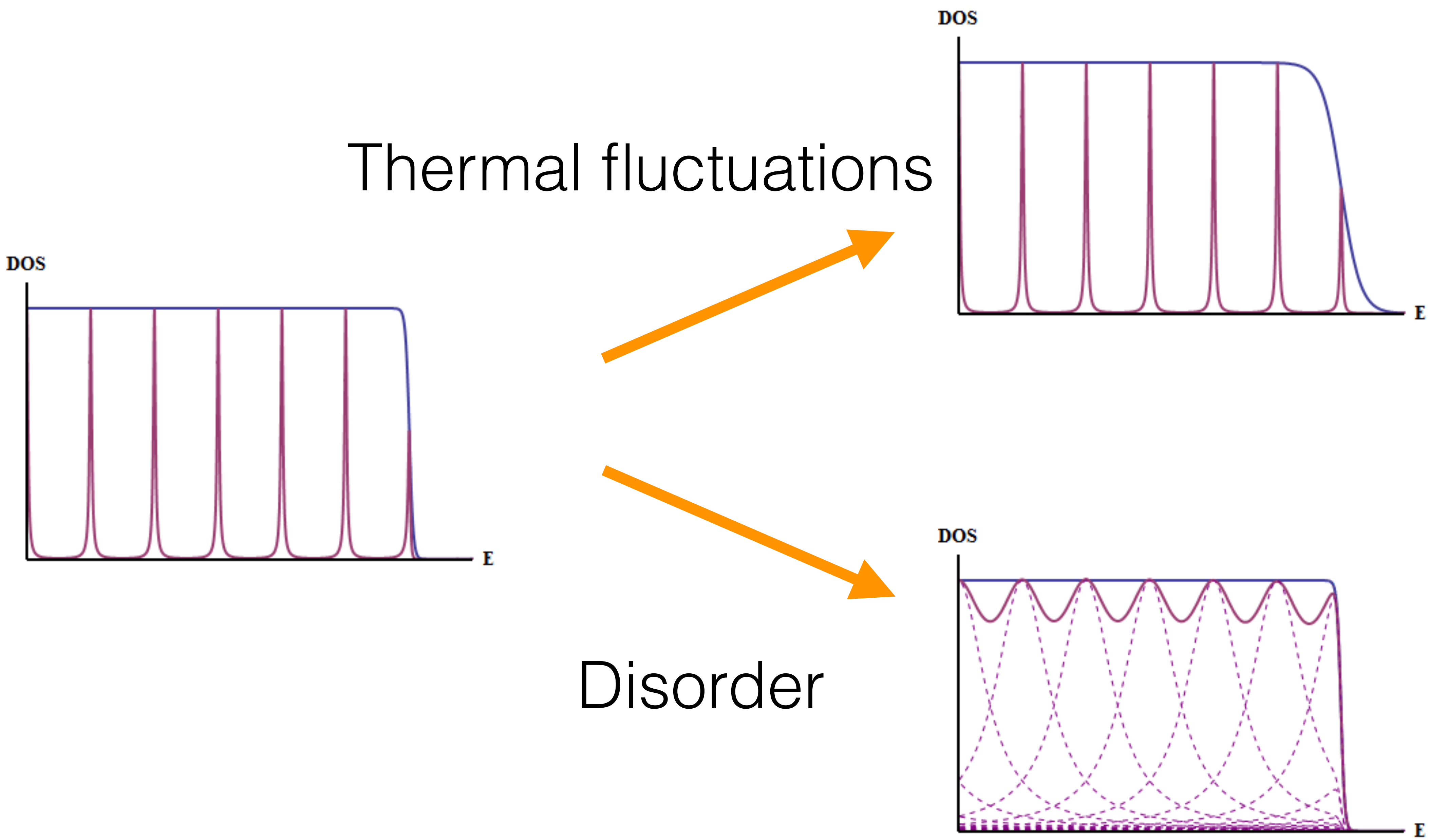


$$\omega_c = \frac{eB}{m^*}$$

$$\varepsilon_c t_c = (n + \gamma) 2\pi\hbar$$



Disorder and thermal fluctuations



Quantum oscillations are a litmus test for metal purity

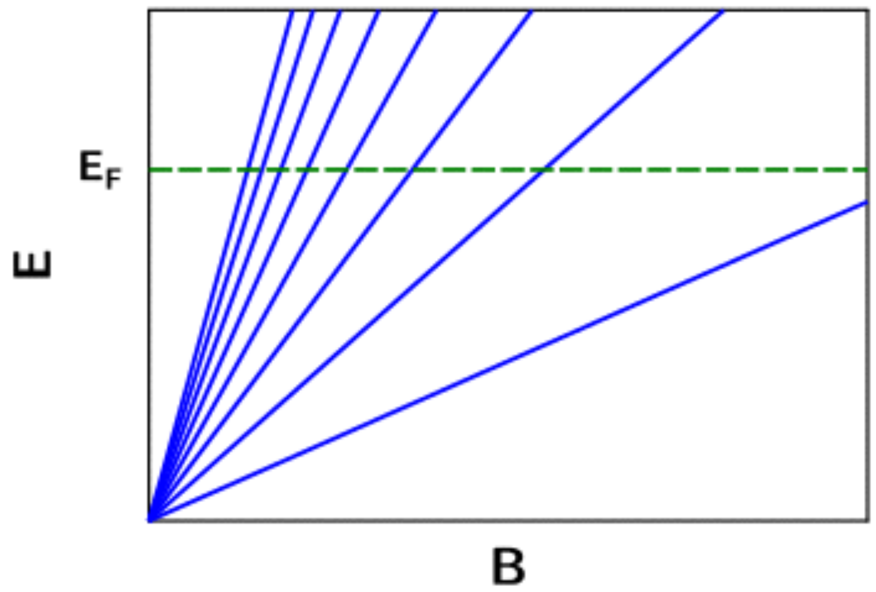
- Consider a really clean metal with a Fermi velocity of 1×10^6m/s and a mean free path 100nm.
- At 10T, the quantum oscillations are suppressed to **1:1000.**

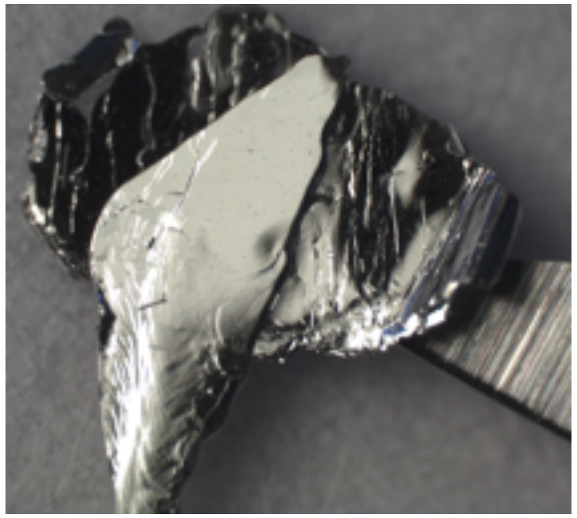
Quantum oscillations are a litmus test for metal purity

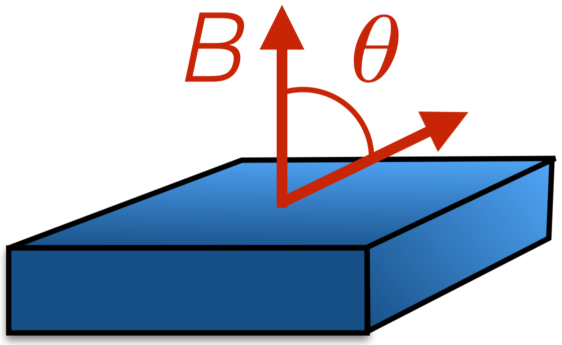
- Consider a really clean metal with a Fermi velocity of 1×10^6 m/s and a mean free path 100nm.
- At 10T, the quantum oscillations are suppressed to **1:1000.**

- Now consider a pretty good metal with a Fermi velocity of 1×10^6 m/s and a mean free path 10nm.
- At 10T, the quantum oscillations are suppressed to **1:10^{27}!**

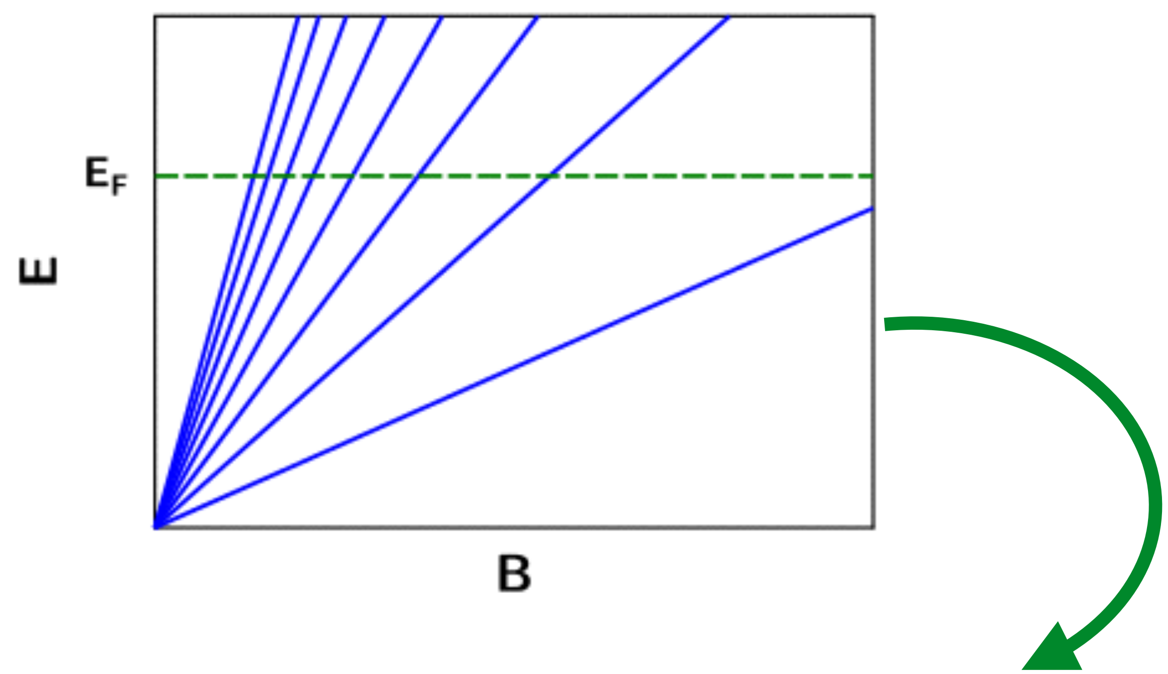
Bulk Quantum oscillations in Bi_2Se_3

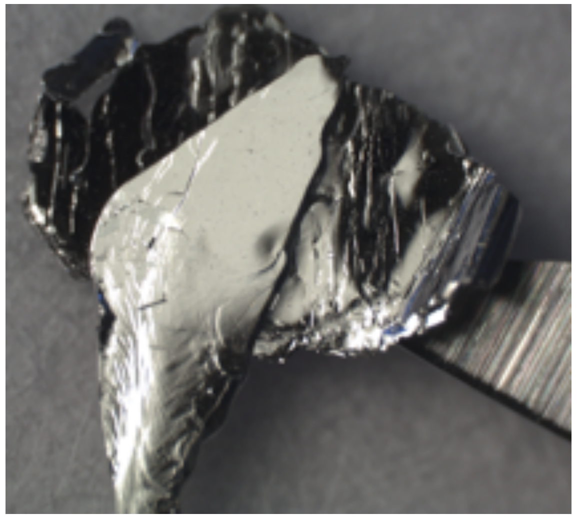


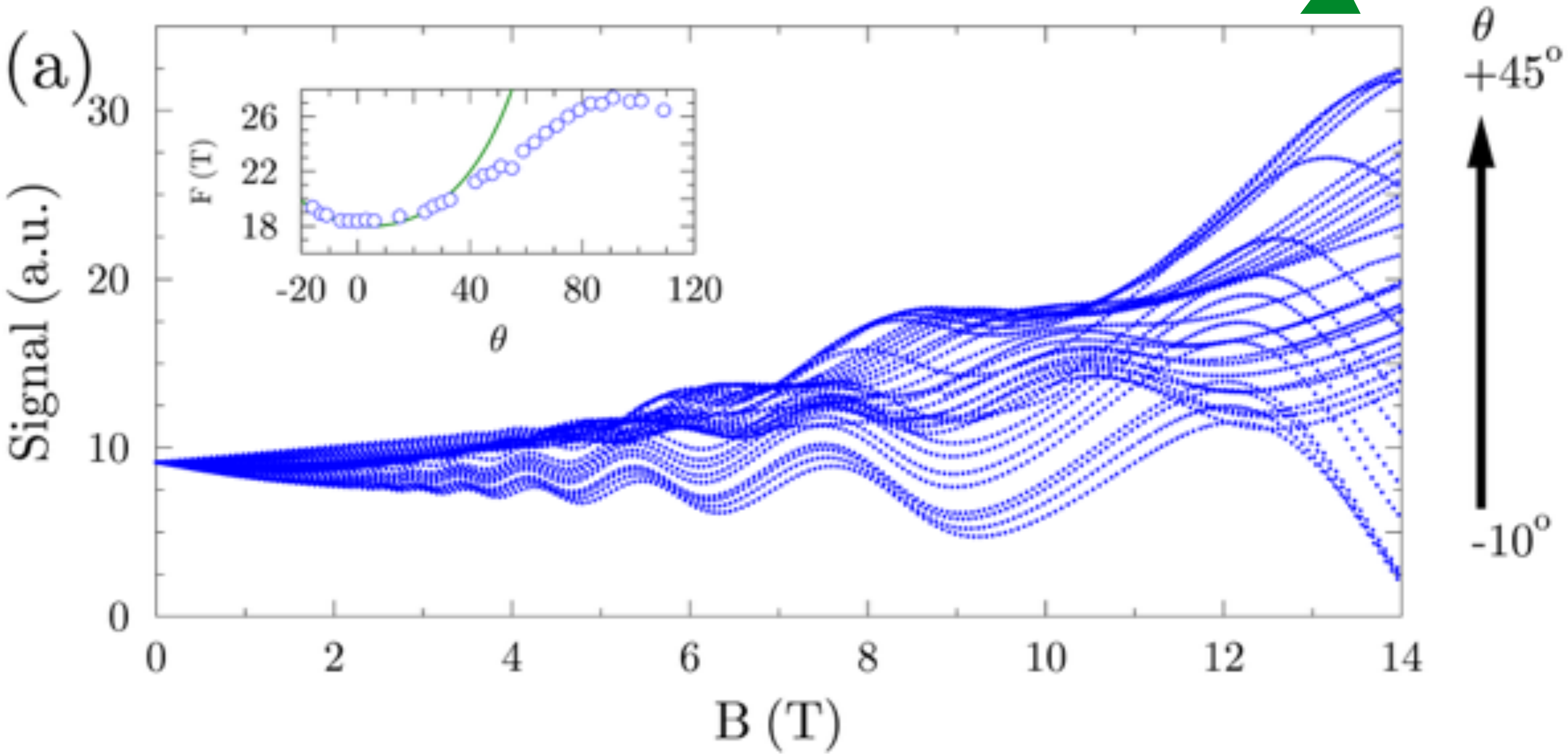


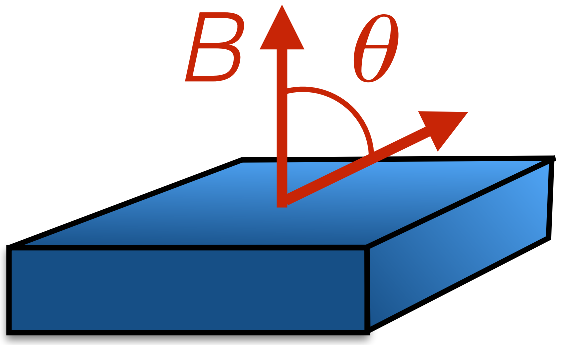


Bulk Quantum oscillations in Bi_2Se_3

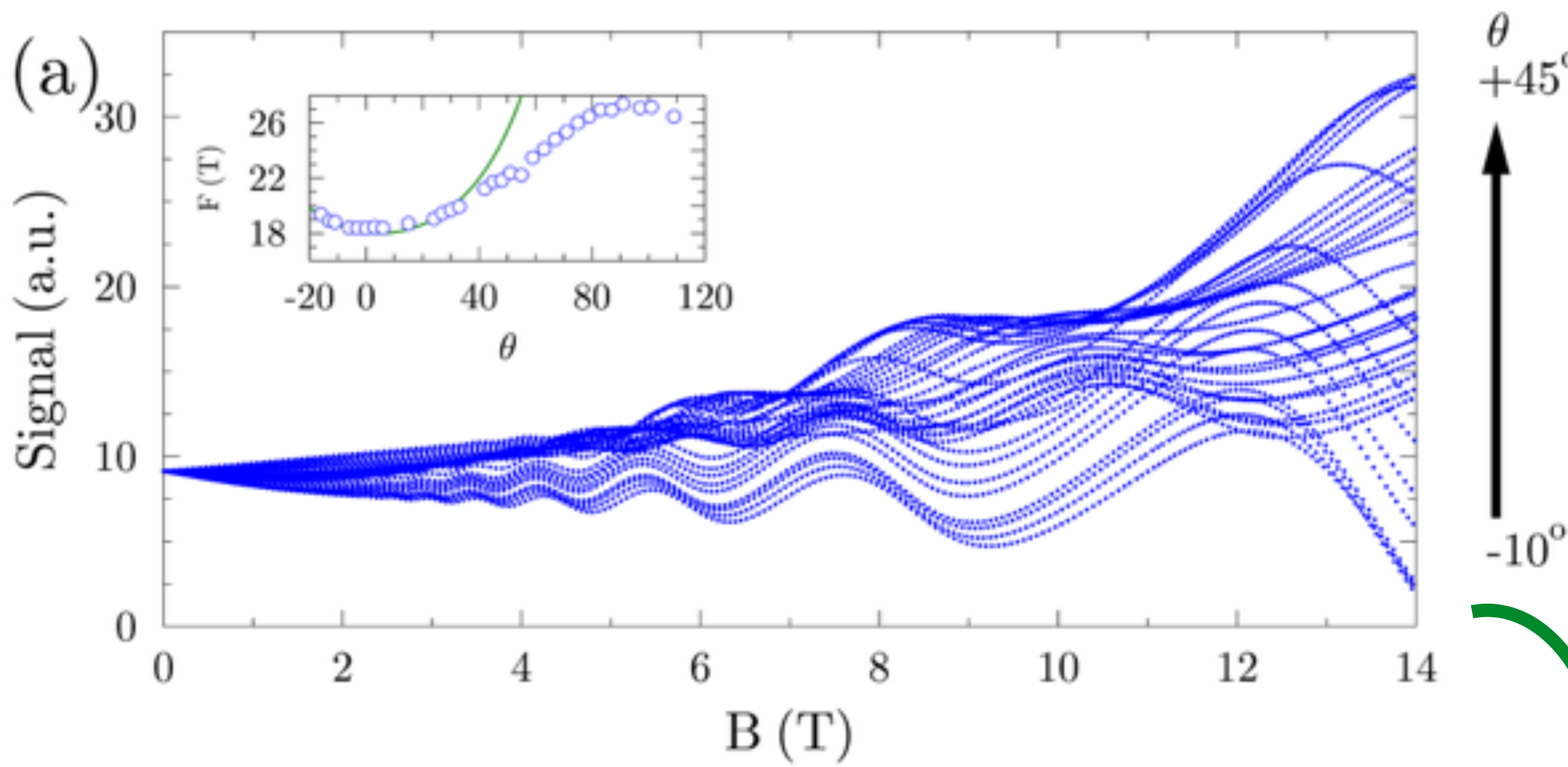


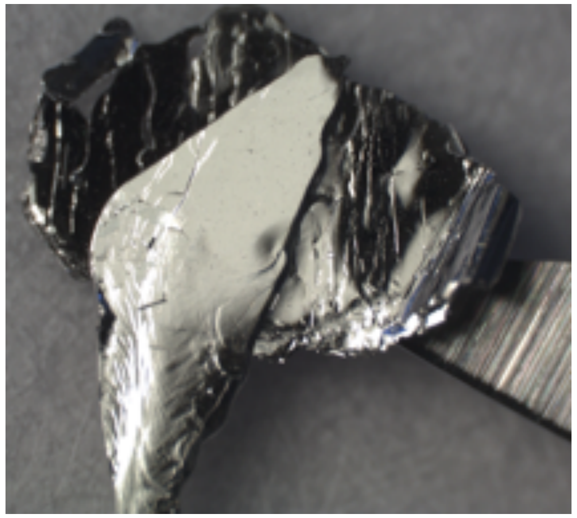


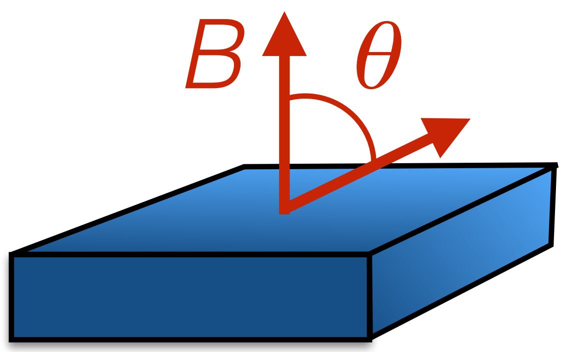


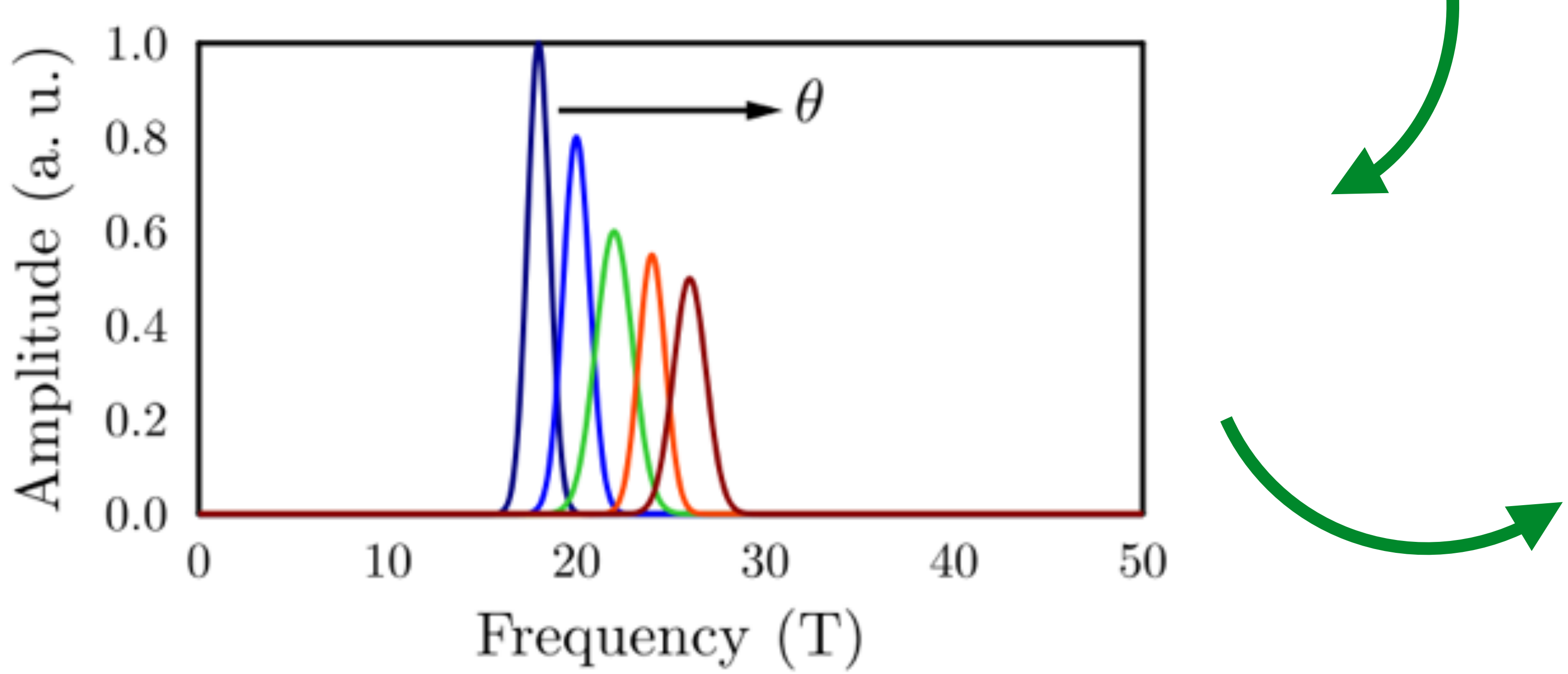


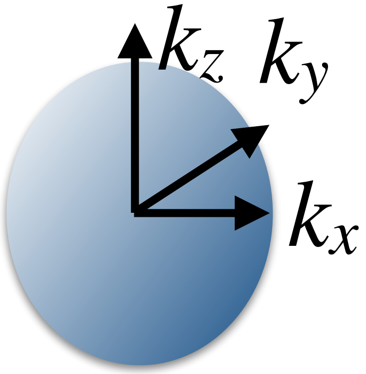
Bulk Quantum oscillations in Bi_2Se_3



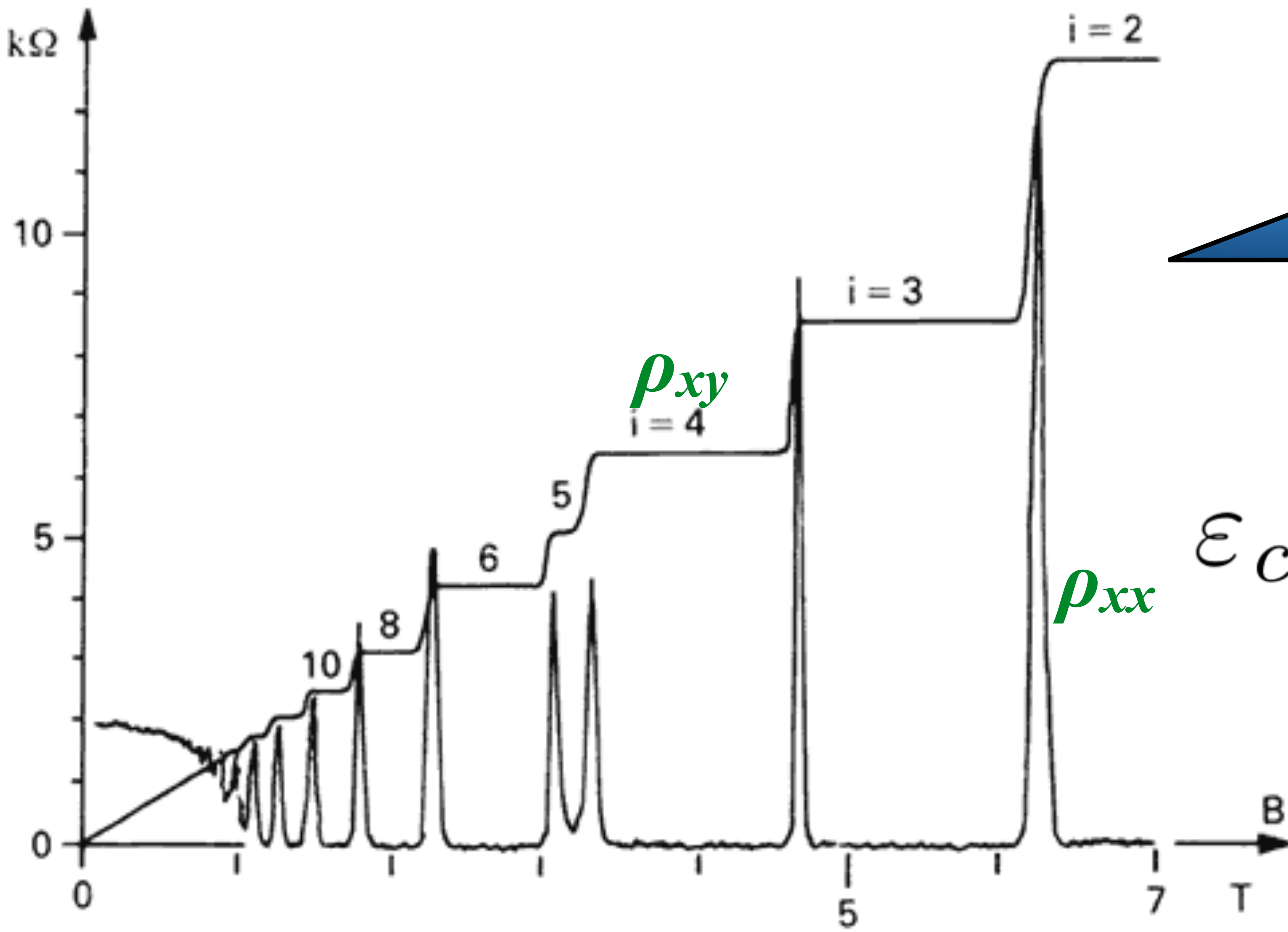


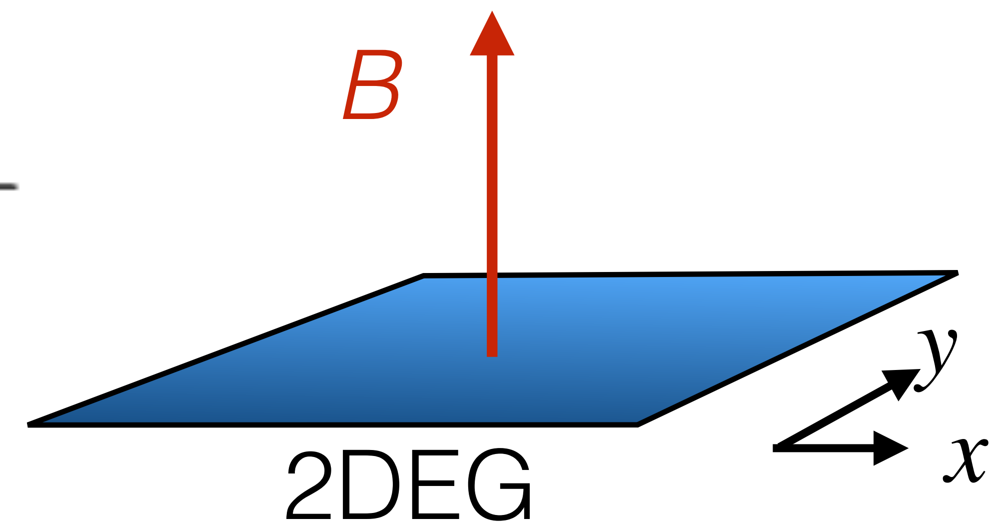






Integer quantum Hall effect



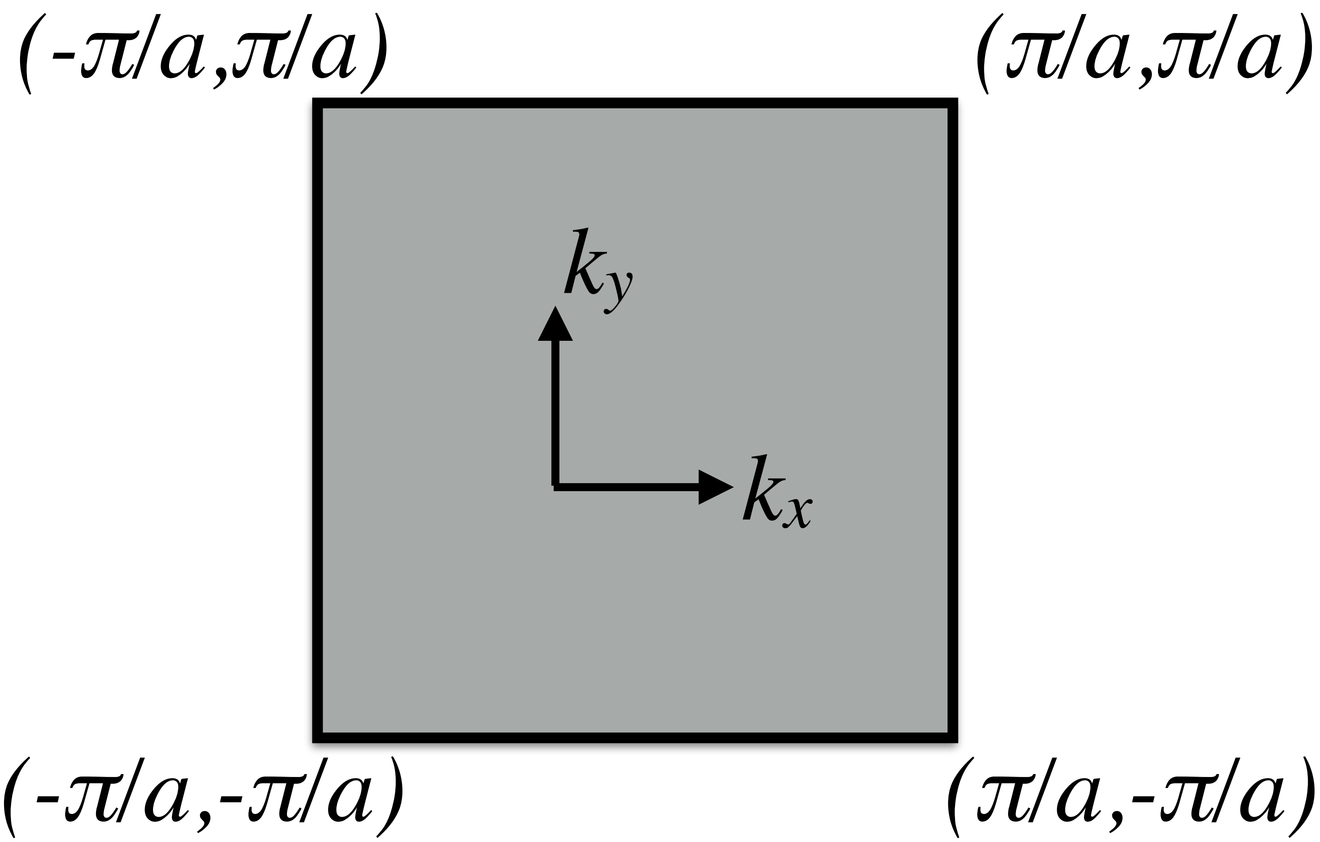


$$\varepsilon_c = (n + \gamma)\hbar\omega_c$$

$$\gamma = 1/2$$

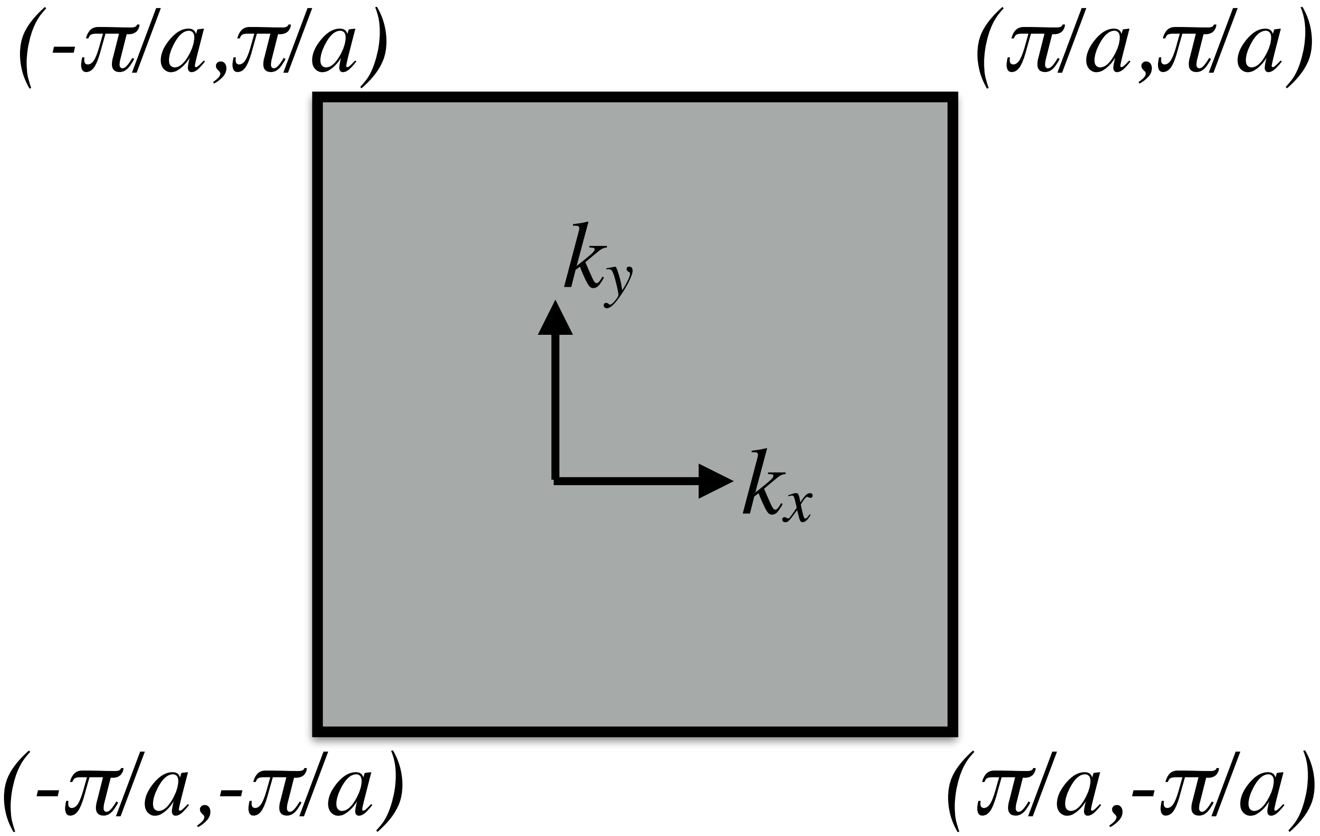
von Klitzing, Dorda and Pepper, PRL 1980

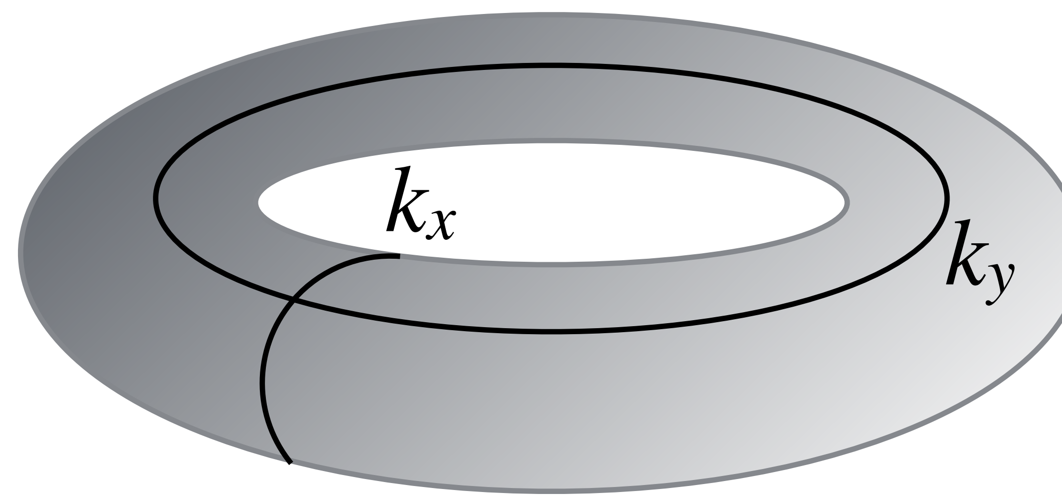
Berry curvature



- IQHE can be understood as a topological invariant arising from the topology of wave functions, which in this case exist in a (magnetic) Brillouin zone with the topology of a torus.

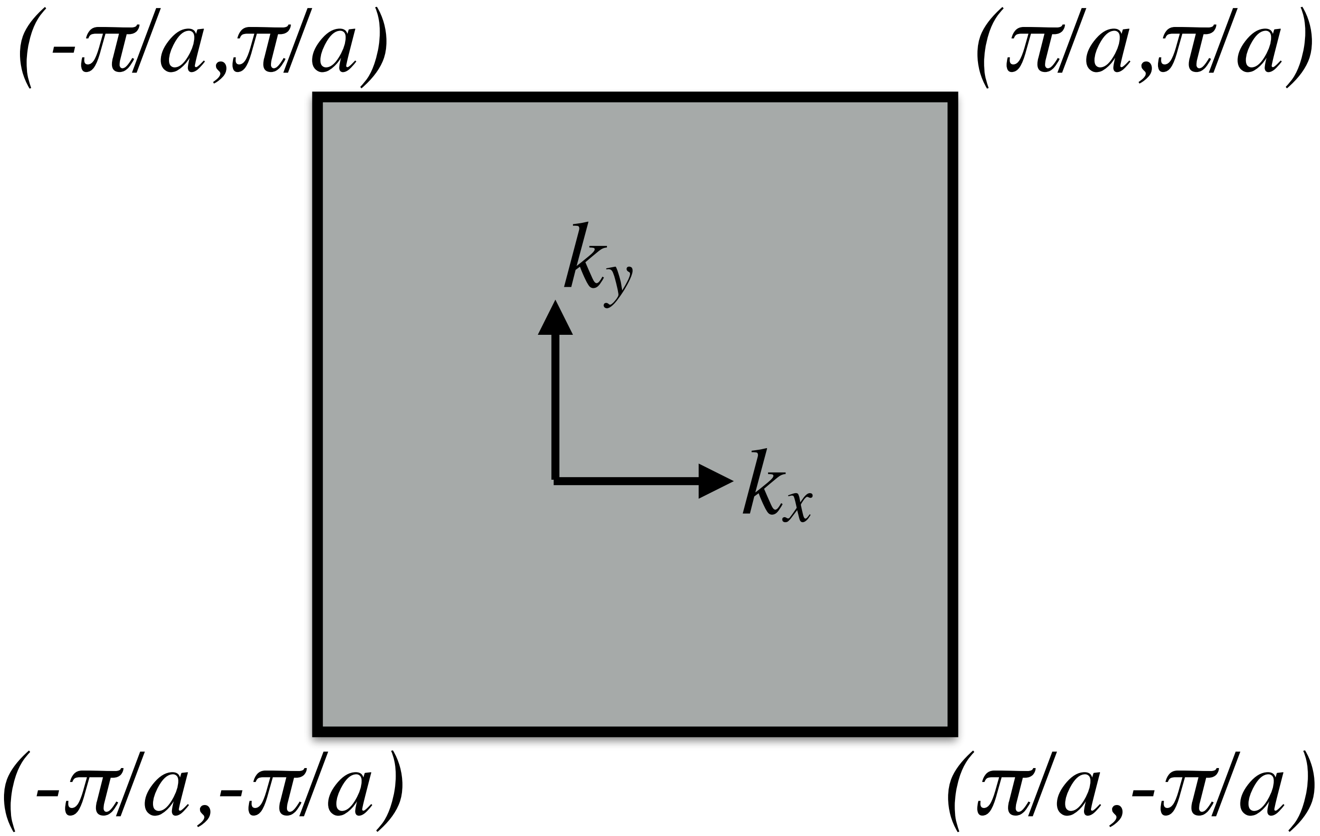
Berry curvature

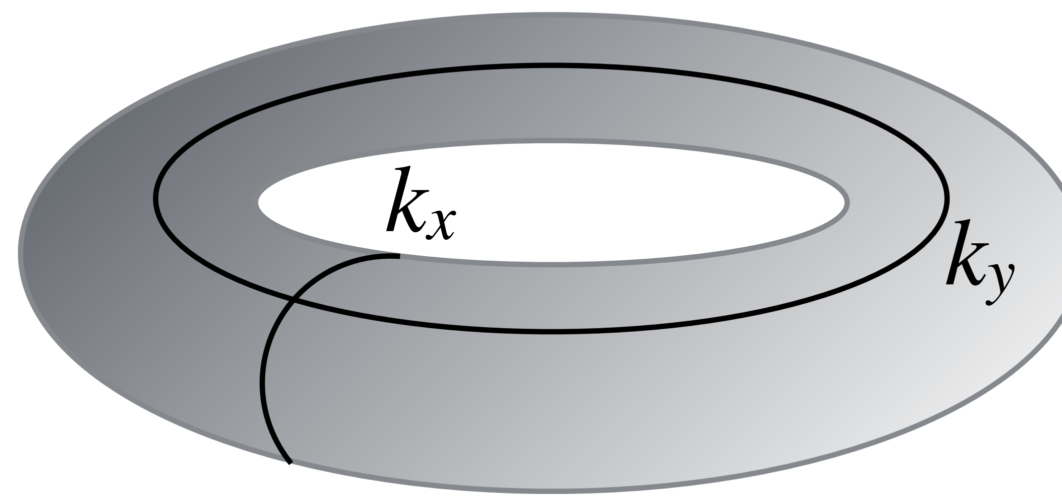




- IQHE can be understood as a topological invariant arising from the topology of wave functions, which in this case exist in a (magnetic) Brillouin zone with the topology of a torus.

Berry curvature

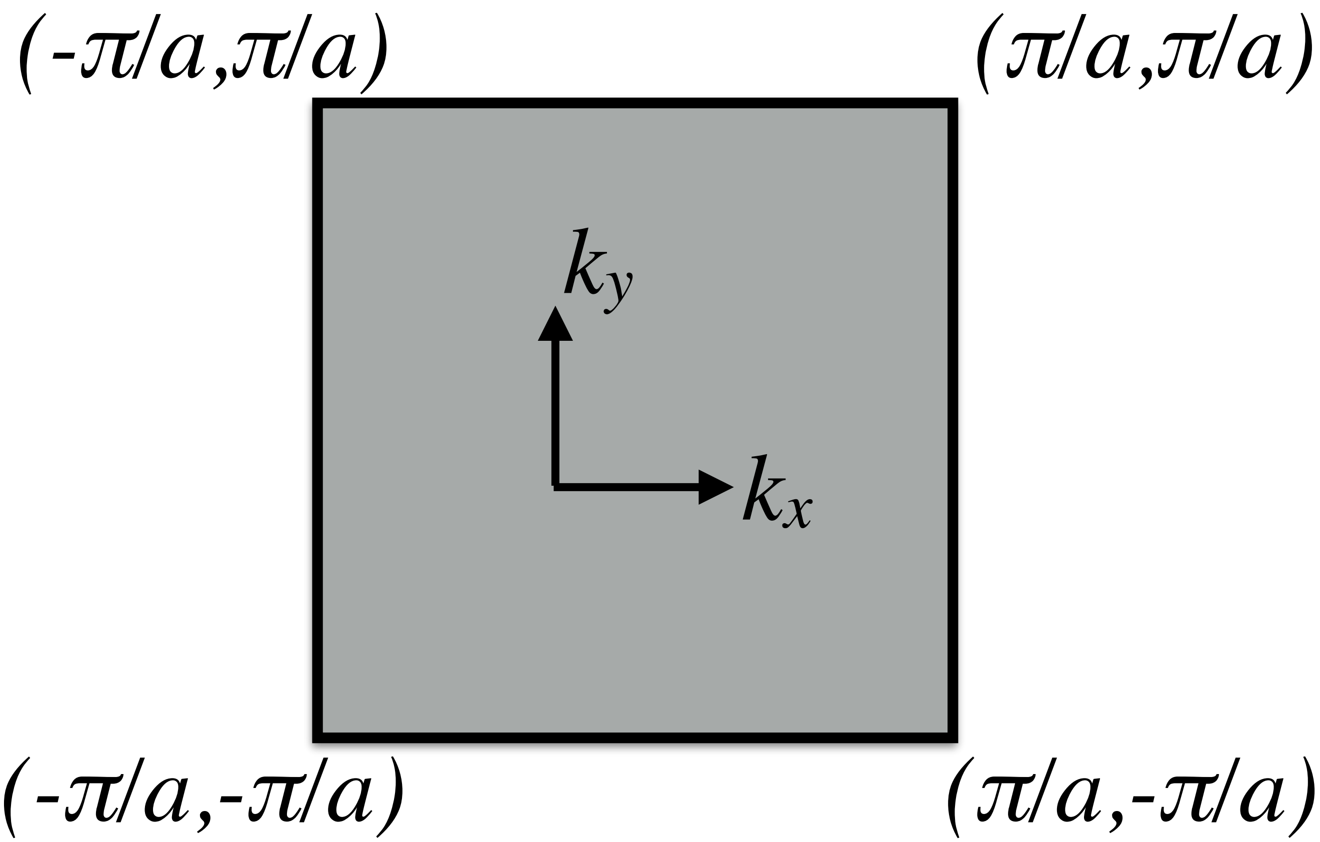


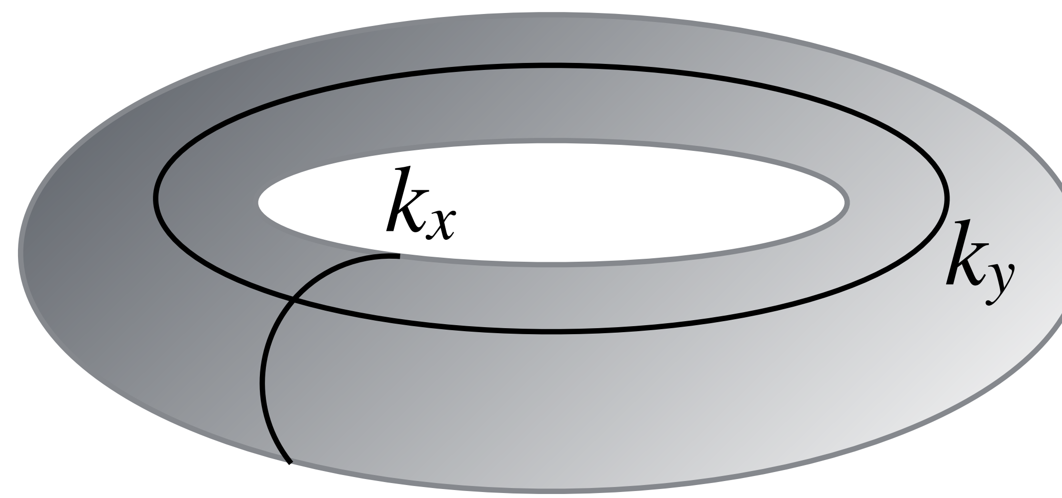


$$\gamma_B(C) = i \oint_C \langle n(\mathbf{R}) | \nabla_R | n(\mathbf{R}) \rangle$$

- IQHE can be understood as a topological invariant arising from the topology of wave functions, which in this case exist in a (magnetic) Brillouin zone with the topology of a torus.

Berry curvature



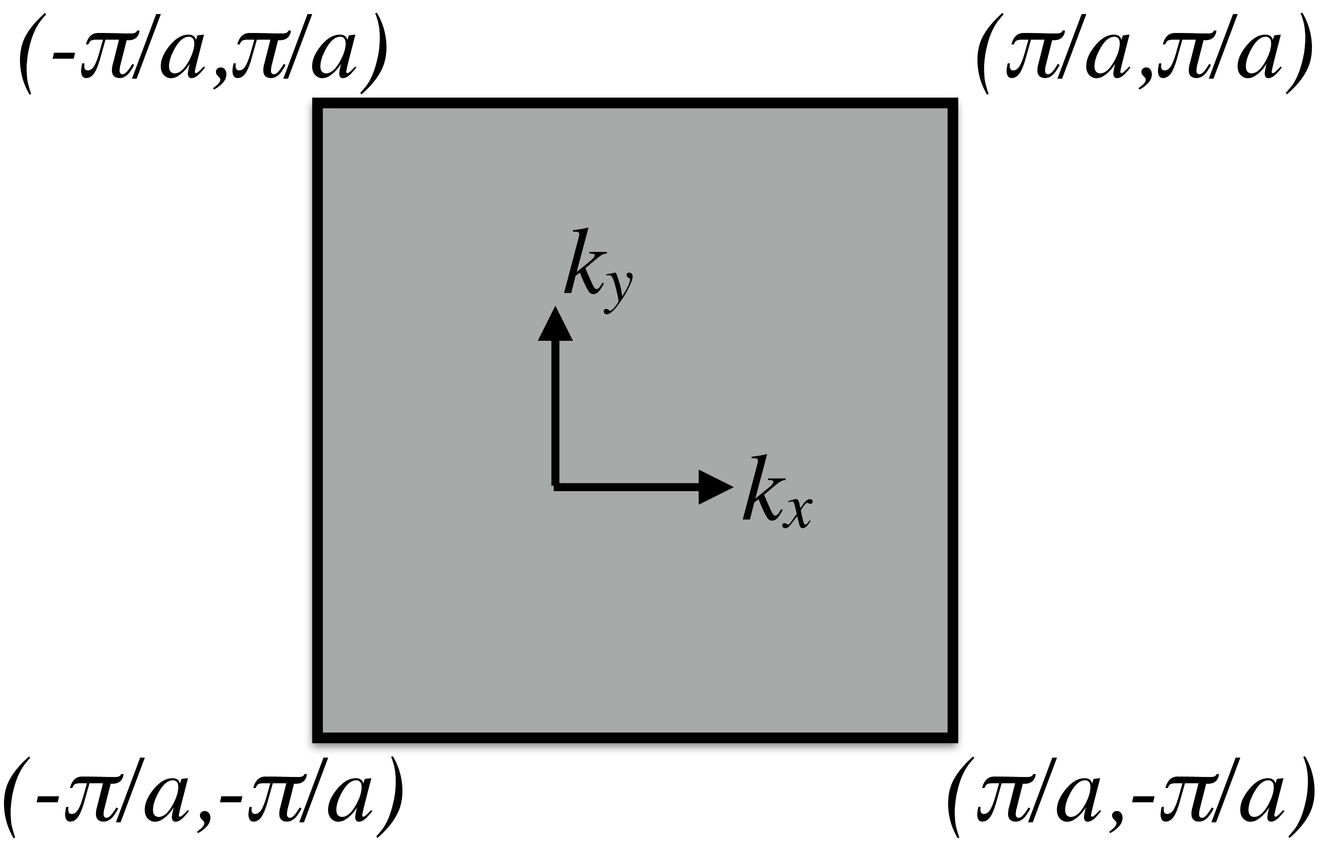


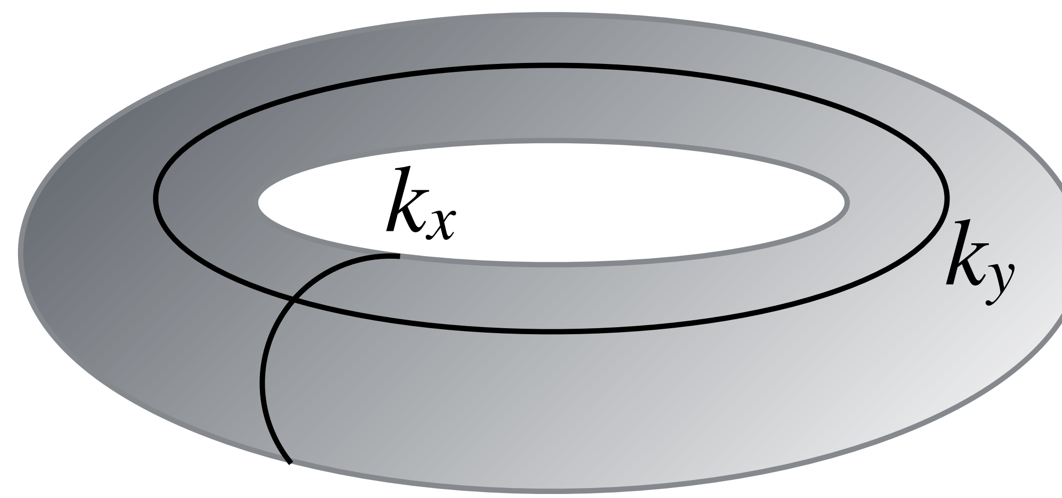
$$\gamma_B(C) = i \oint_C \langle n(\mathbf{R})|\nabla_R|n(\mathbf{R})\rangle$$

$$\Omega = \nabla \times \langle n(\mathbf{R})|\nabla_R|n(\mathbf{R})\rangle$$

- IQHE can be understood as a topological invariant arising from the topology of wave functions, which in this case exist in a (magnetic) Brillouin zone with the topology of a torus.

Berry curvature





$$\gamma_B(C) = i \oint_C \langle n(\mathbf{R}) | \nabla_R | n(\mathbf{R}) \rangle$$

$$\Omega = \nabla \times \langle n(\mathbf{R}) | \nabla_R | n(\mathbf{R}) \rangle$$

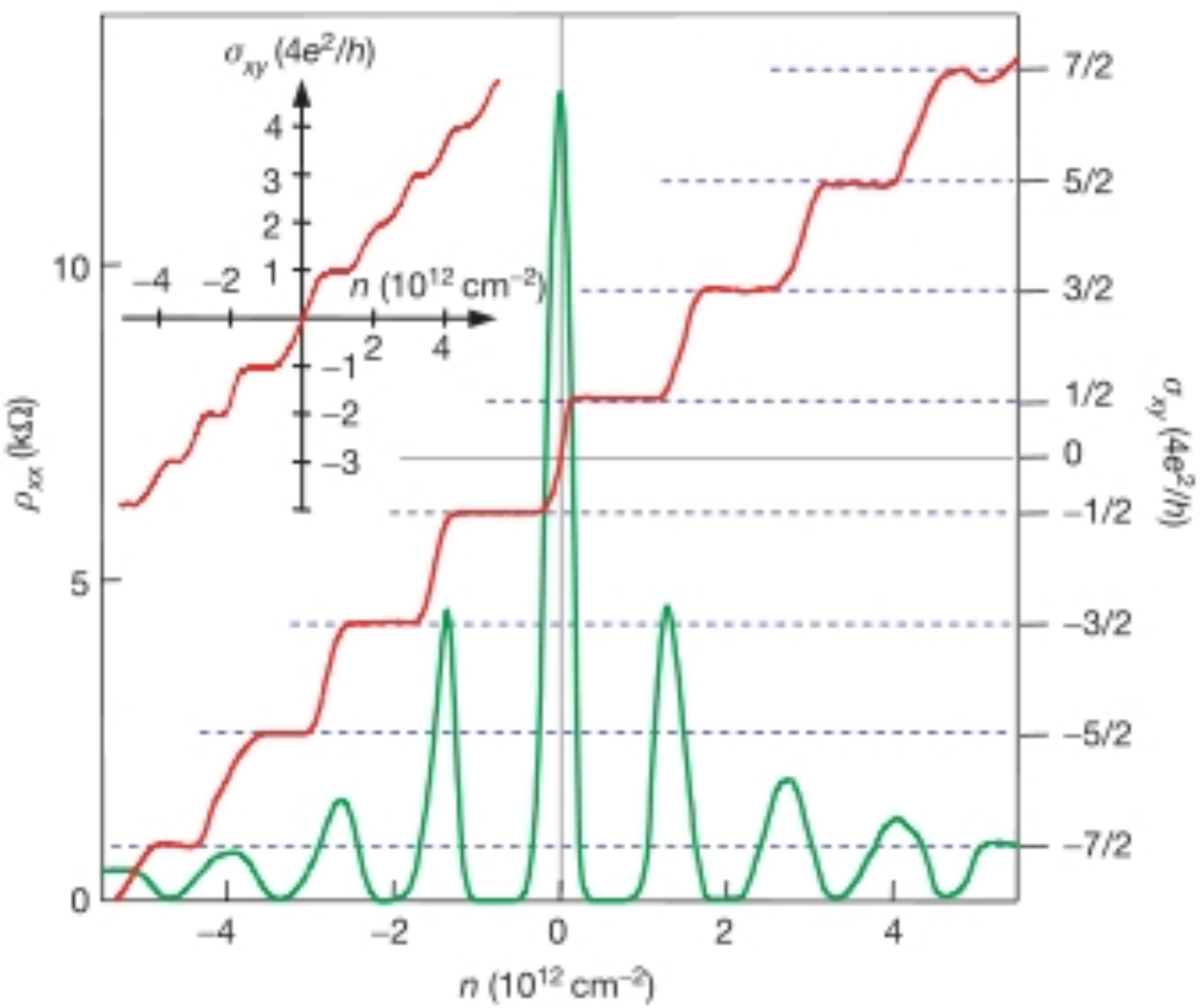
$$\sigma_{xy} = \frac{e^2}{\hbar} \int_{BZ} \frac{d^2k}{(2\pi)^2} \Omega$$

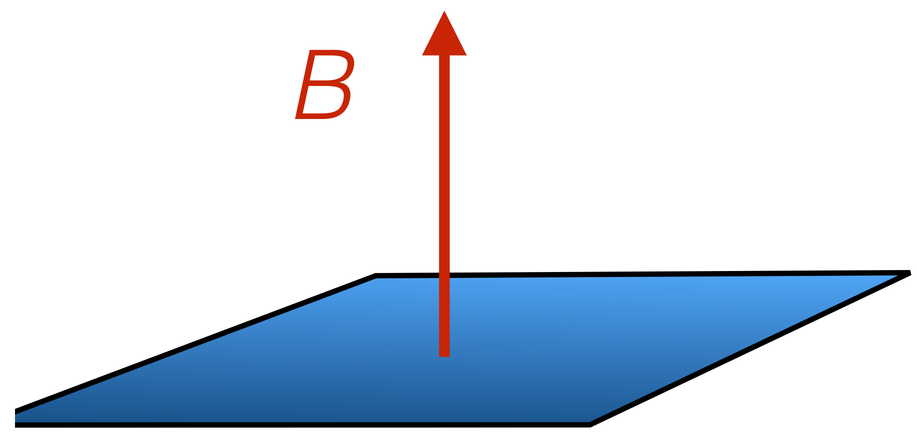
- IQHE can be understood as a topological invariant arising from the topology of wave functions, which in this case exist in a (magnetic) Brillouin zone with the topology of a torus.

$$\gamma = \gamma(0) - \frac{\gamma_B}{2\pi} \approx 1/2 - \frac{\gamma_B}{2\pi}$$

Mikitik and Sharlai PRL 1999, also Laura Roth PRB 1966

Integer quantum Hall effect (graphene)



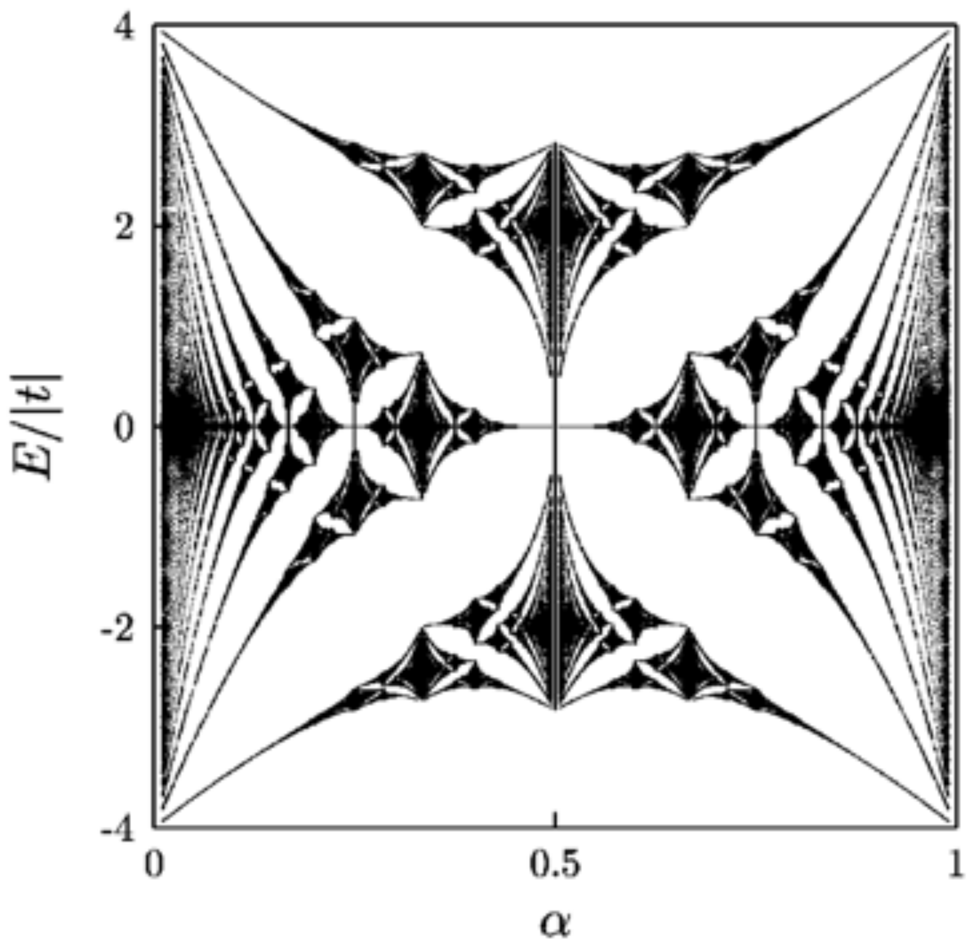


$$\varepsilon_n = \pm v_F \sqrt{2ne\hbar B}$$

$$\gamma = 0$$

P. Kim & Novoselov/Geim *Nature* 2005

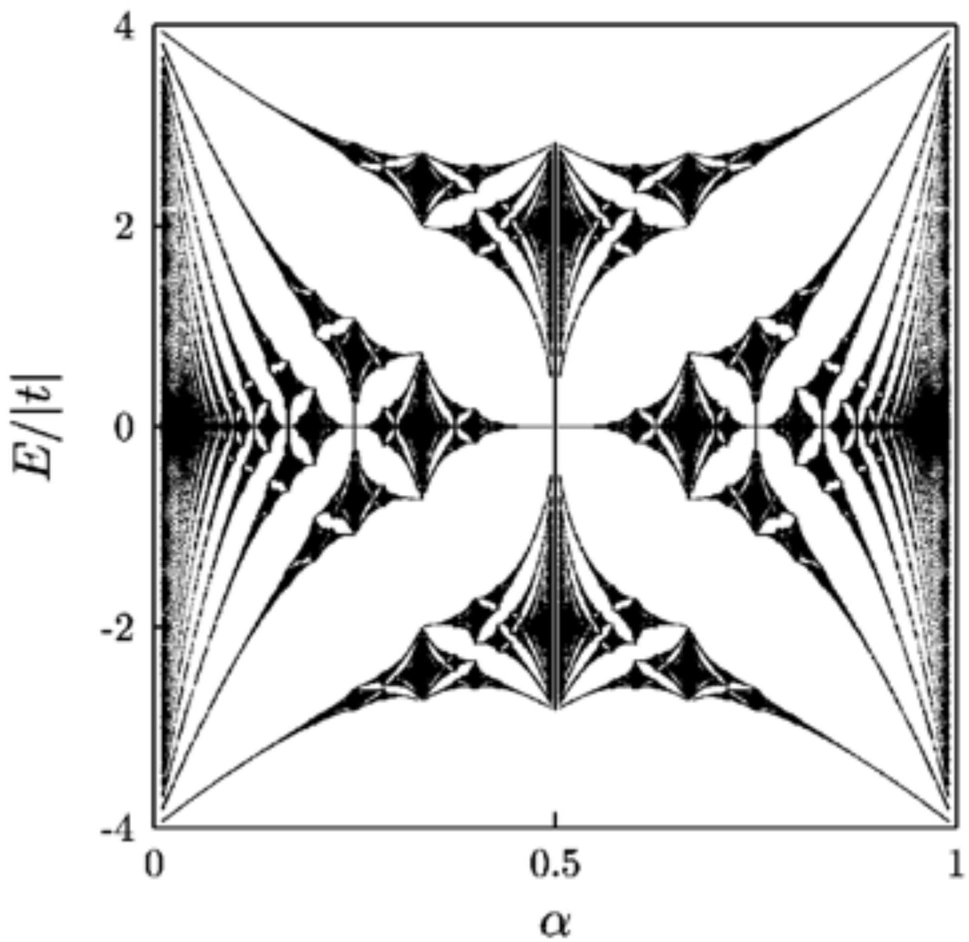
Landau quantization in a 2D square lattice



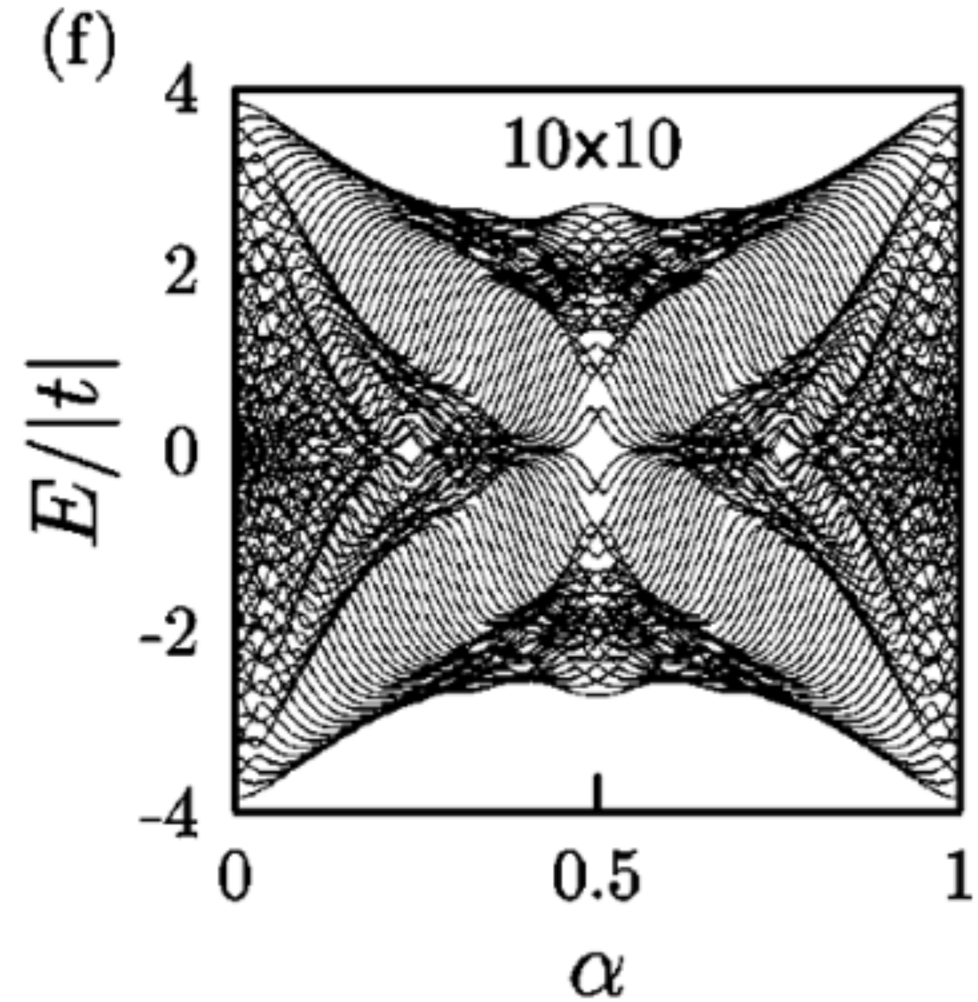
Hofstadter Butterfly (PRB 1976)

Analytis AMJP 2004

Landau quantization in a 2D square lattice

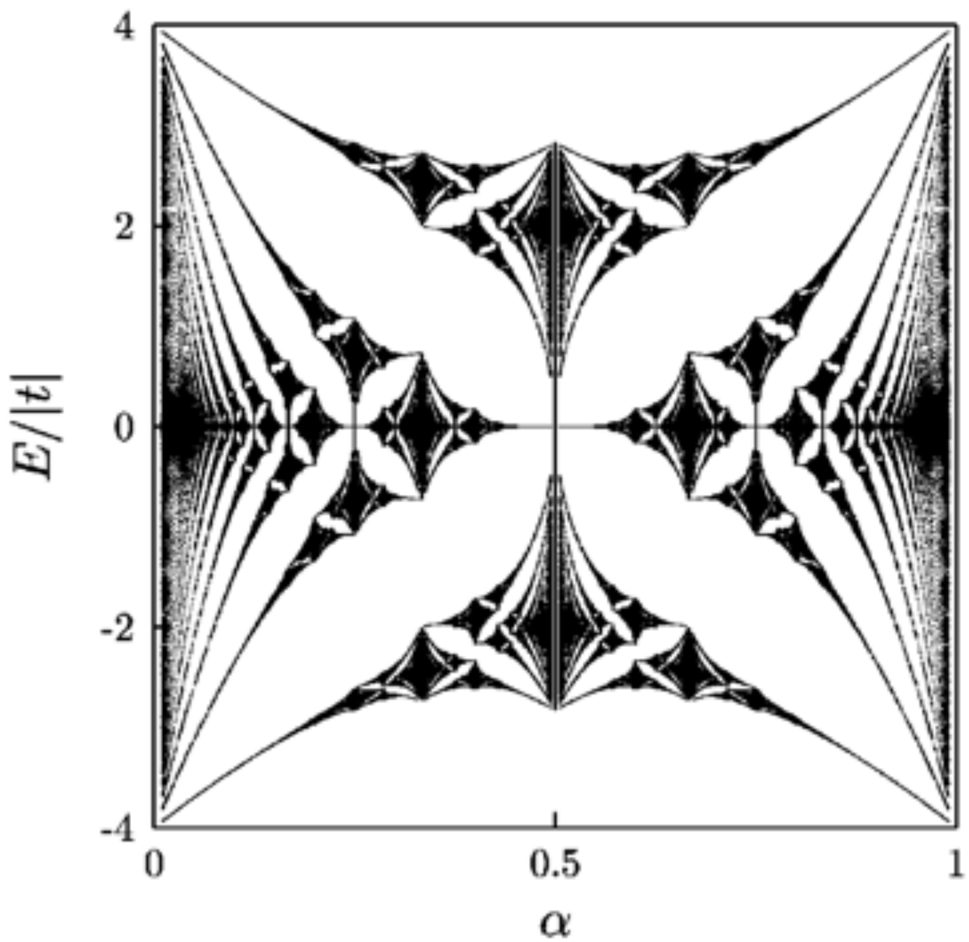


Hofstadter Butterfly (PRB 1976)

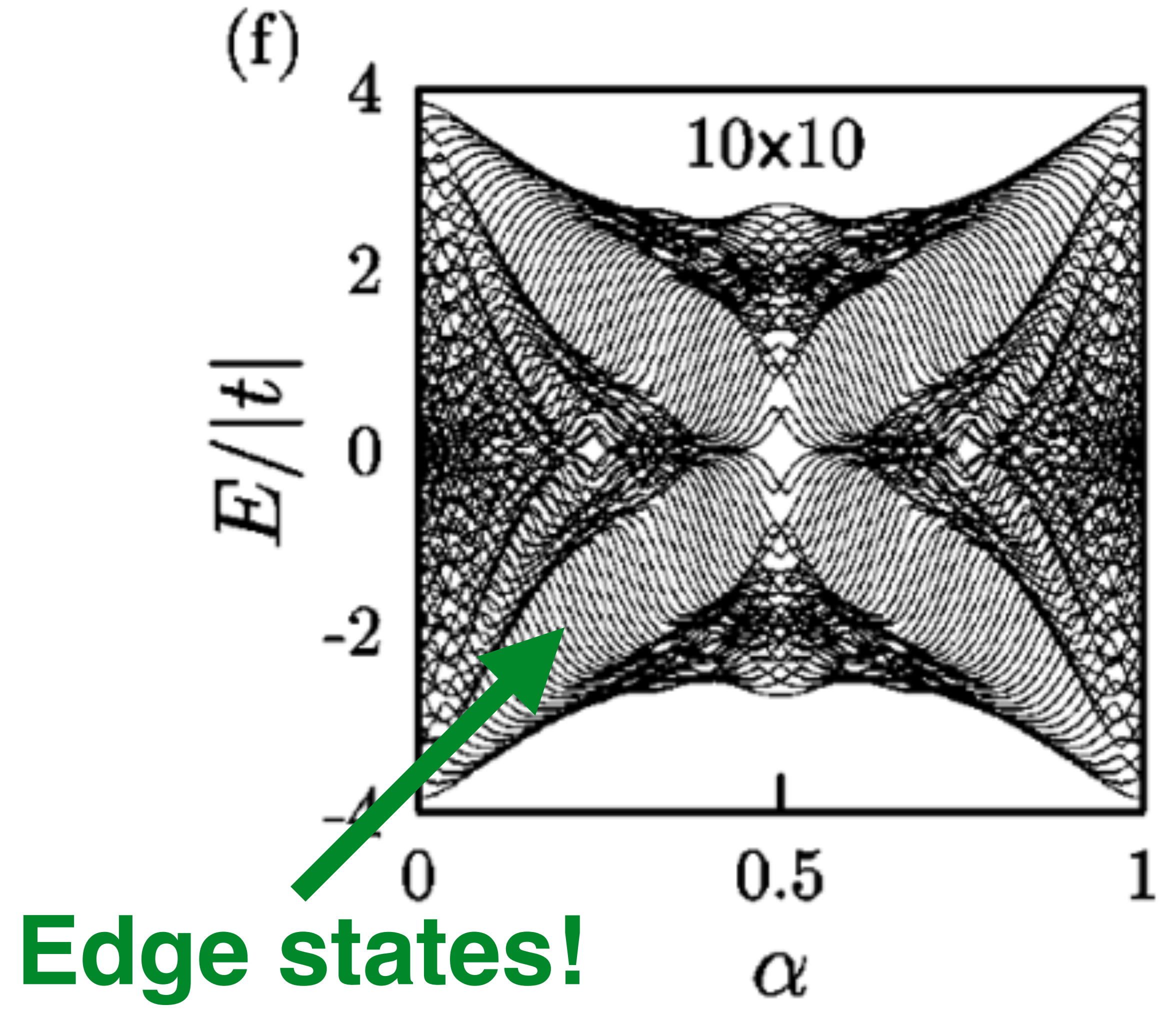


Analytis AMJP 2004

Landau quantization in a 2D square lattice

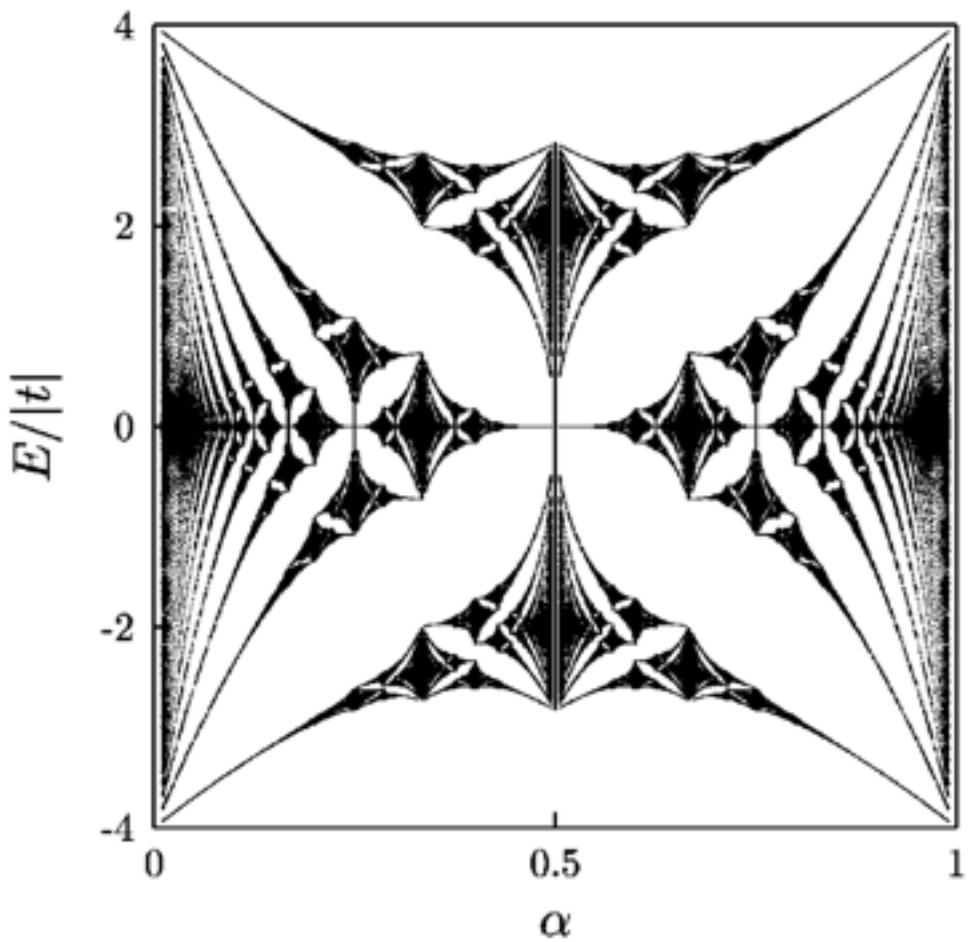


Hofstadter Butterfly (PRB 1976)

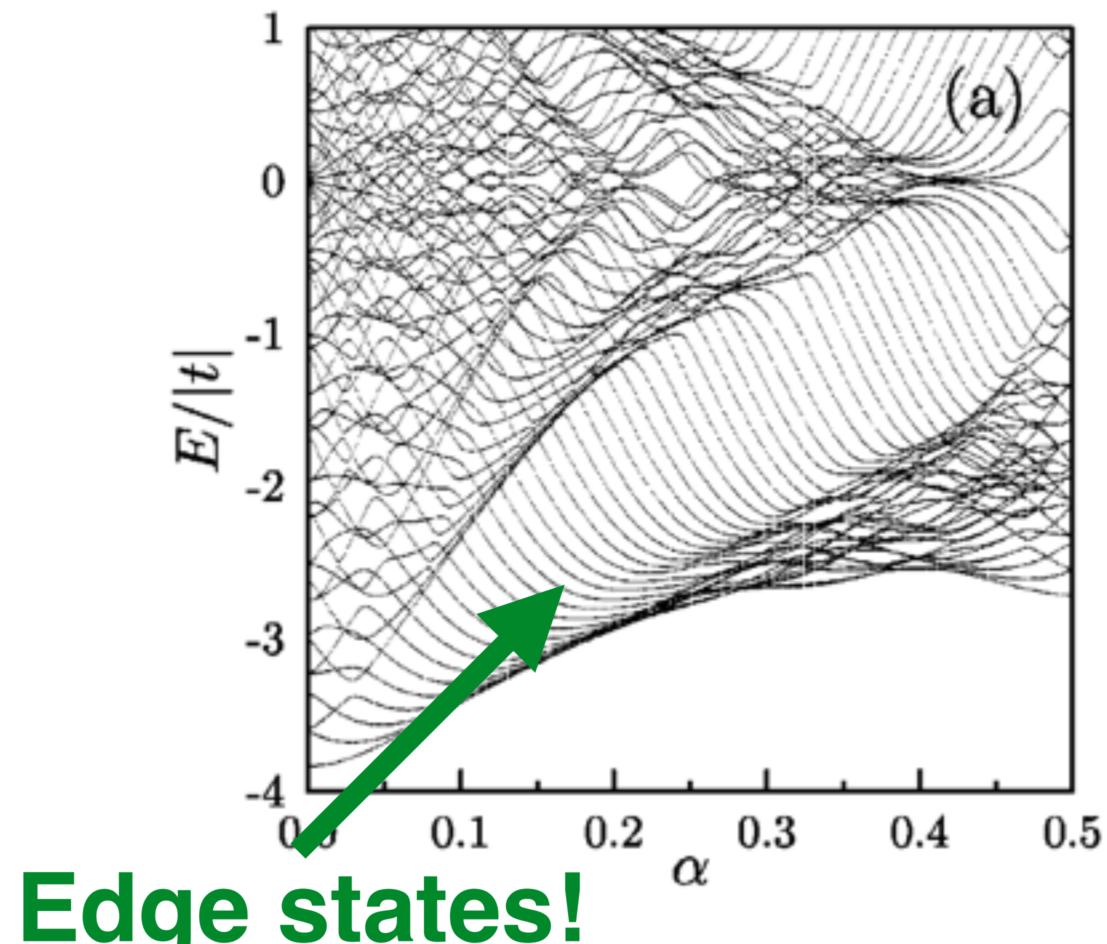


Analytis AMJP 2004

Landau quantization in a 2D square lattice

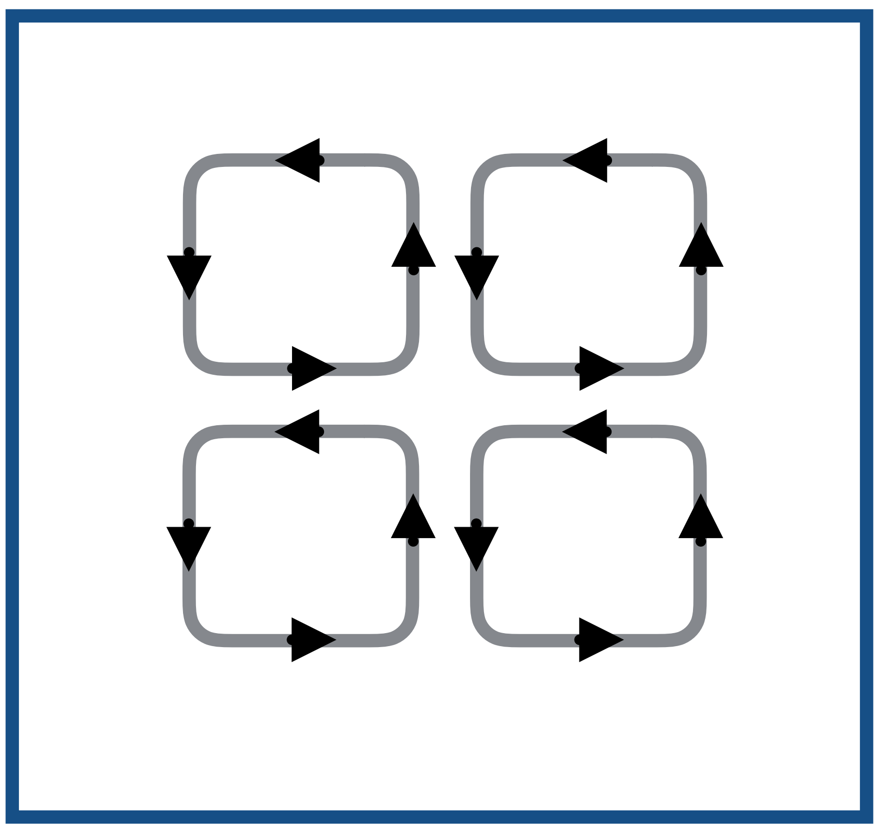


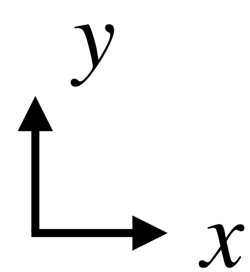
Hofstadter Butterfly (PRB 1976)

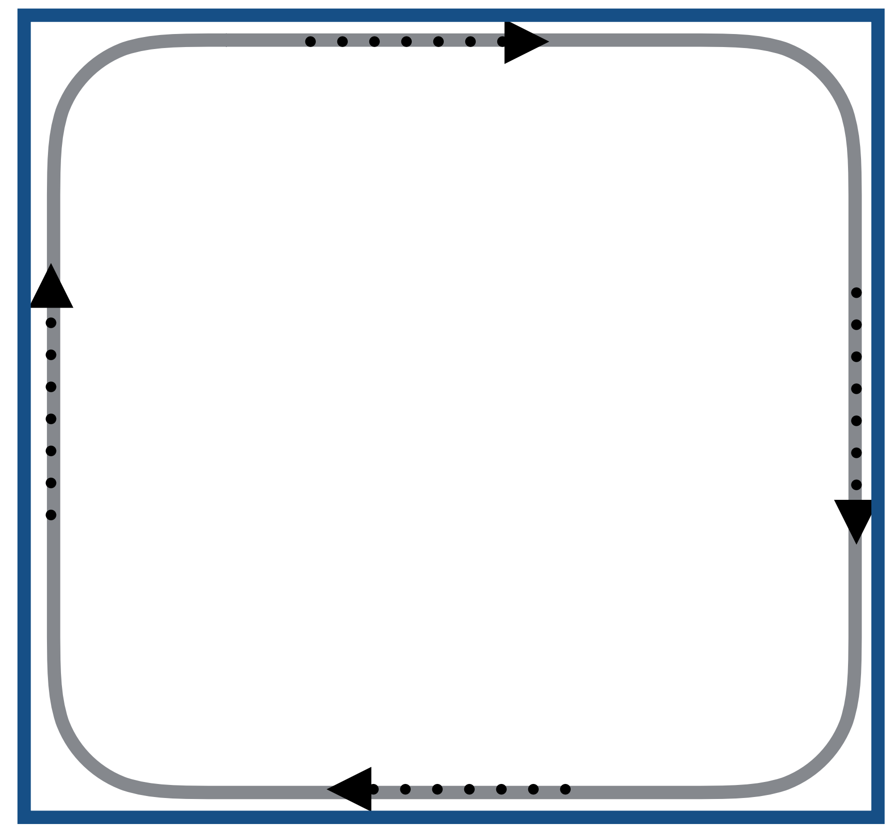


Analytis AMJP 2004

Landau quantization in a 2D square lattice







Bulk states

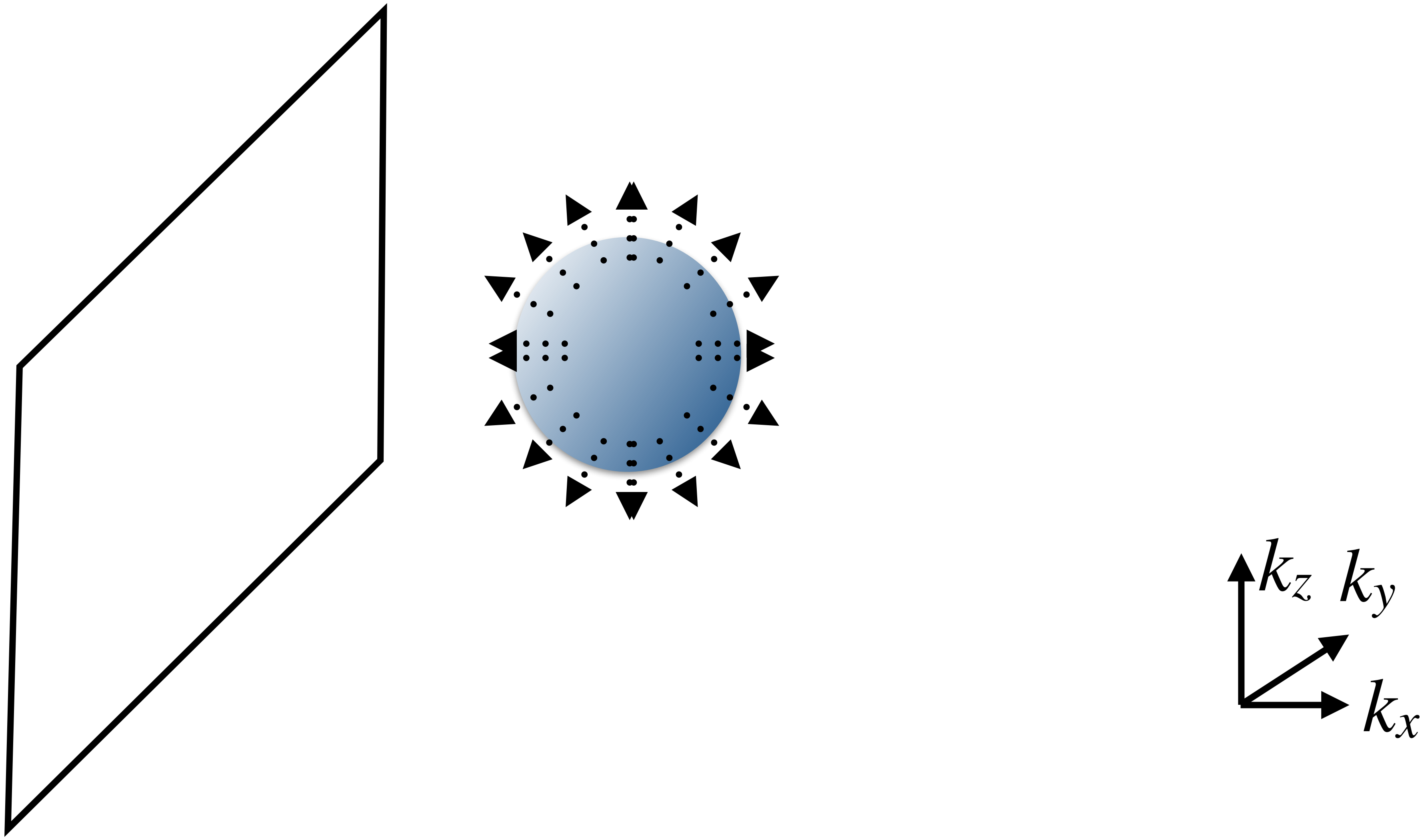
Edge states

By breaking the translation symmetry at the boundary, we immediately get edge states that connect the bulk states

Part II: Fermi arcs and Weyl orbits

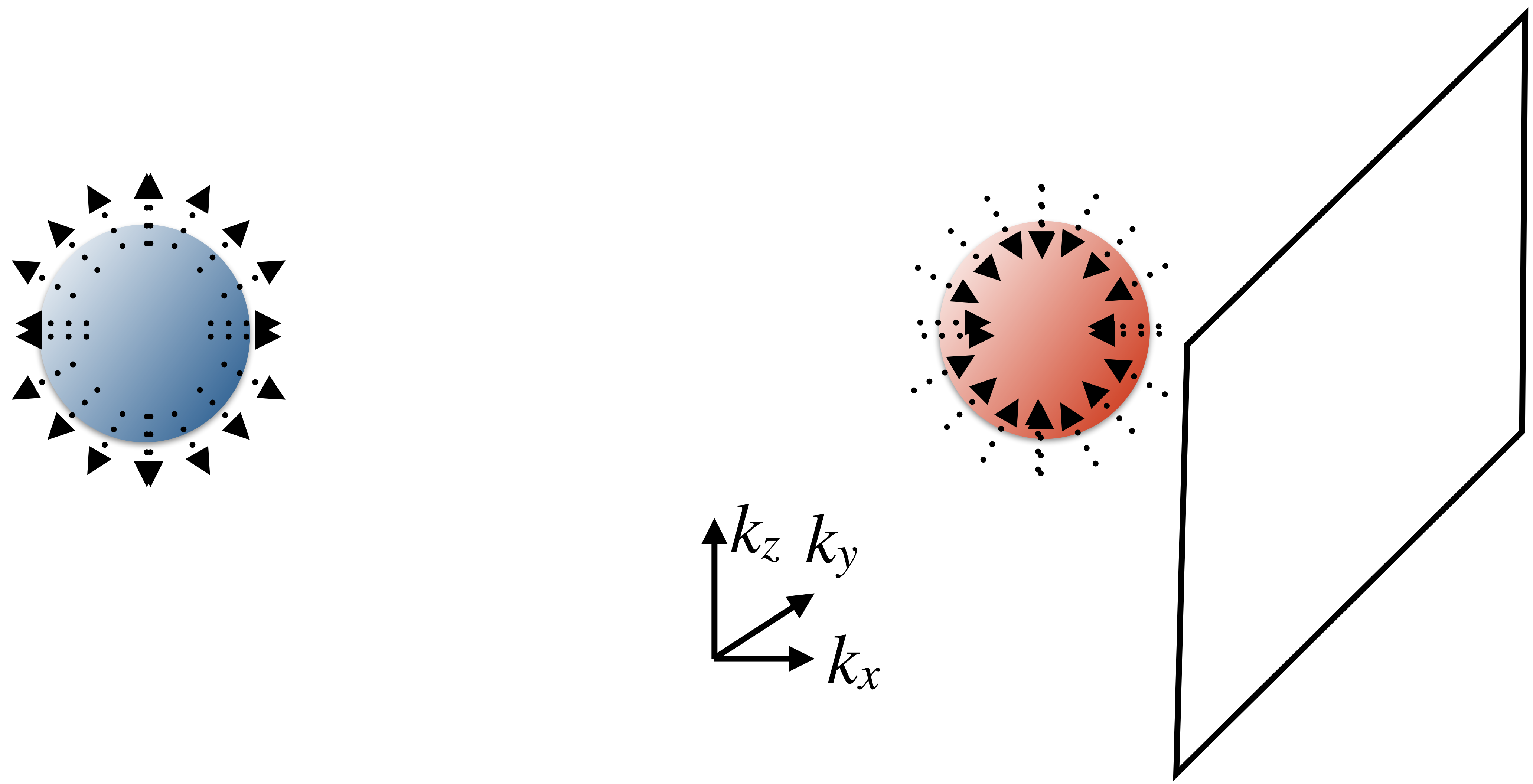
Weyl nodes - ‘magnetic monopoles’ in reciprocal space

$$H_\pm = \pm v_F(k_x\sigma_x + k_y\sigma_y + k_z\sigma_z)$$



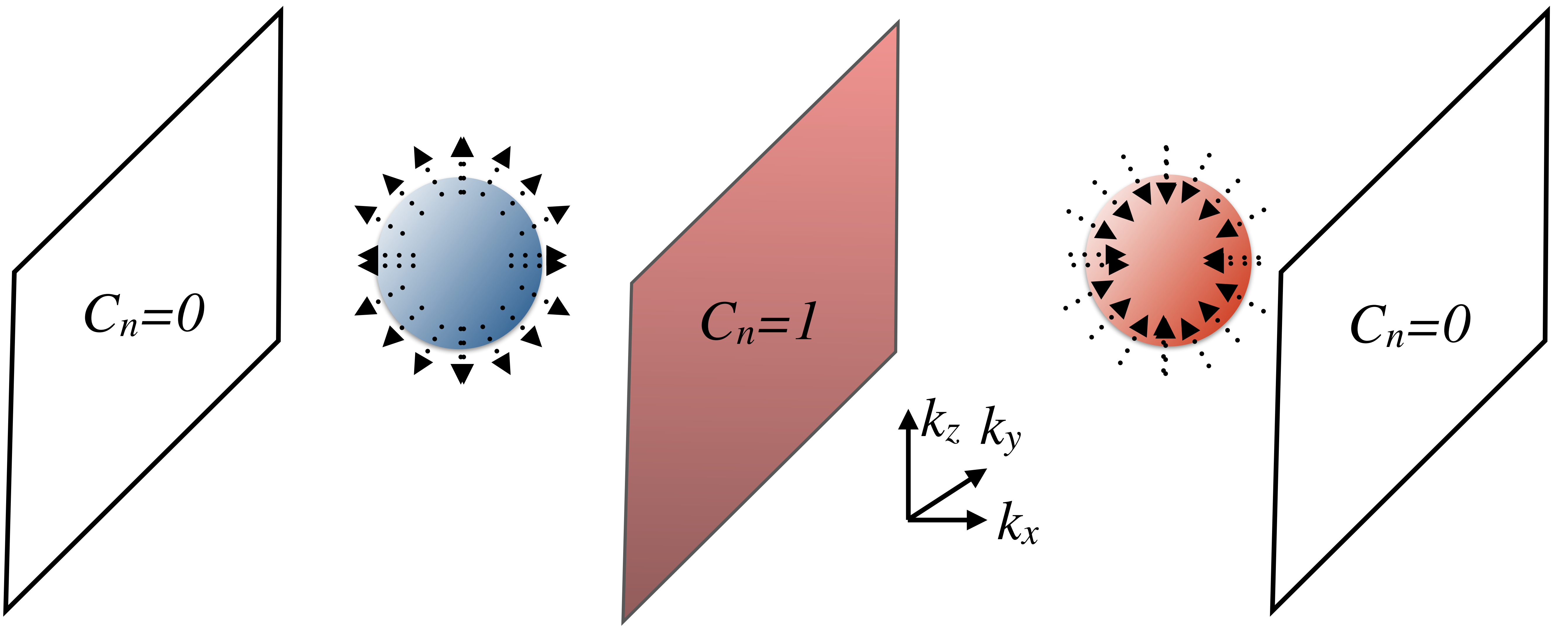
Weyl nodes - ‘magnetic monopoles’ in reciprocal space

$$H_{\pm} = \pm v_F(k_x\sigma_x + k_y\sigma_y + k_z\sigma_z)$$

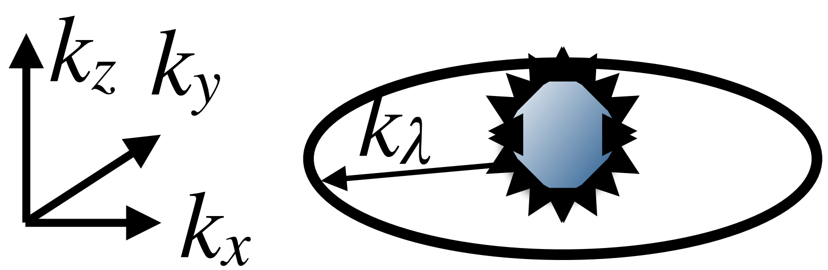


Weyl nodes - ‘magnetic monopoles’ in reciprocal space

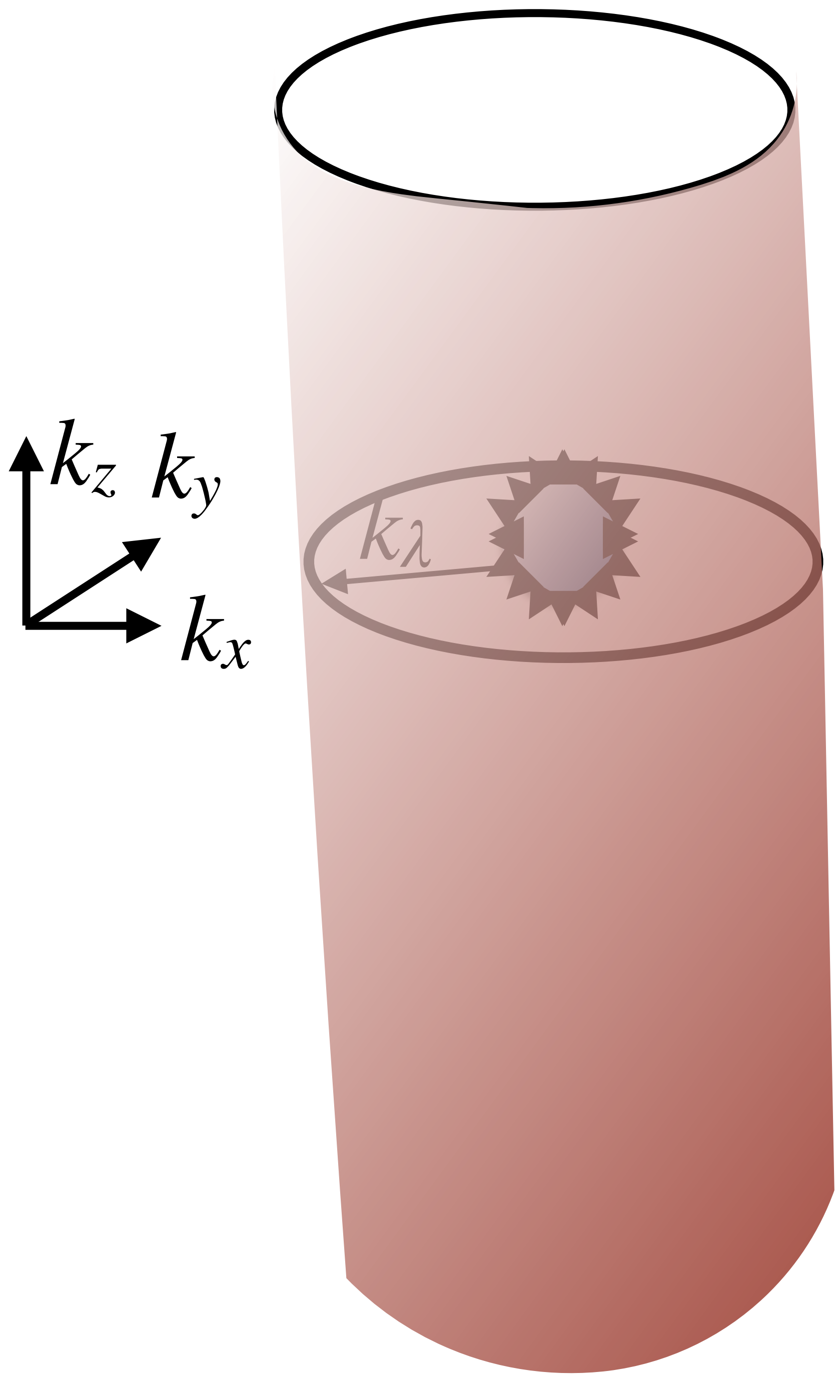
$$H_{\pm} = \pm v_F(k_x\sigma_x + k_y\sigma_y + k_z\sigma_z)$$



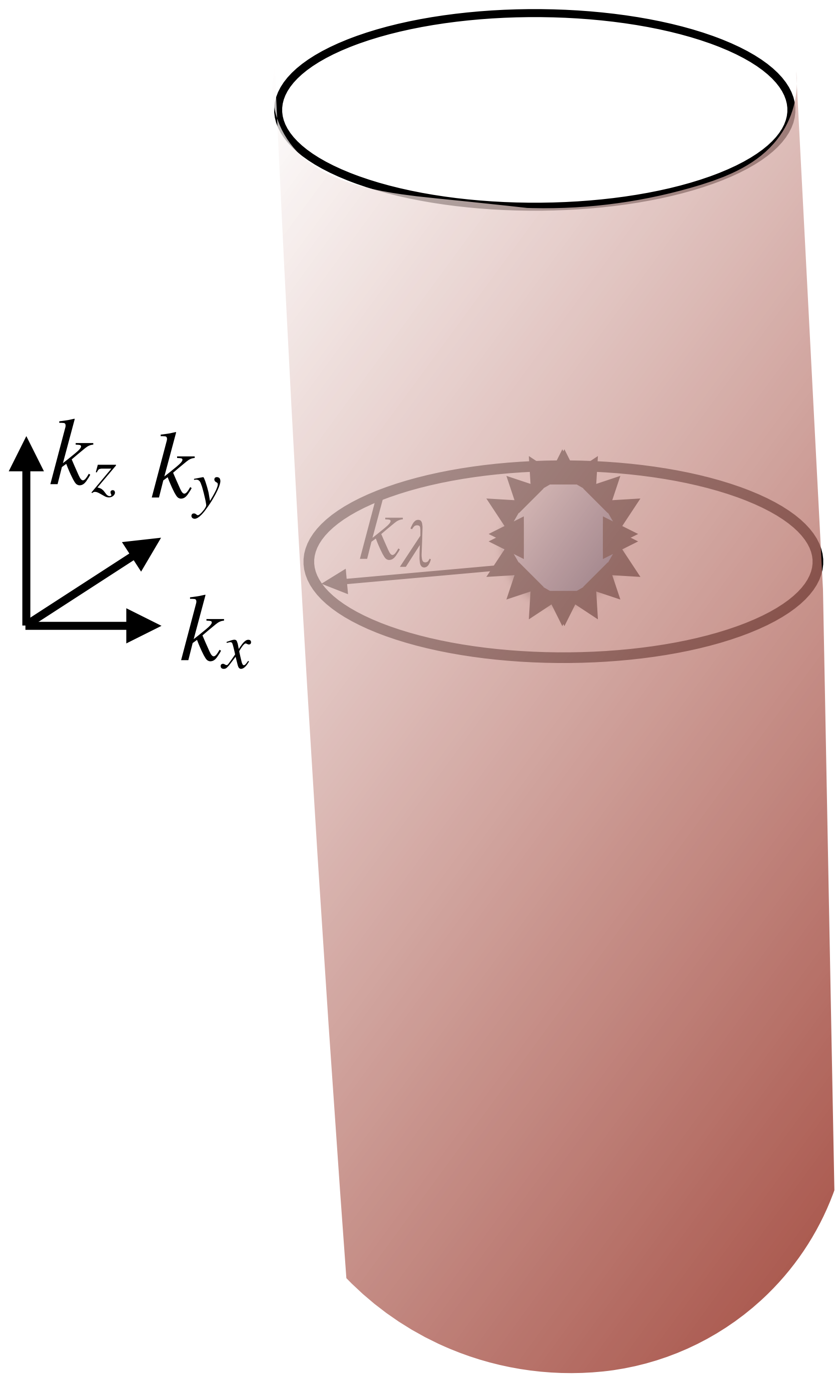
Weyl nodes - ‘magnetic monopoles’ in reciprocal space

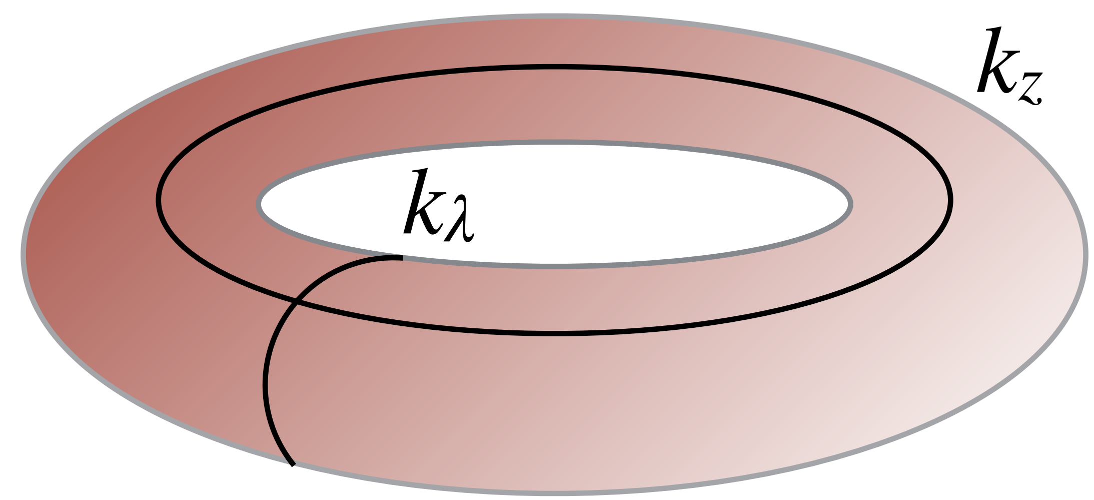


Weyl nodes - ‘magnetic monopoles’ in reciprocal space

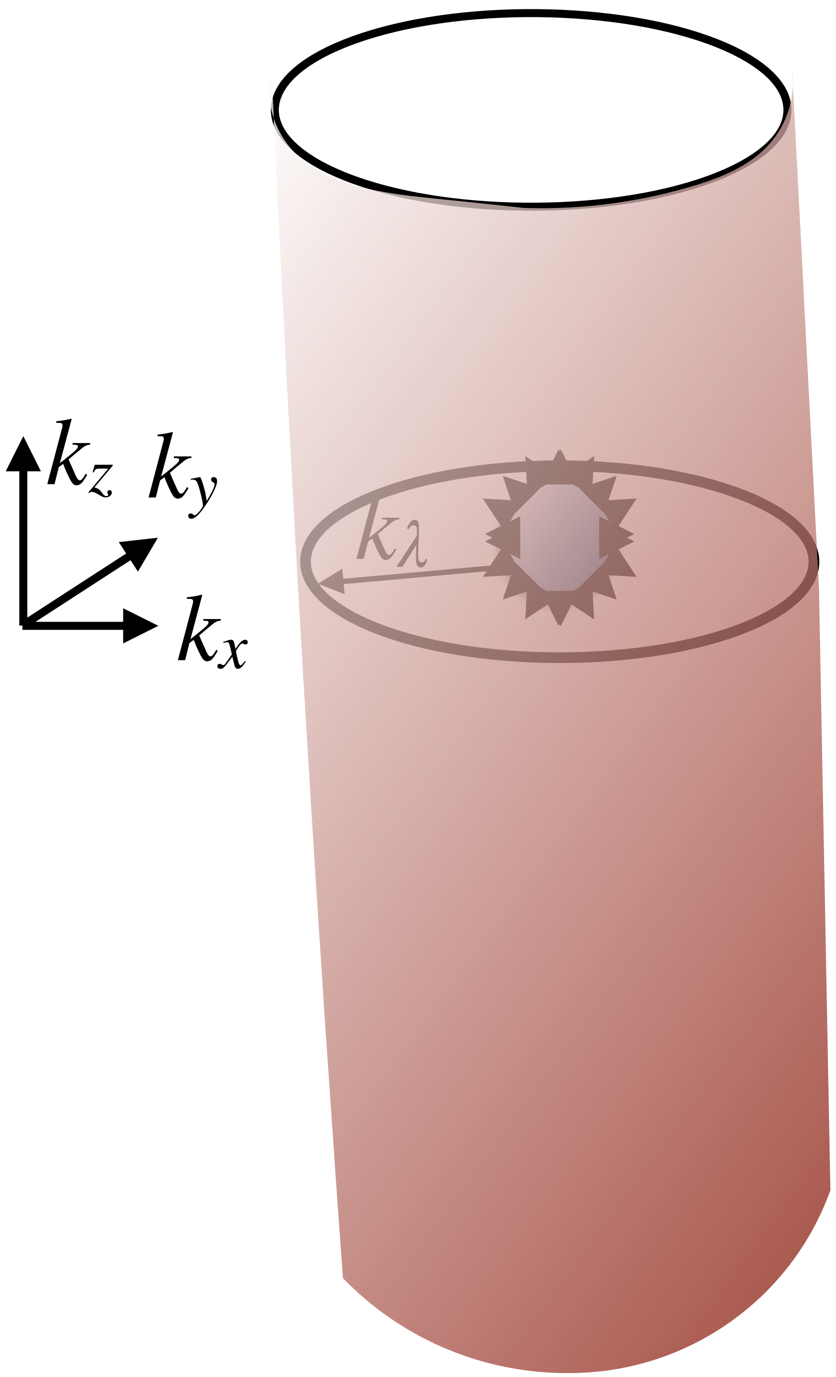


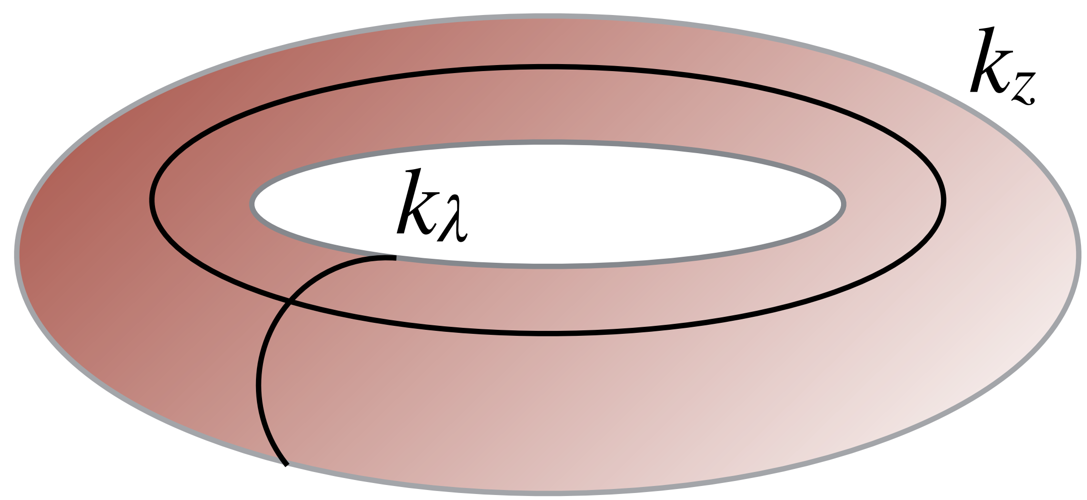
Weyl nodes - ‘magnetic monopoles’ in reciprocal space





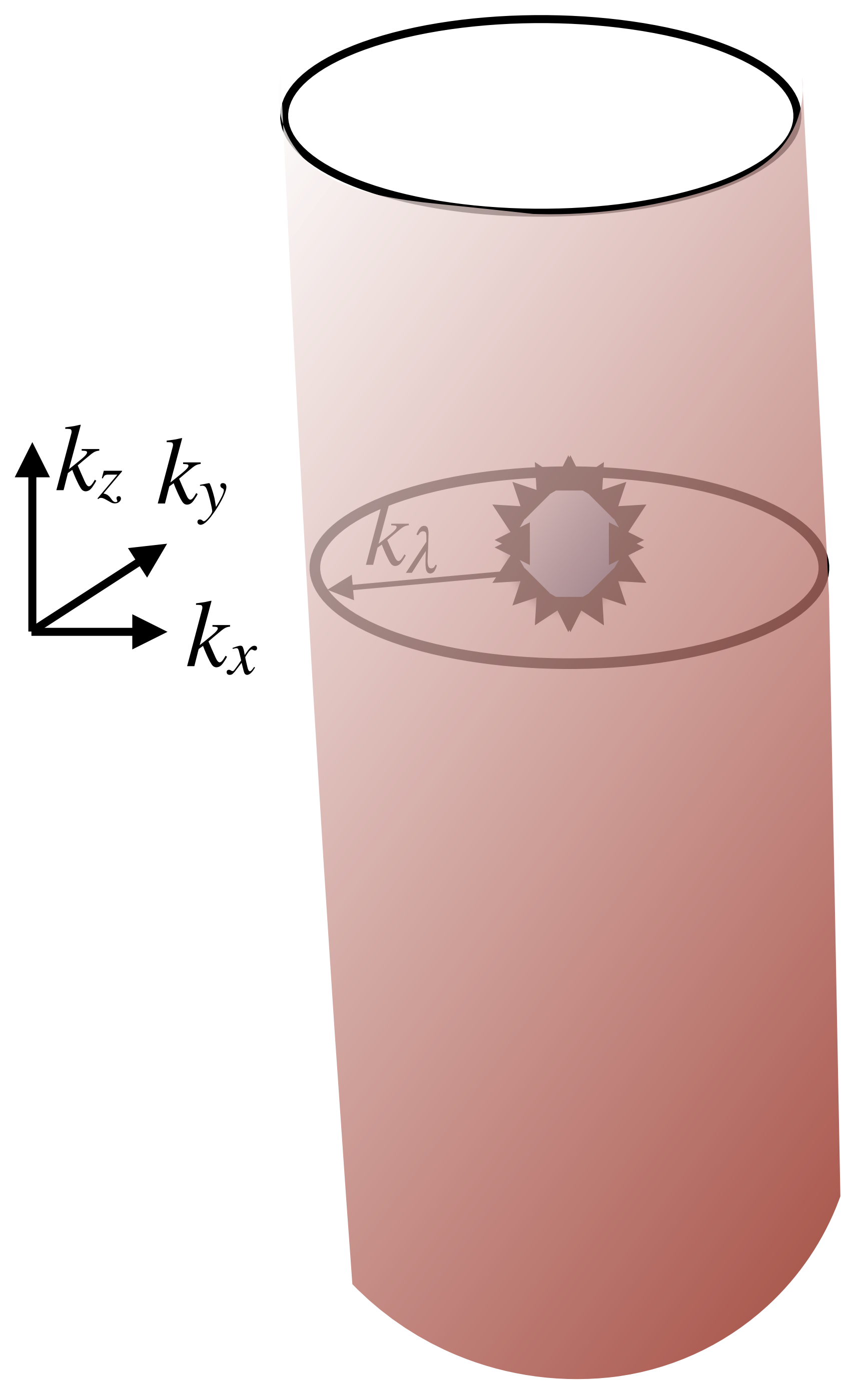
Weyl nodes - ‘magnetic monopoles’ in reciprocal space

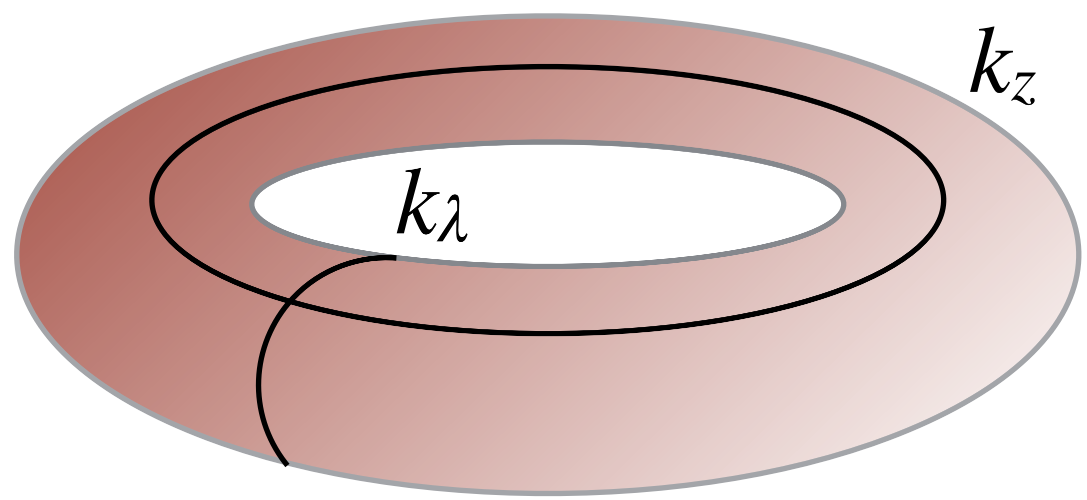




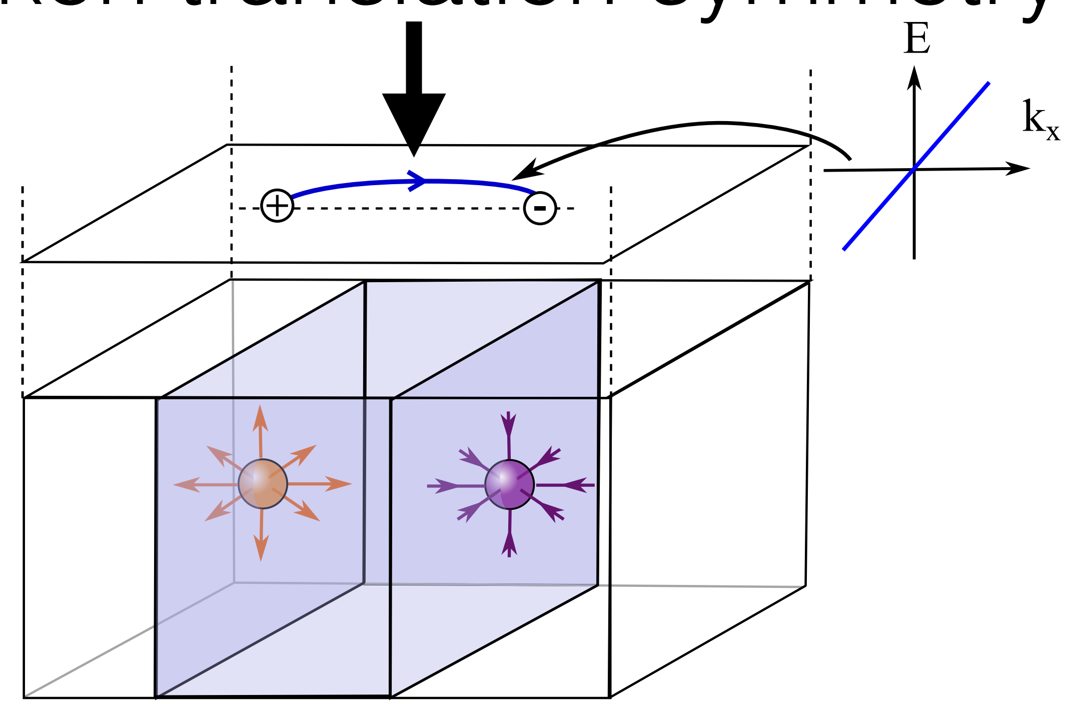
+ broken translation symmetry

Weyl nodes - ‘magnetic monopoles’ in reciprocal space

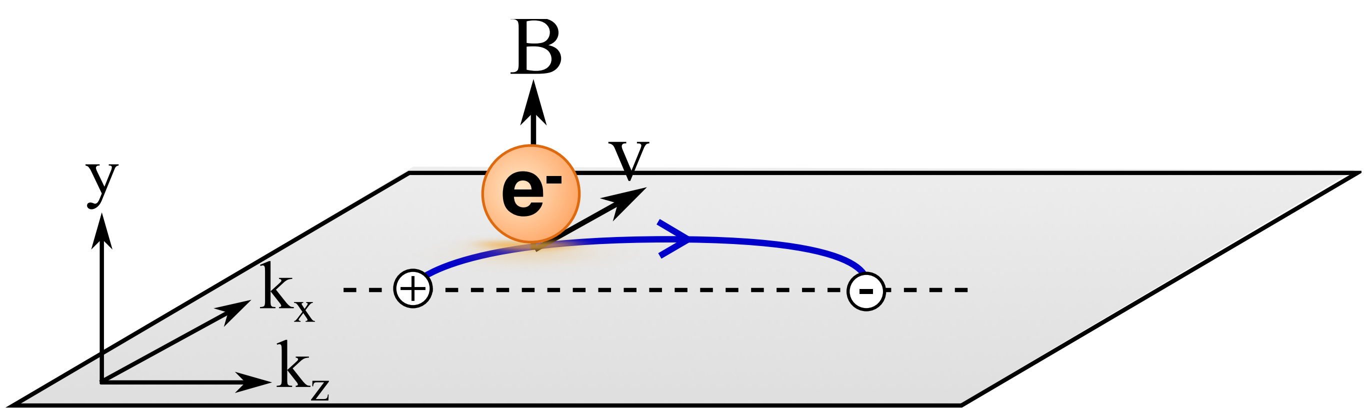




+ broken translation symmetry

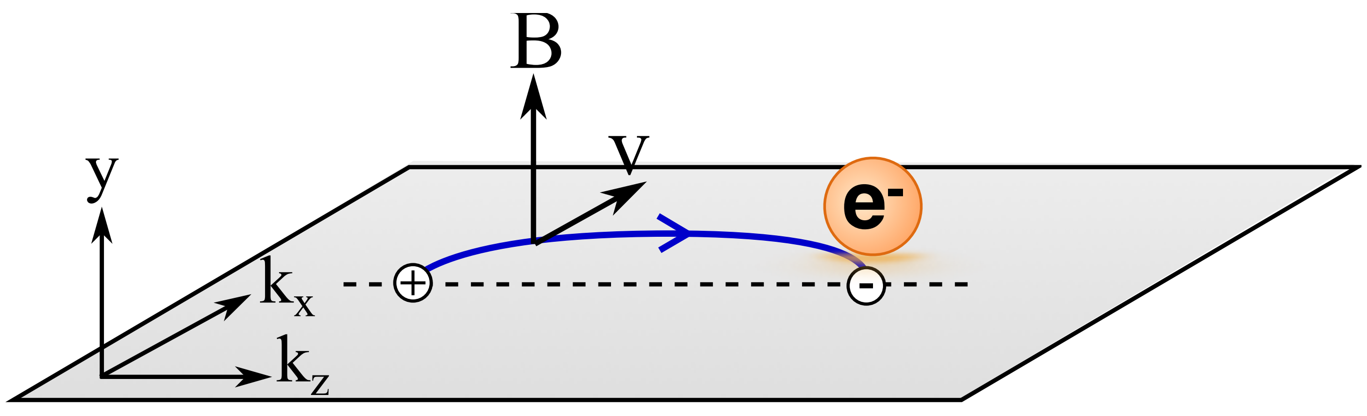


The chirality conveyor belt



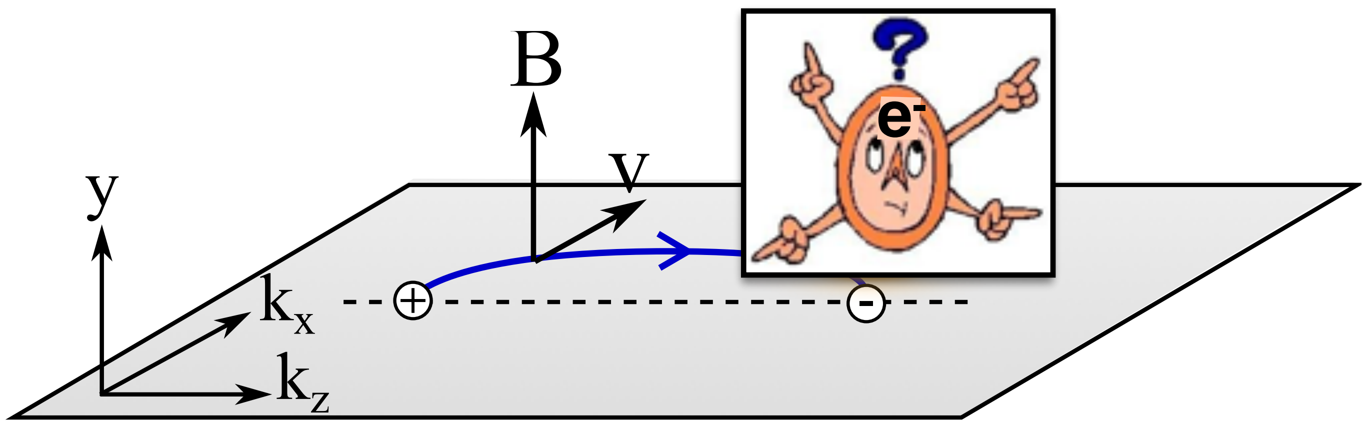
Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

The chirality conveyor belt



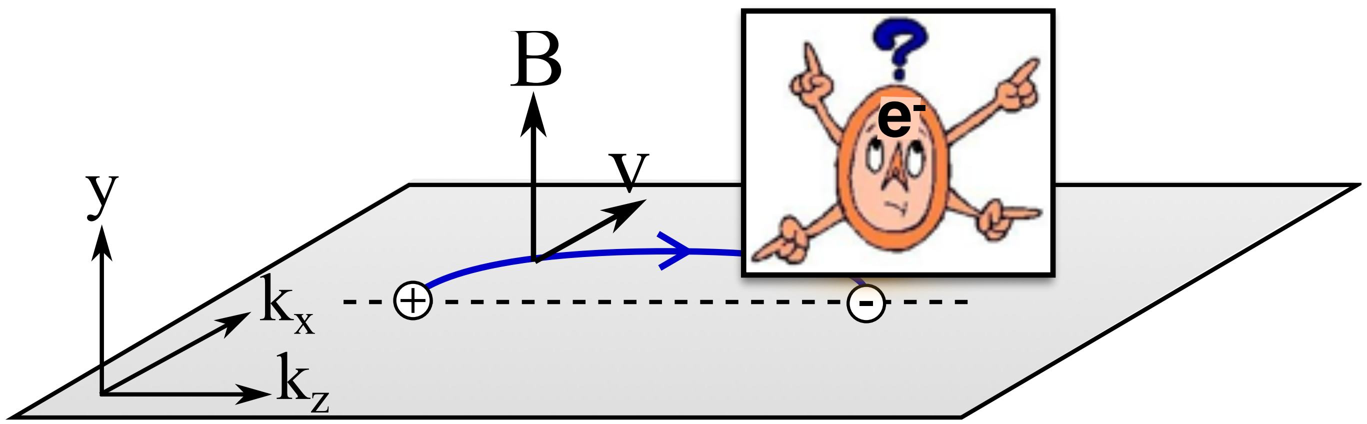
Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

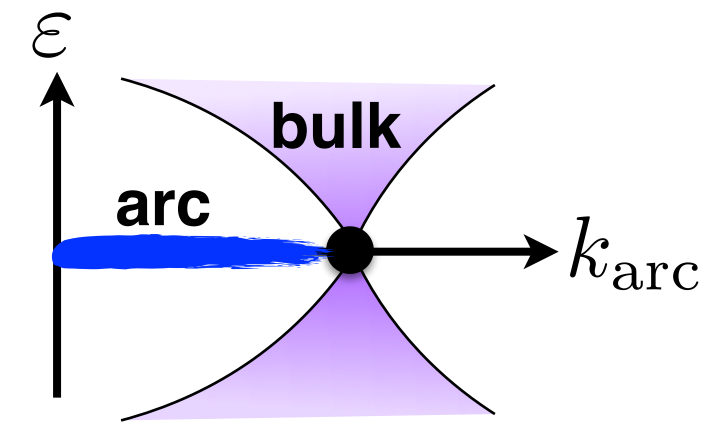
The chirality conveyor belt



Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

The chirality conveyor belt



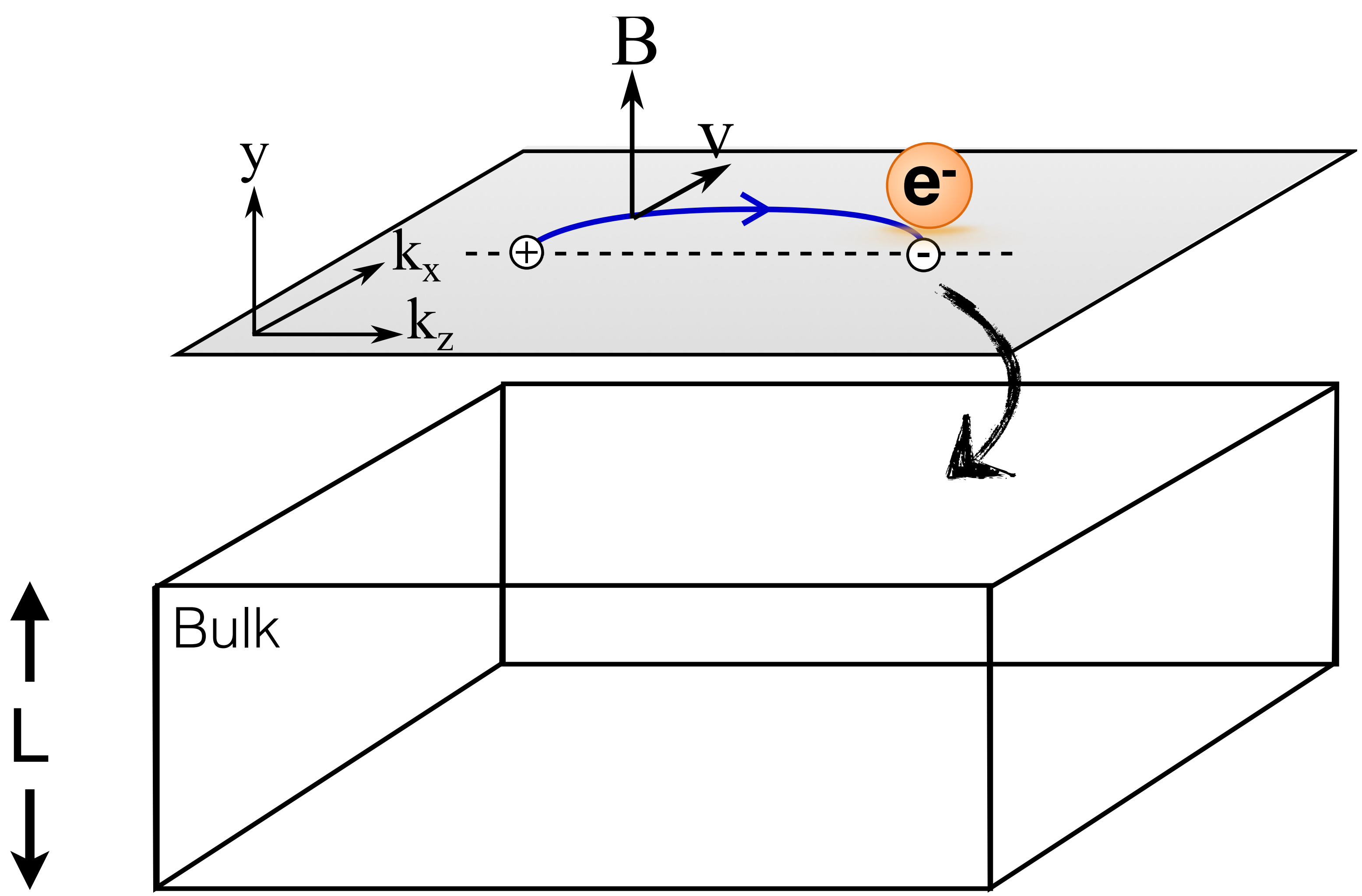


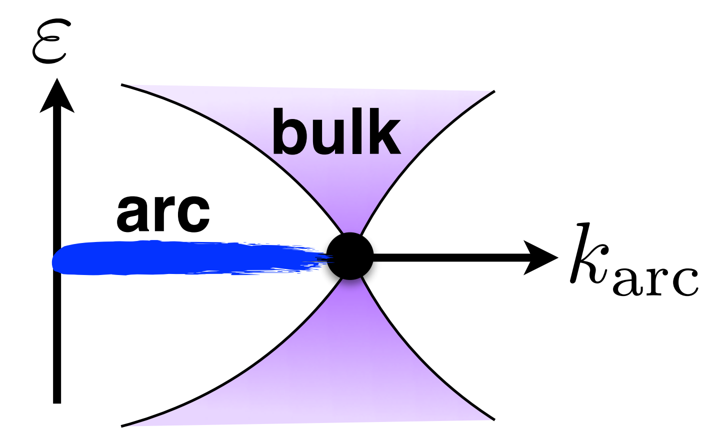
Surface-to-Bulk Transfer

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

The chirality conveyor belt

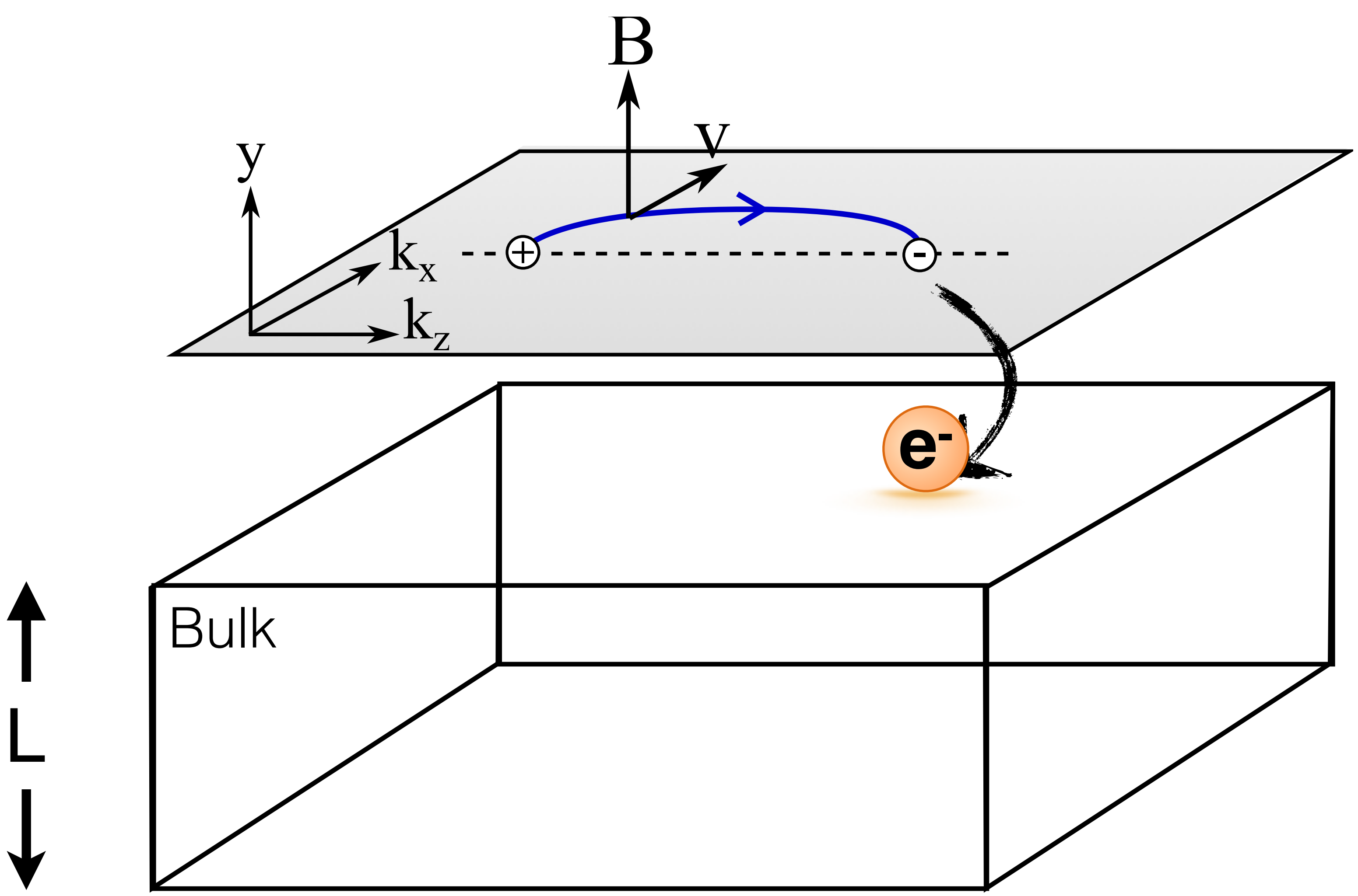
Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

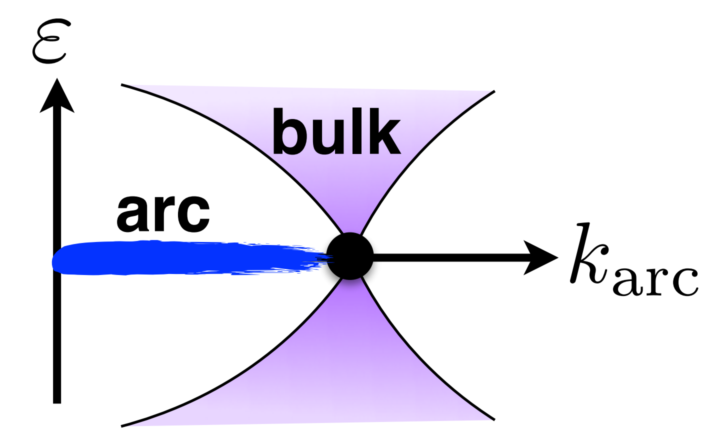




Surface-to-Bulk Transfer

The chirality conveyor belt

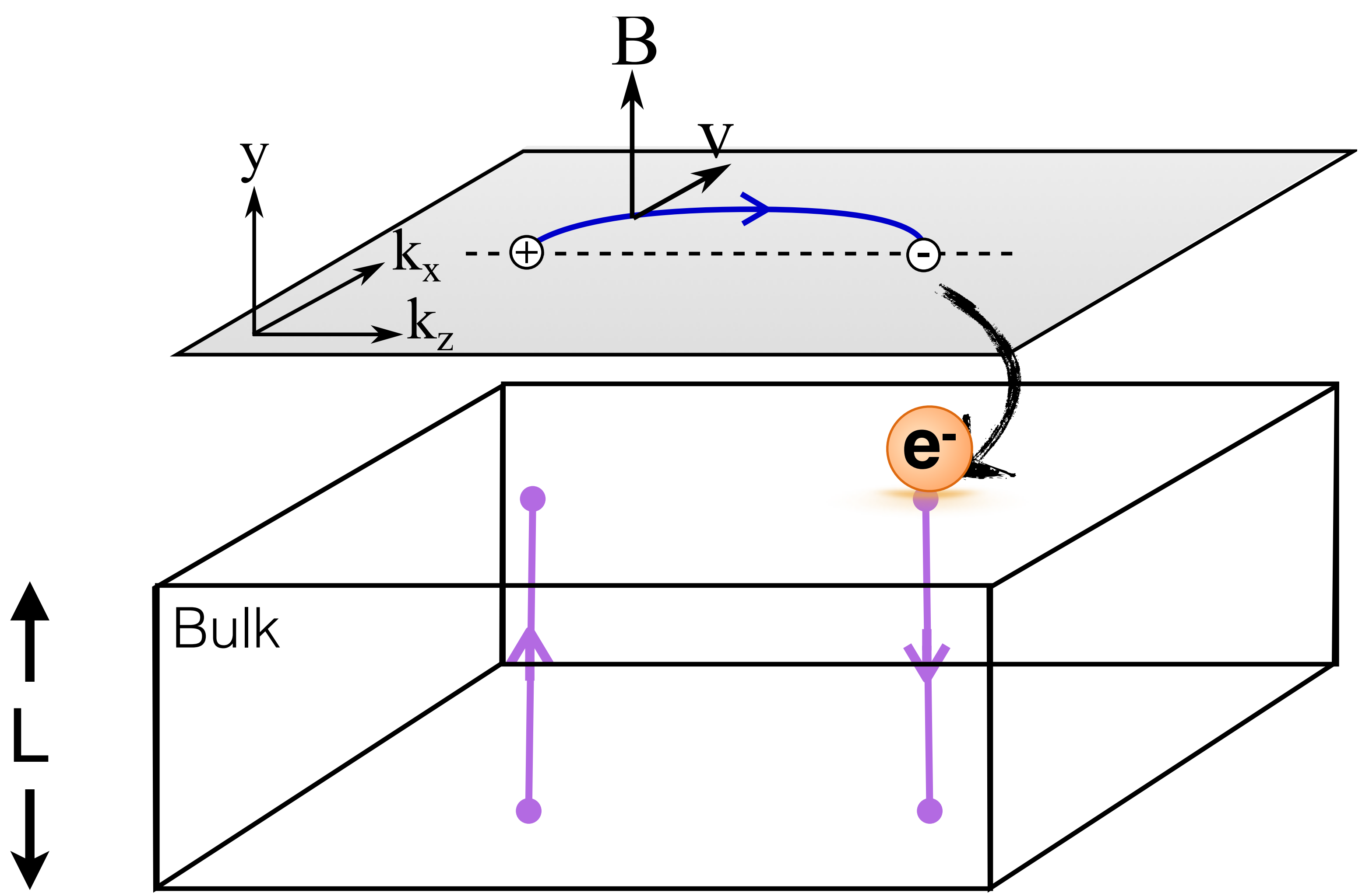


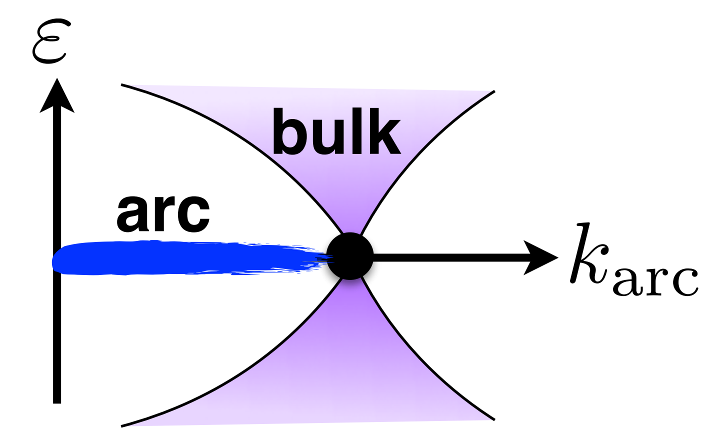


Surface-to-Bulk Transfer

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

The chirality conveyor belt



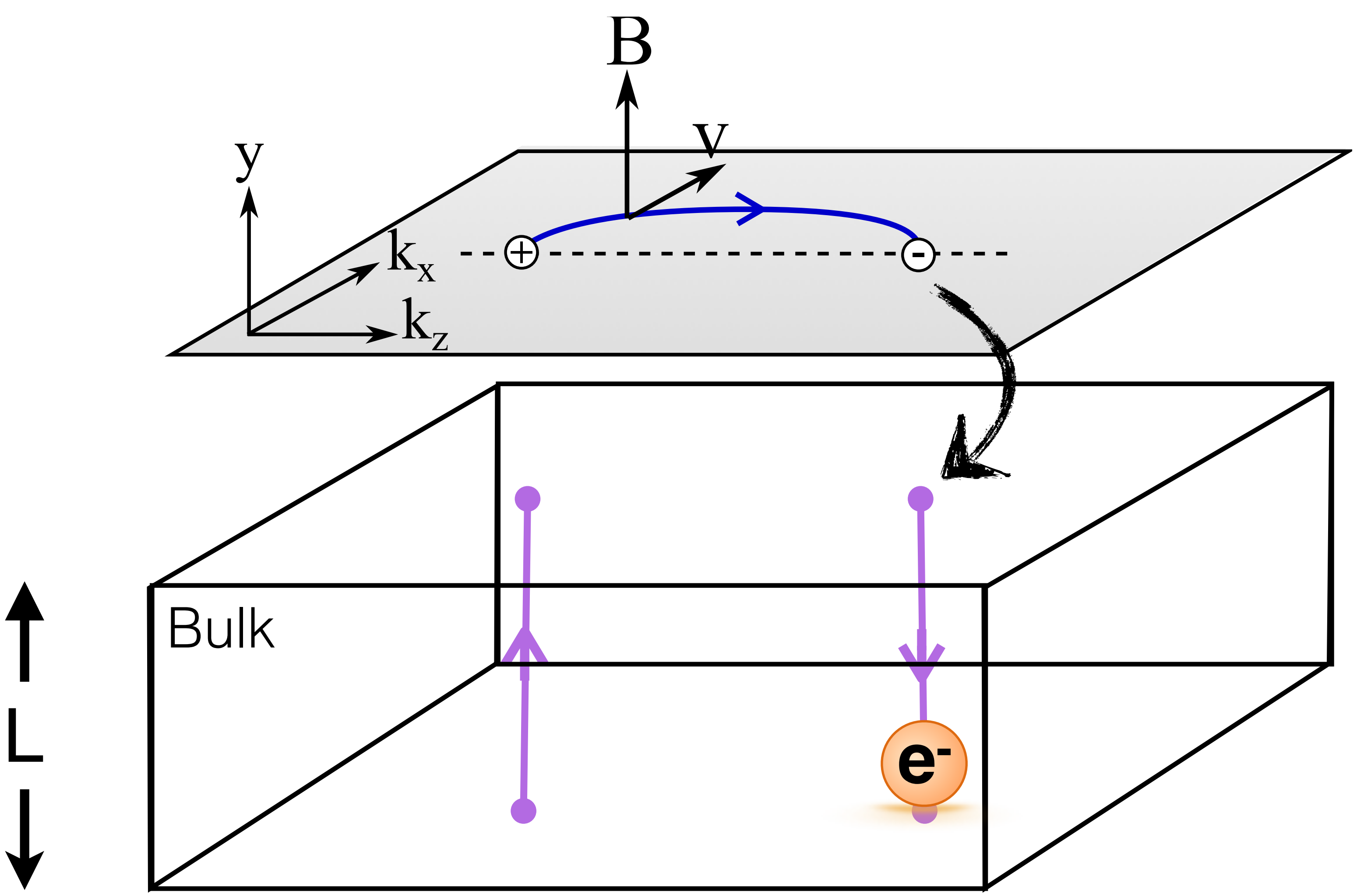


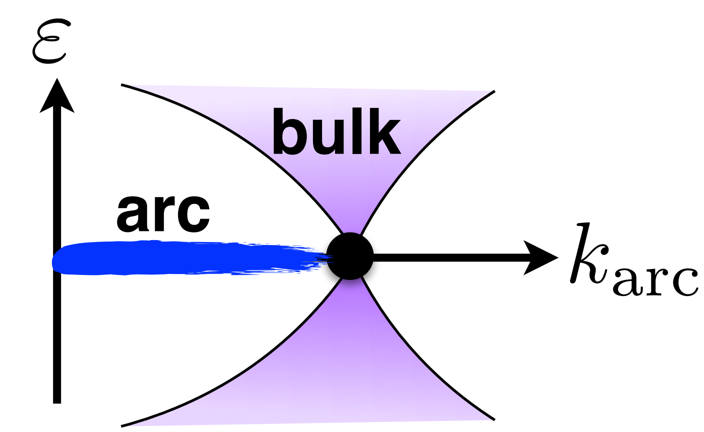
Surface-to-Bulk Transfer

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

The chirality conveyor belt

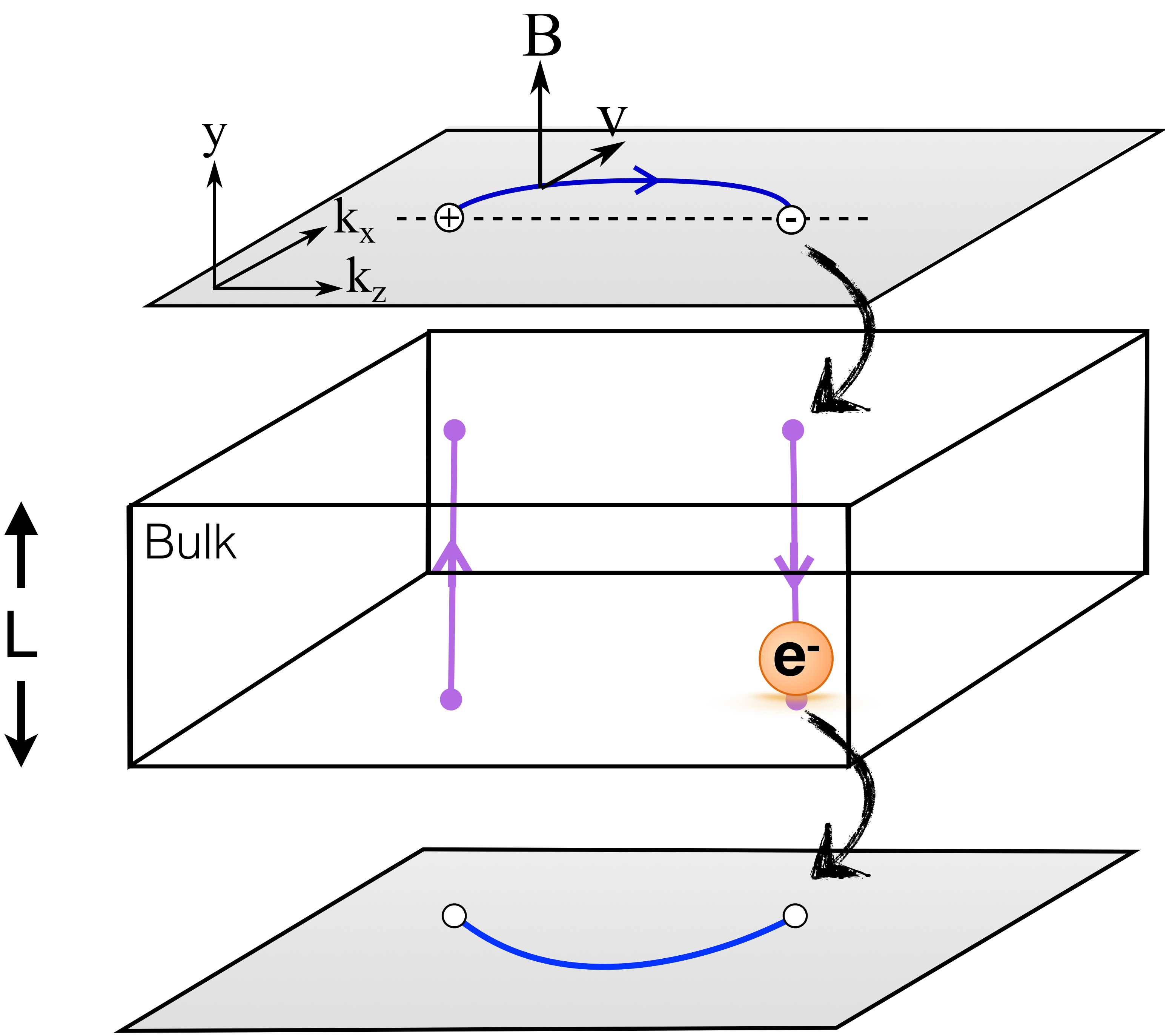
Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

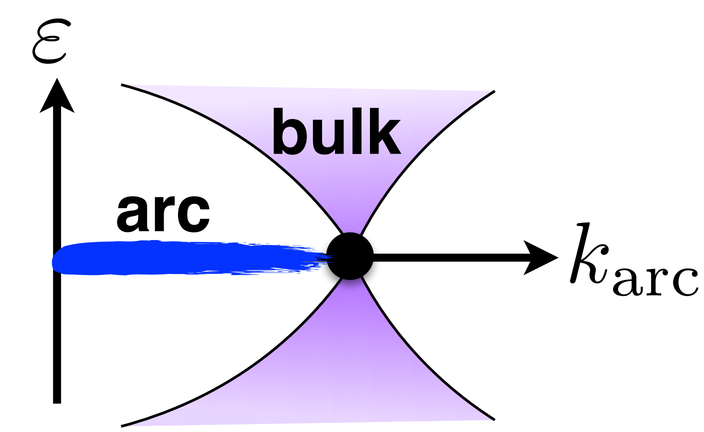




Surface-to-Bulk Transfer

The chirality conveyor belt

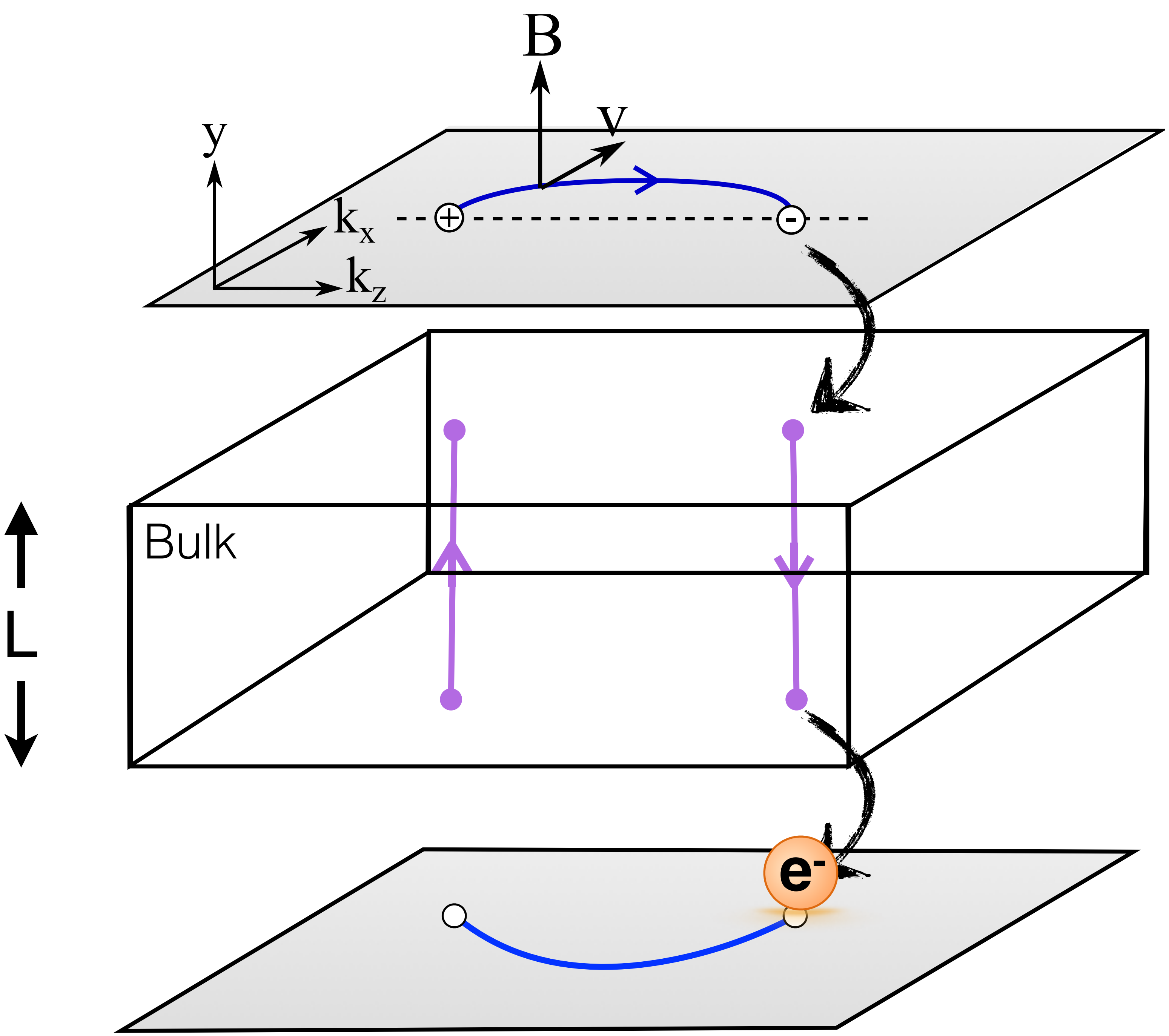


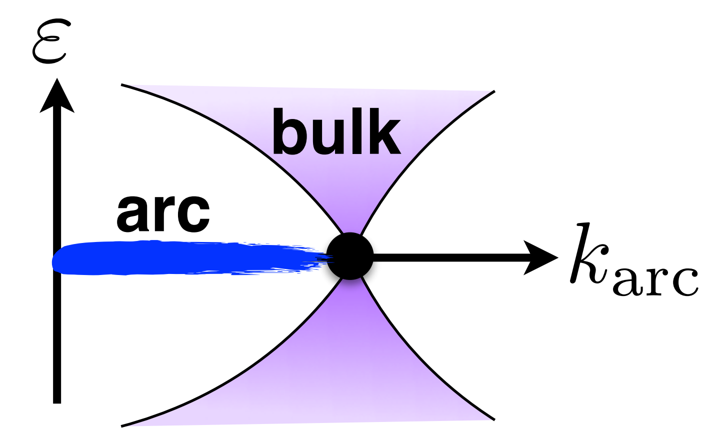


Surface-to-Bulk Transfer

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

The chirality conveyor belt



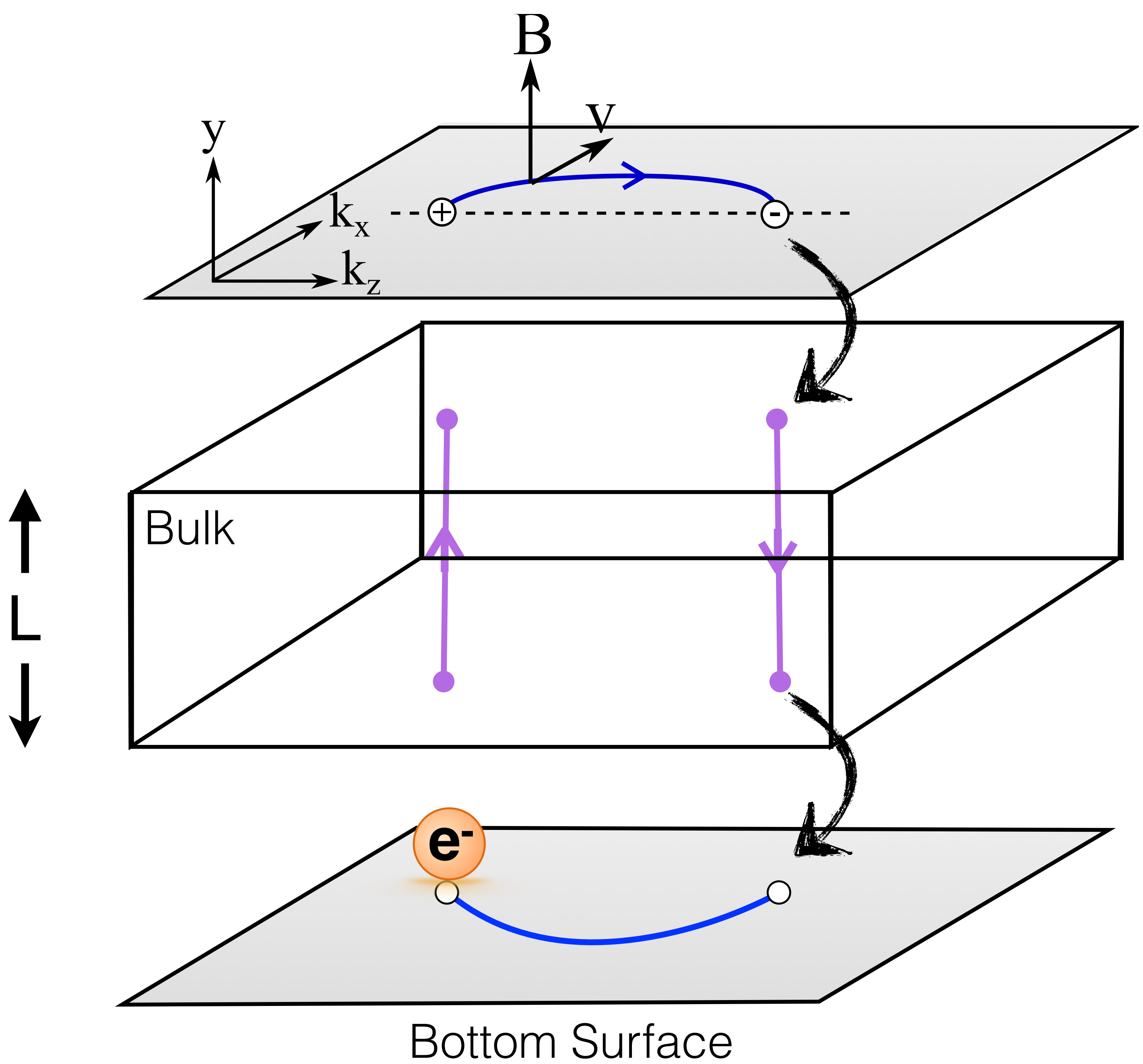


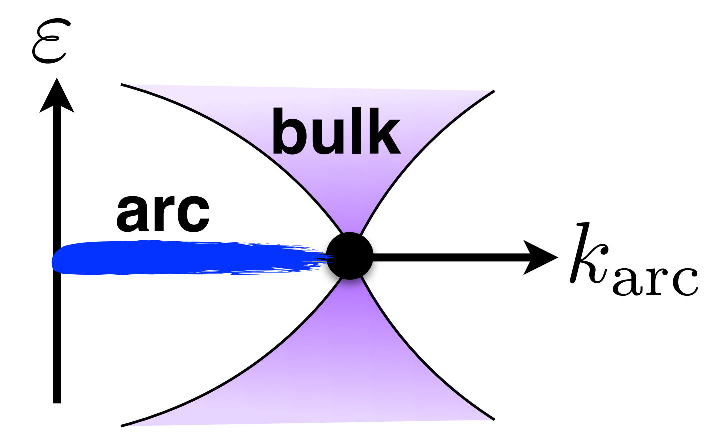
Surface-to-Bulk Transfer

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

The chirality conveyor belt

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

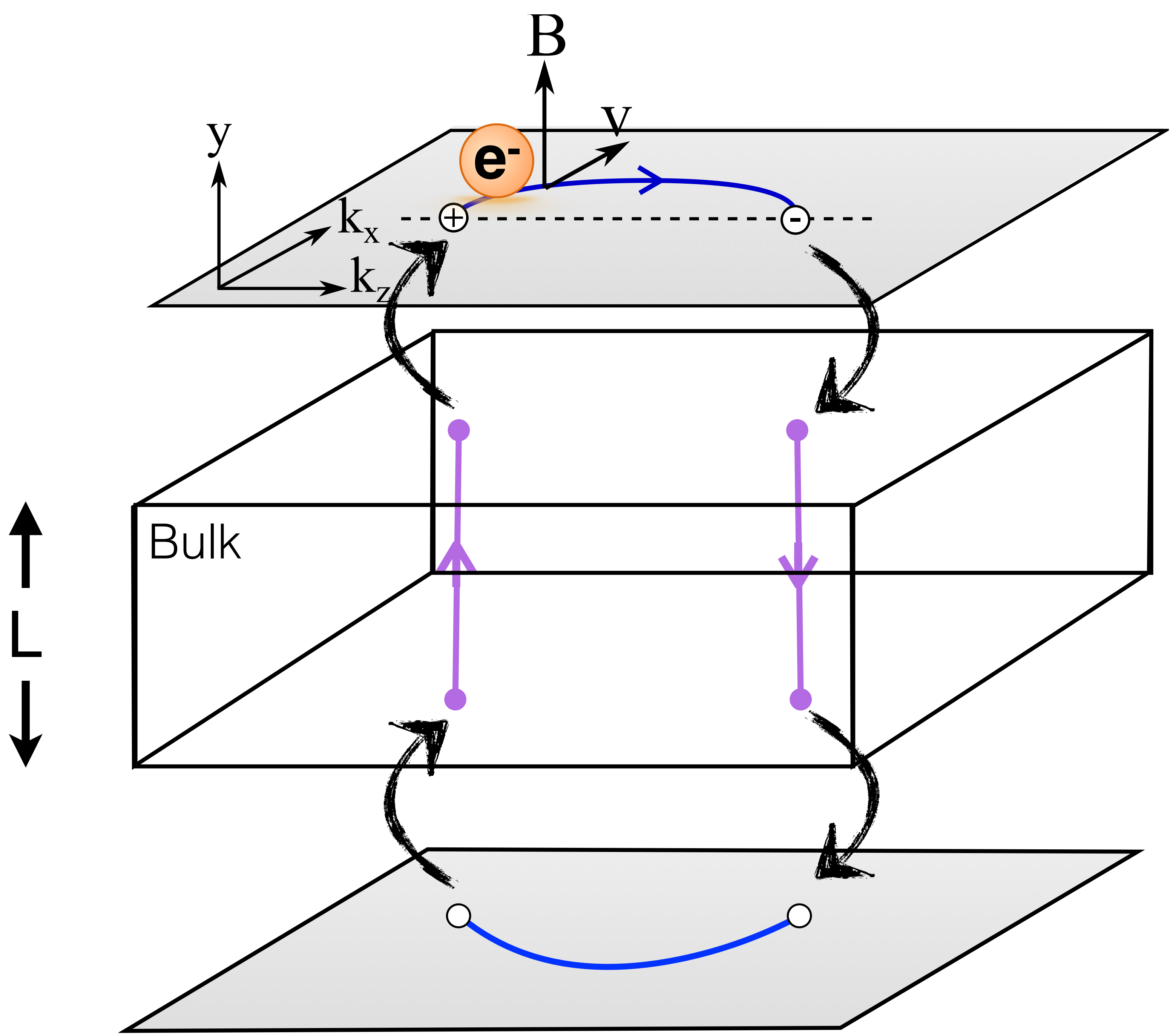




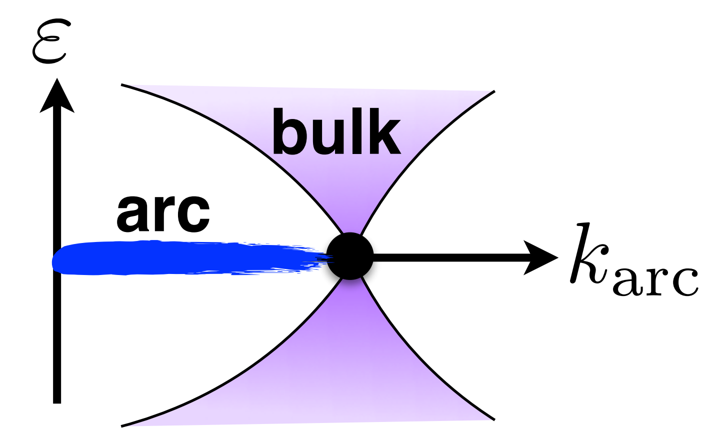
Surface-to-Bulk Transfer

The chirality conveyor belt

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)



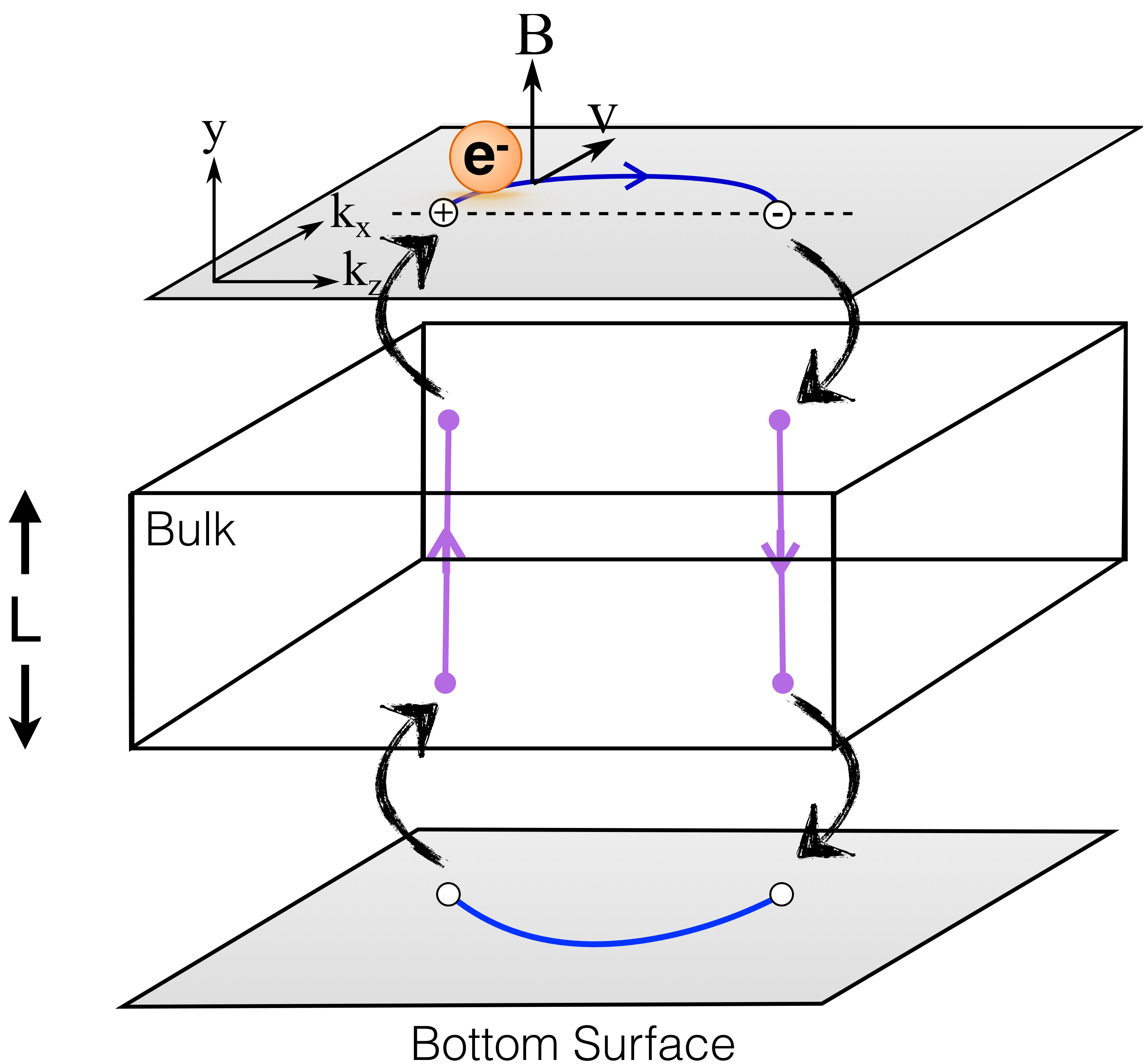
Bottom Surface

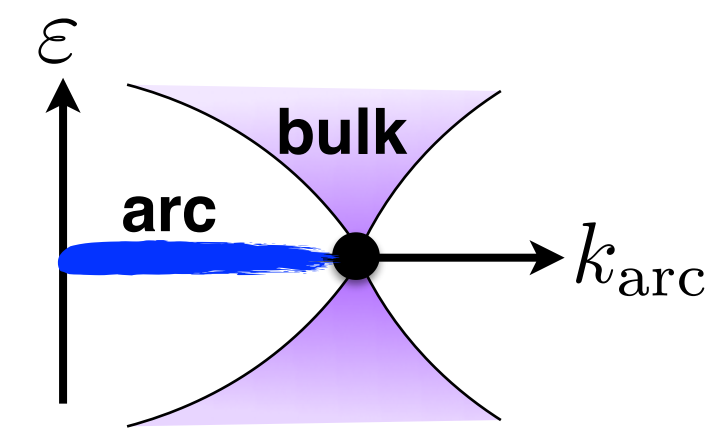


Surface-to-Bulk Transfer

The chirality conveyor belt

Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)





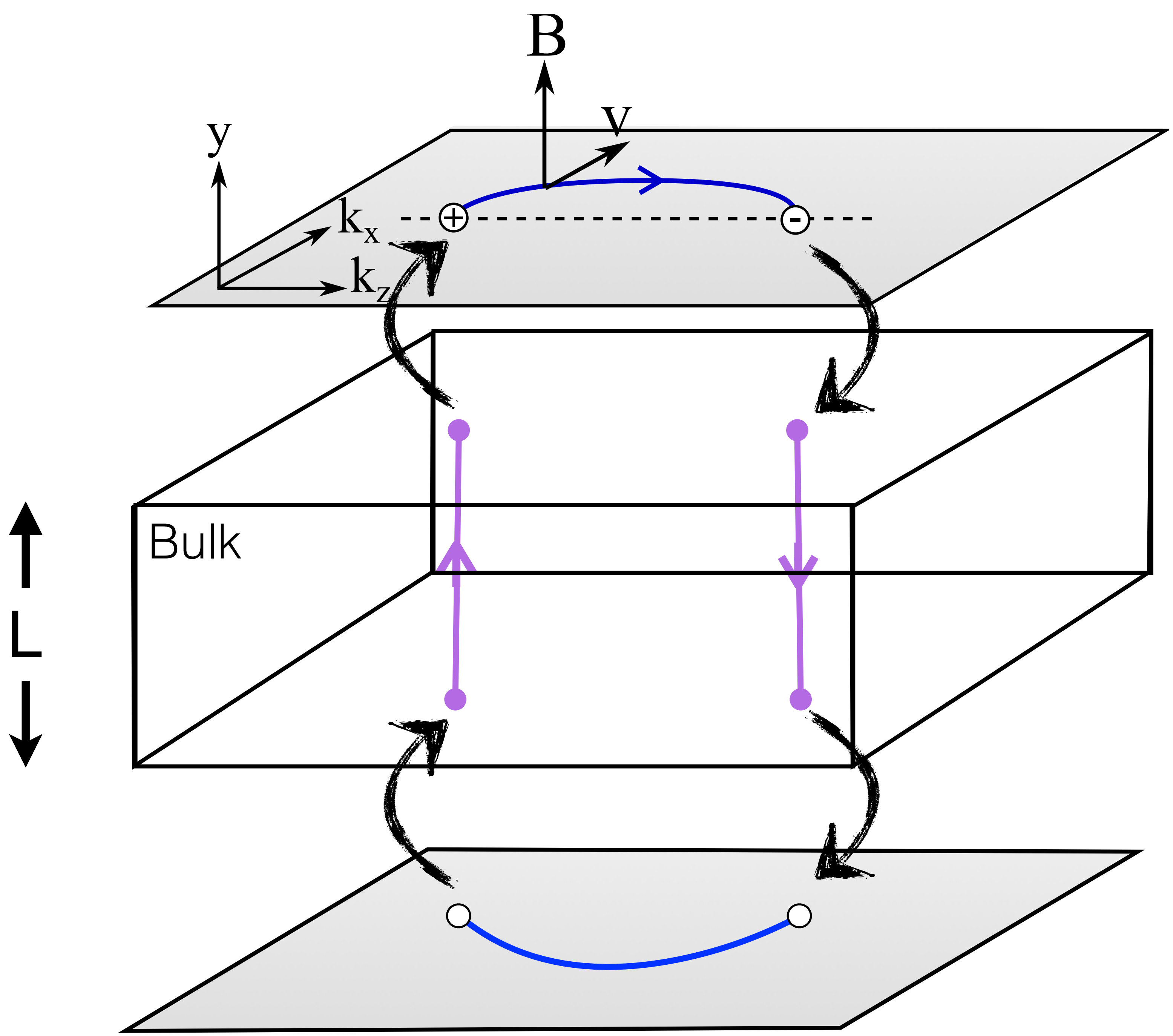
Surface-to-Bulk Transfer

$$t = 2\left(\frac{k_{\rm arc}}{evB} + \frac{L}{v}\right)$$

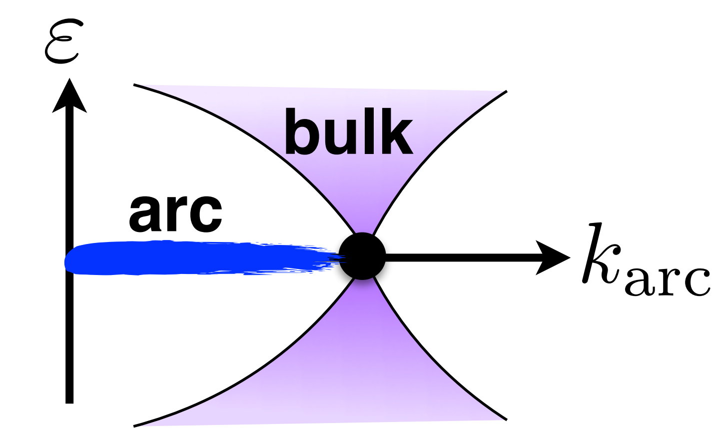
Arc **Bulk**

Weyl orbits

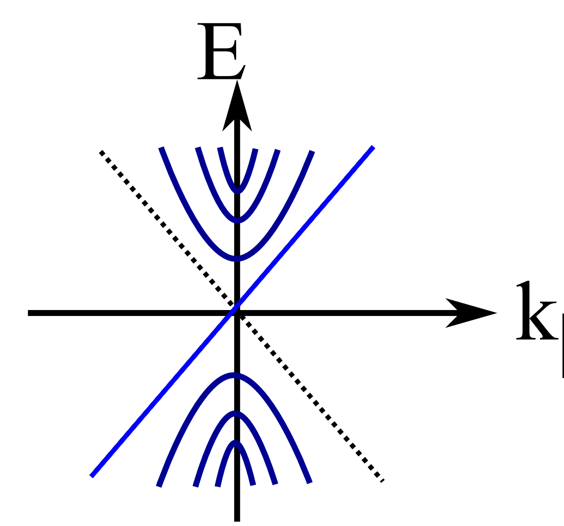
Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)



Bottom Surface

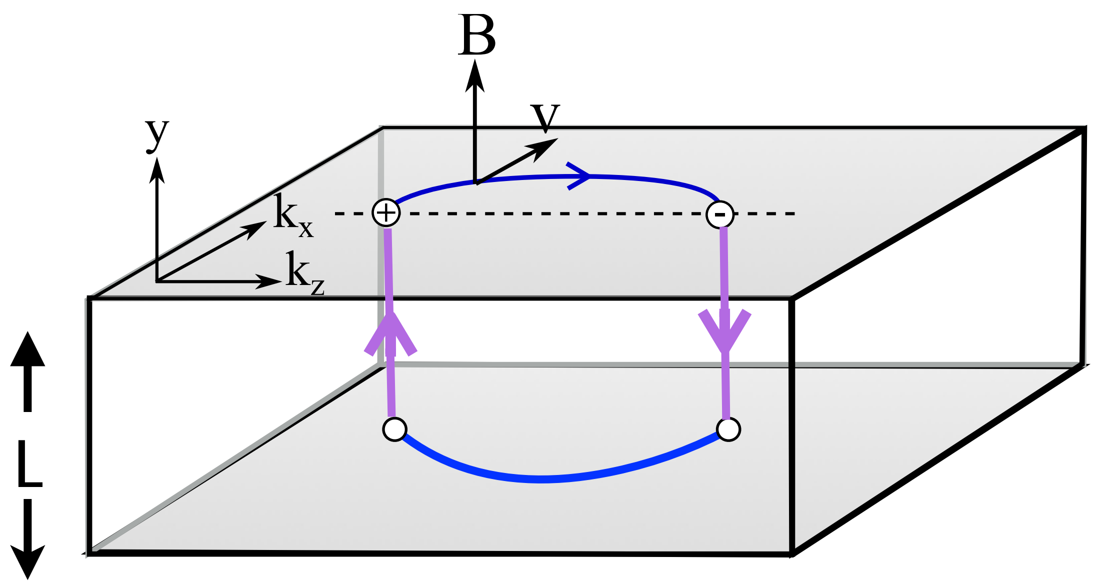


Surface-to-Bulk Transfer

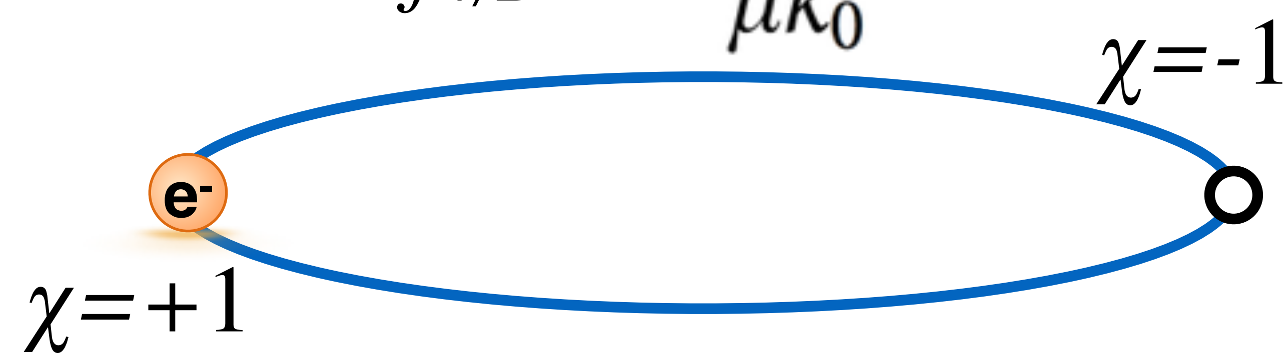


Bulk Landau Levels

Some remarks



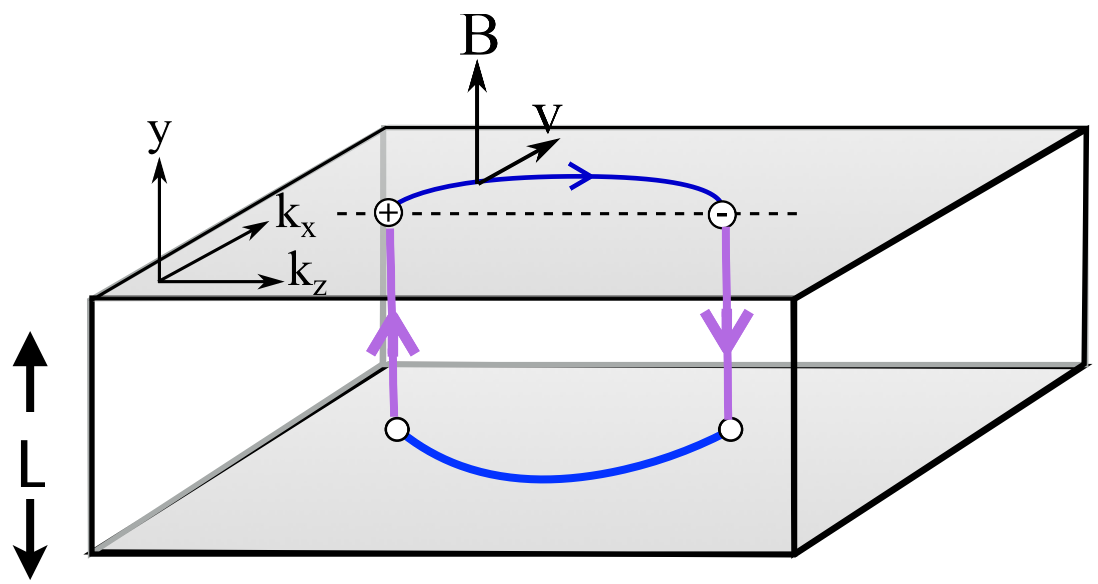
$$f_{1/B} \approx \frac{e\pi v}{\mu k_0}$$

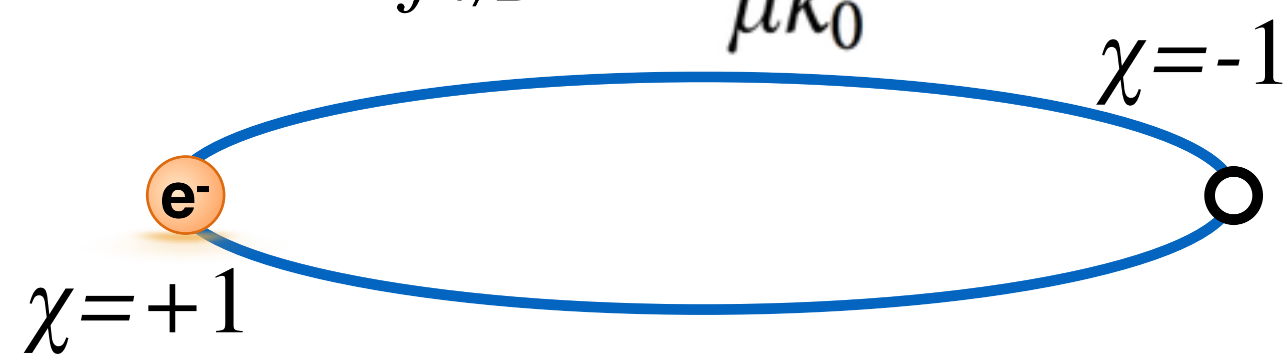


- The cyclotron “Weyl” orbit involves a real space and k-space path.
- Real space trajectory encloses no flux (Lorentz force free path).
- From a quantum oscillatory point of view, it looks a lot like a 2D orbit with area A_k, or equivalently frequency $f_{1/B}$

Some remarks

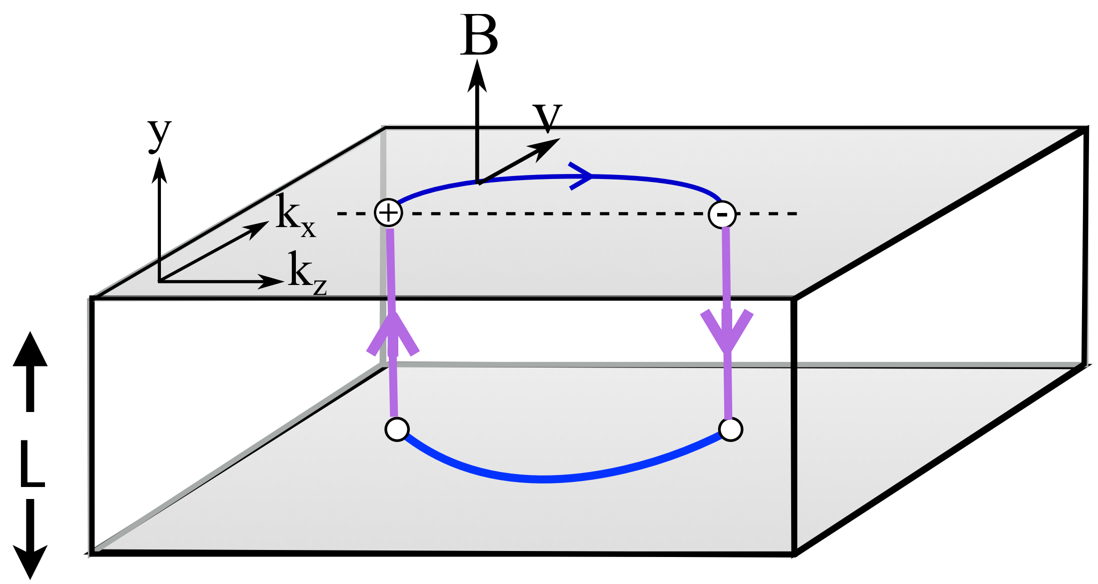
$$f_{1/B} \approx \frac{e\pi v}{\mu k_0}$$

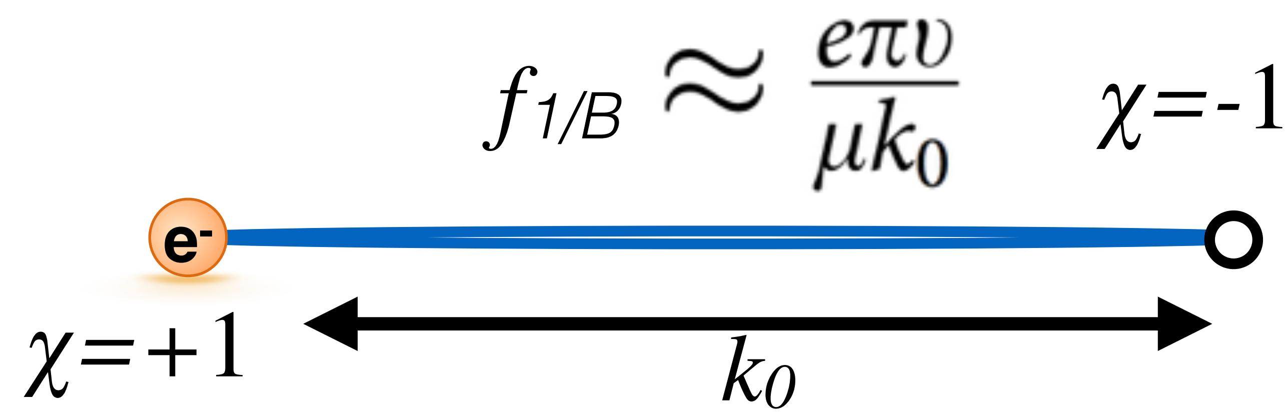




- The cyclotron “Weyl” orbit involves a real space and k-space path.
- Real space trajectory encloses no flux (Lorentz force free path).
- From a quantum oscillatory point of view, it looks a lot like a 2D orbit with area A_k, or equivalently frequency $f_{1/B}$

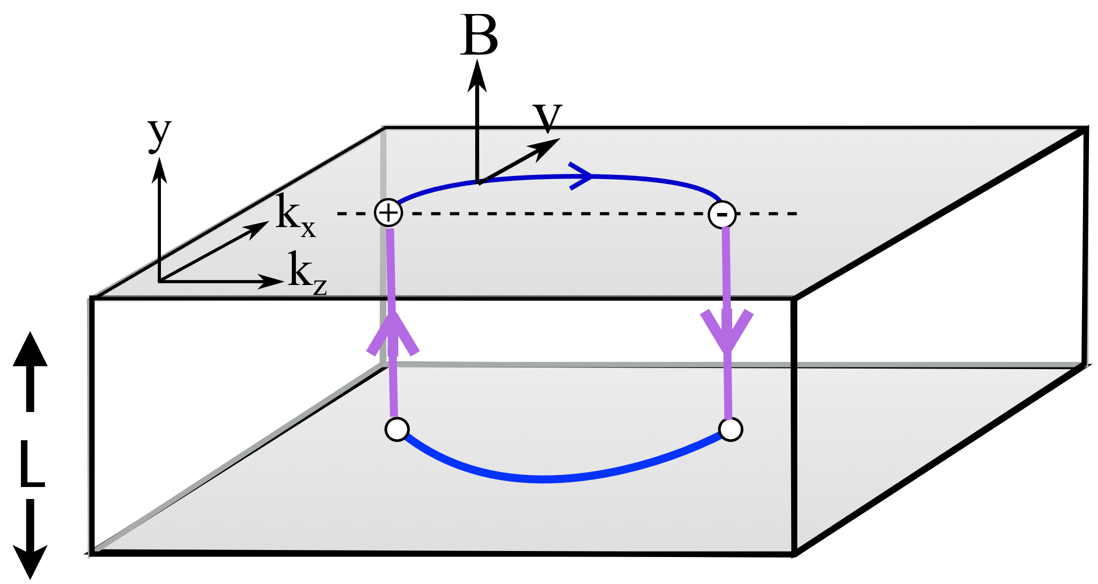
Some remarks

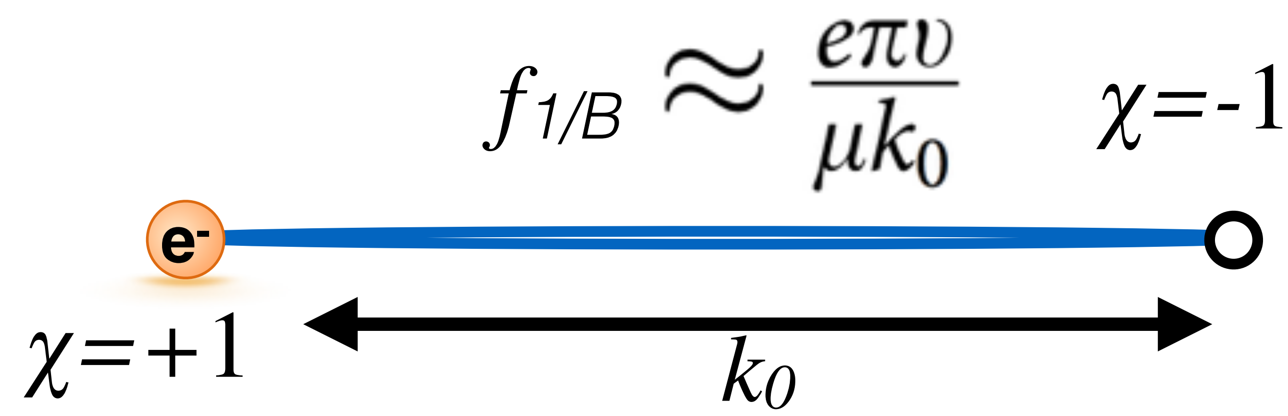




- The cyclotron “Weyl” orbit involves a real space and k-space path.
- Real space trajectory encloses no flux (Lorentz force free path).
- From a quantum oscillatory point of view, it looks a lot like a 2D orbit with area A_k, or equivalently frequency $f_{1/B}$

Some remarks



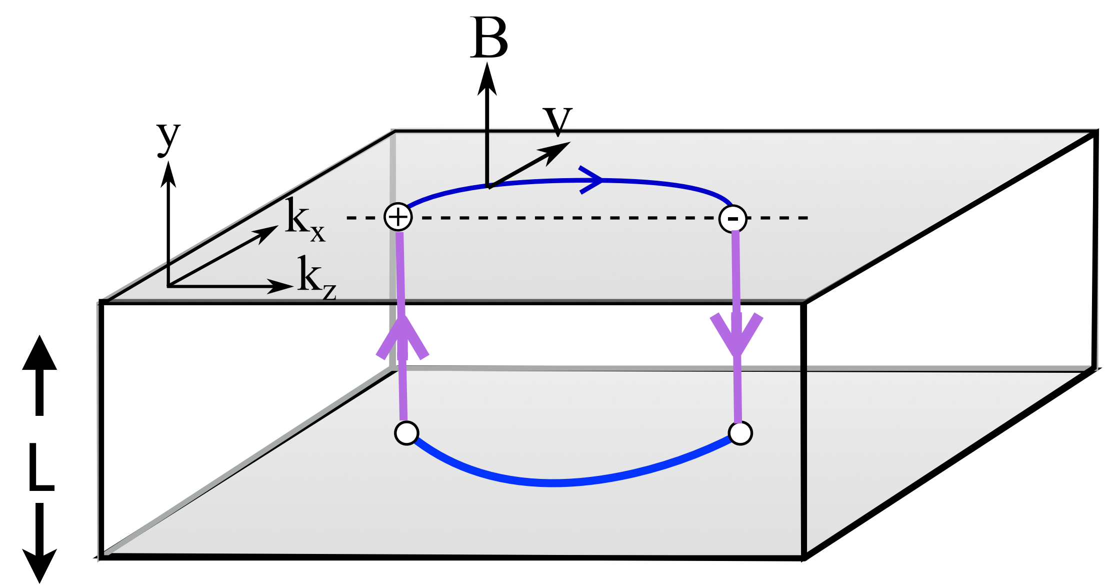


- The cyclotron “Weyl” orbit involves a real space and k-space path.
- Real space trajectory encloses no flux (Lorentz force free path).
- From a quantum oscillatory point of view, it looks a lot like a 2D orbit with area A_k, or equivalently frequency $f_{1/B}$

Distinguishing features

Quantization of semiclassical orbits

$$\varepsilon_c t_c = (n + \gamma) 2\pi\hbar$$



Quantum oscillations $\qquad \frac{1}{B_n} = \frac{2\pi n}{f_{1/B}} - \frac{e}{k_{\text{arc}}} L$

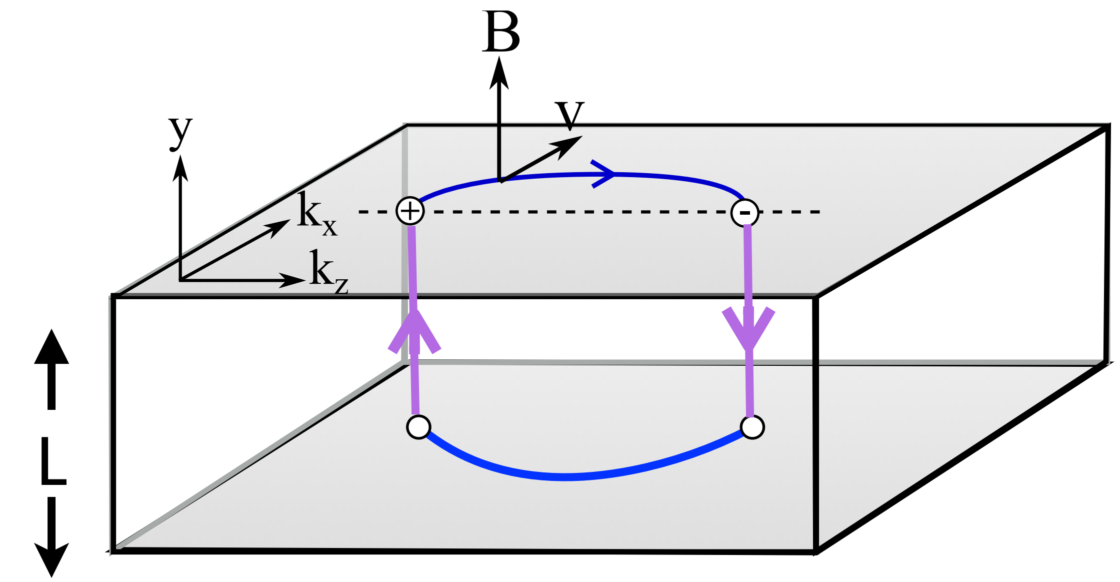
$f_{1/B}$

Distinguishing features

Quantization of semiclassical orbits

$$\varepsilon_n \approx \frac{2\pi n + \gamma}{t} \approx \frac{2\pi n + \gamma}{2\left(\frac{k_{\text{arc}}}{evB} + \frac{L}{v}\right)}$$

Arc **Bulk**



Quantum oscillations $\frac{1}{B_n} = \frac{2\pi n}{f_{1/B}} - \frac{e}{k_{\text{arc}}} L$

$B^* \approx \frac{k_{\text{arc}}}{L}$

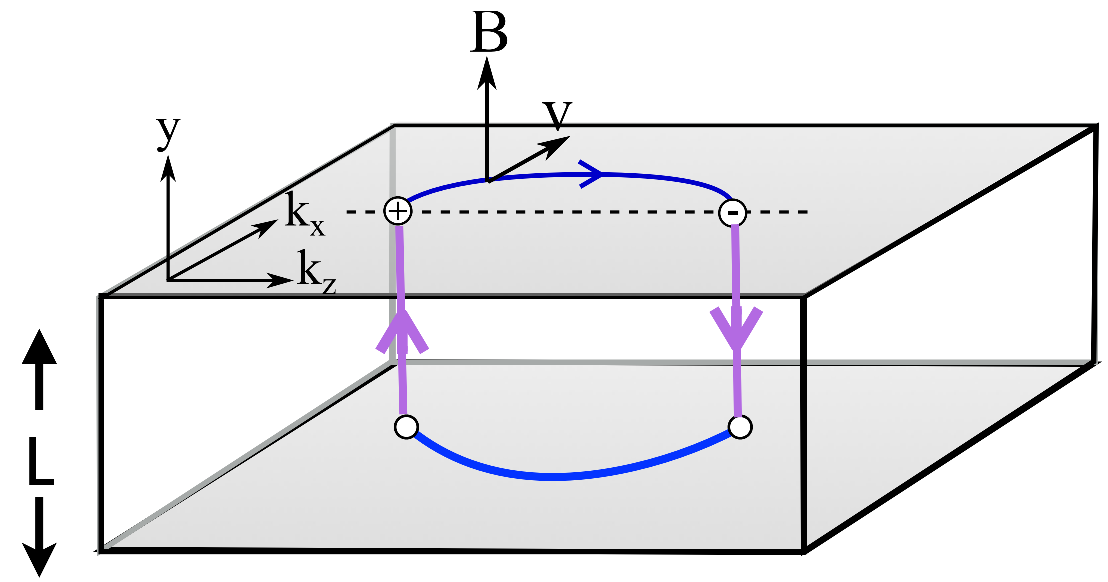
$f_{1/B}$

Distinguishing features

Quantization of semiclassical orbits

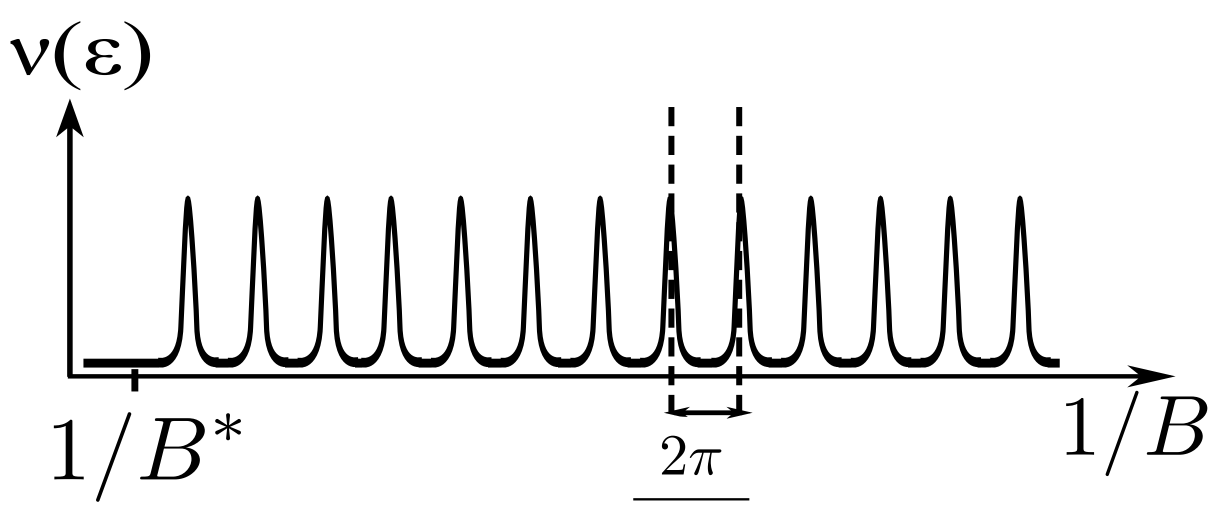
$$\varepsilon_n \approx \frac{2\pi n + \gamma}{t} \approx \frac{2\pi n + \gamma}{2\left(\frac{k_{\text{arc}}}{evB} + \frac{L}{v}\right)}$$

Arc **Bulk**

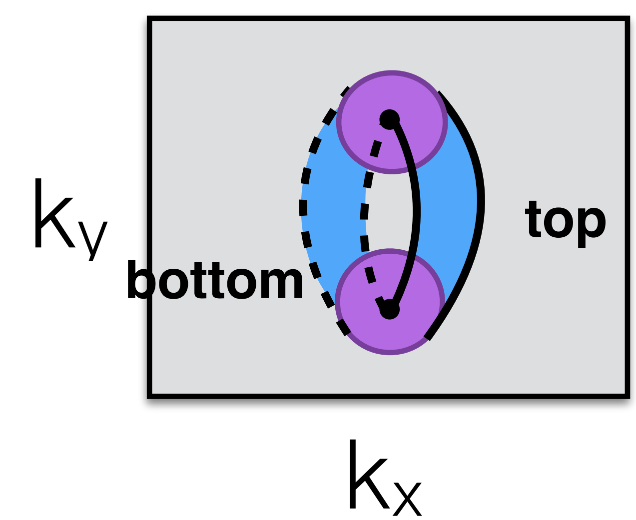


Quantum oscillations $$\frac{1}{B_n} = \frac{2\pi n}{f_{1/B}} - \frac{e}{k_{\text{arc}}} L$$

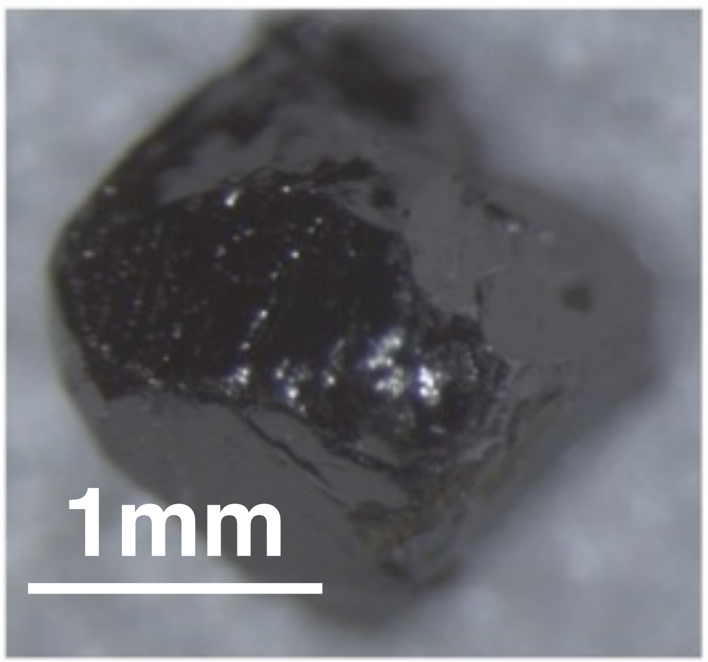
$f_{1/B}$



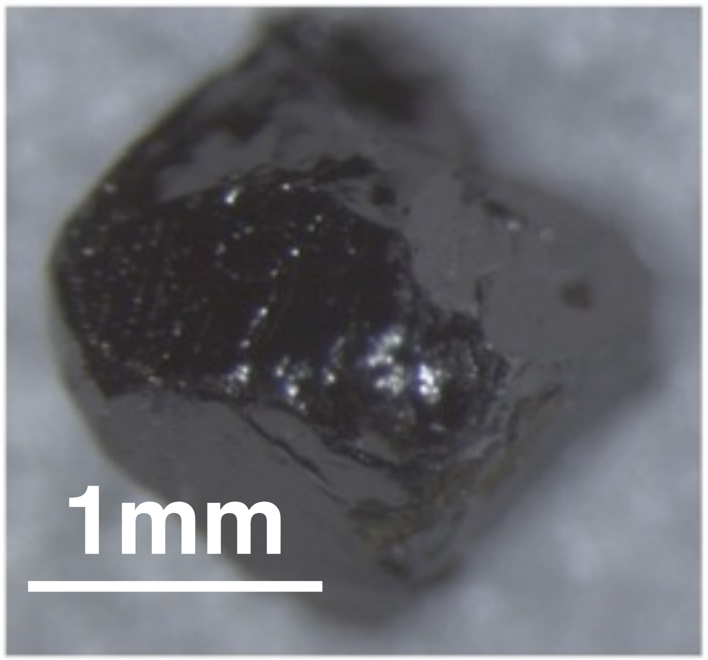
$$B^* \approx \frac{k_{\text{arc}}}{L}$$



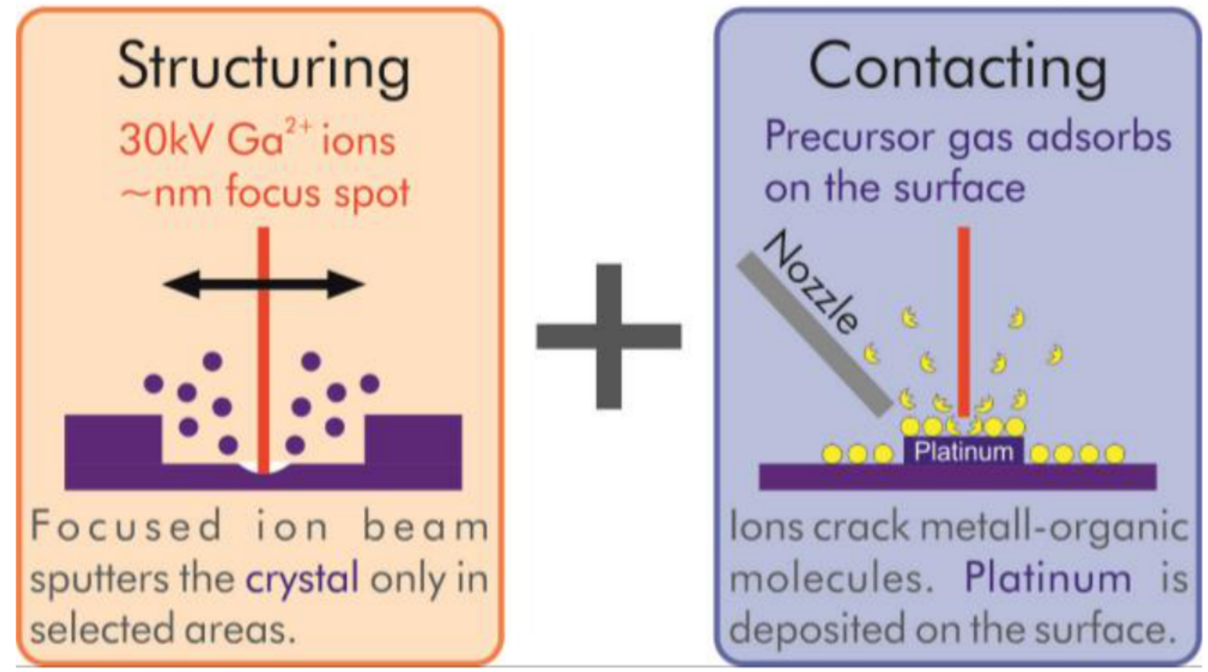
Focused Ion Beam microstructuring Cd_3As_2



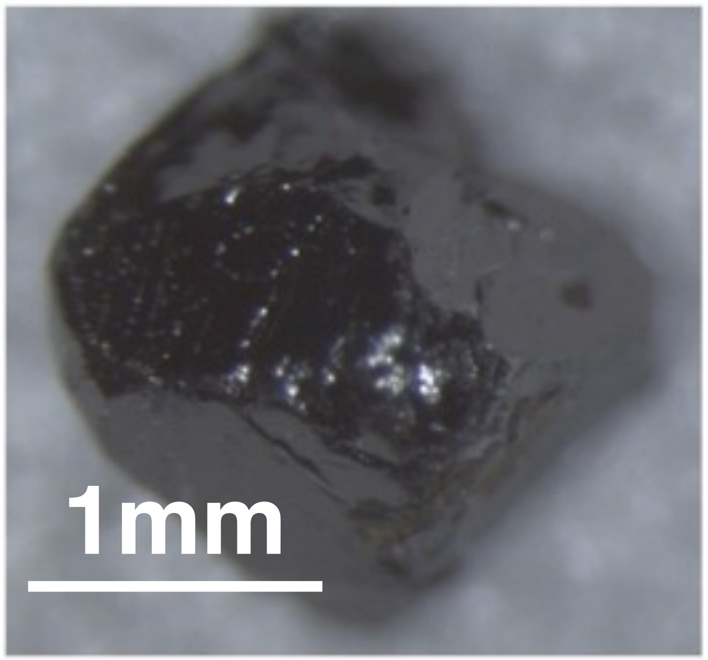
Focused Ion Beam microstructuring Cd_3As_2



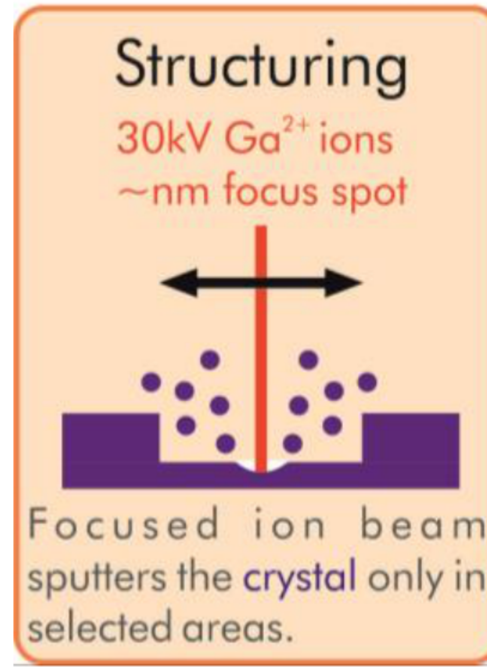
+



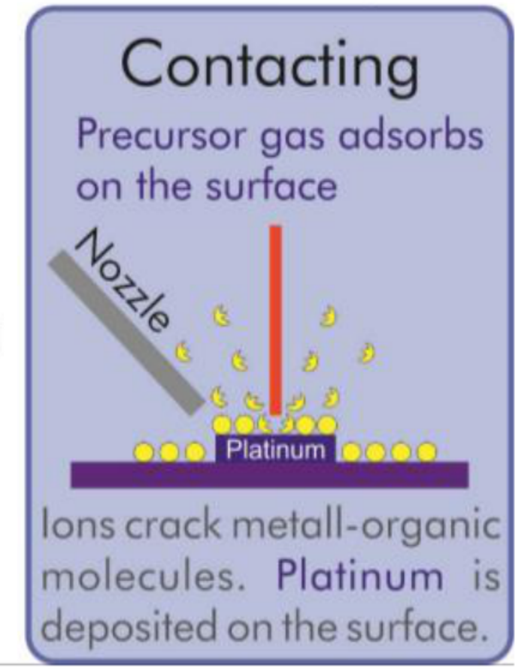
Focused Ion Beam microstructuring Cd_3As_2



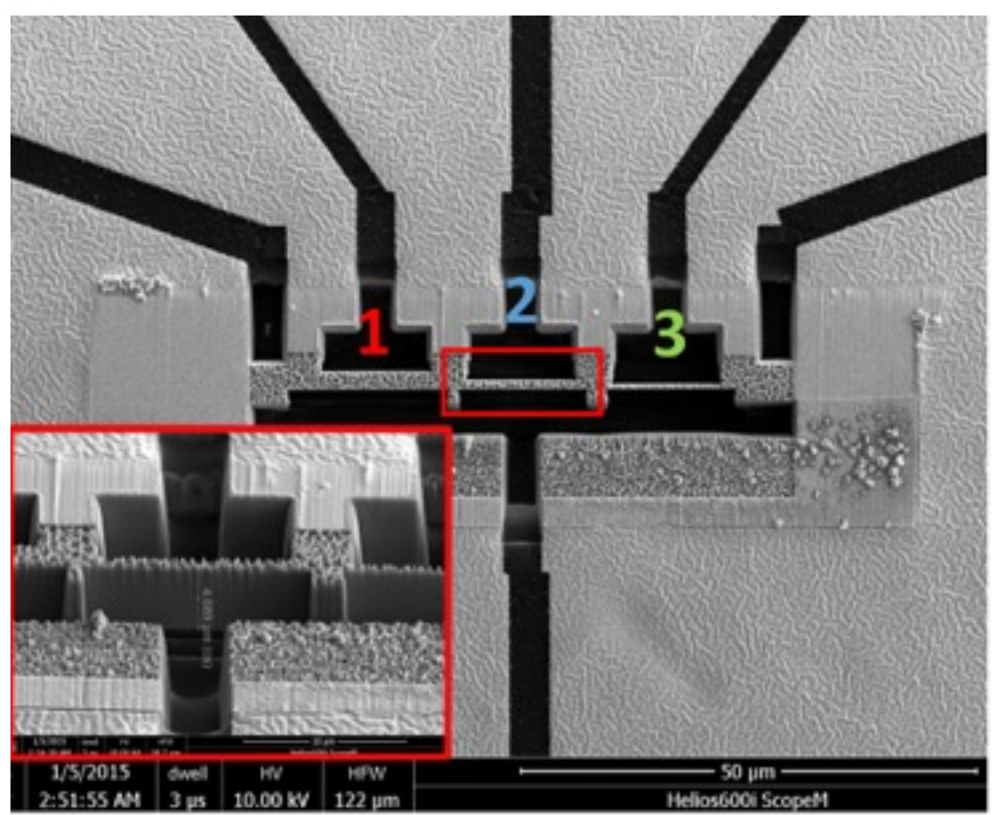
+



+

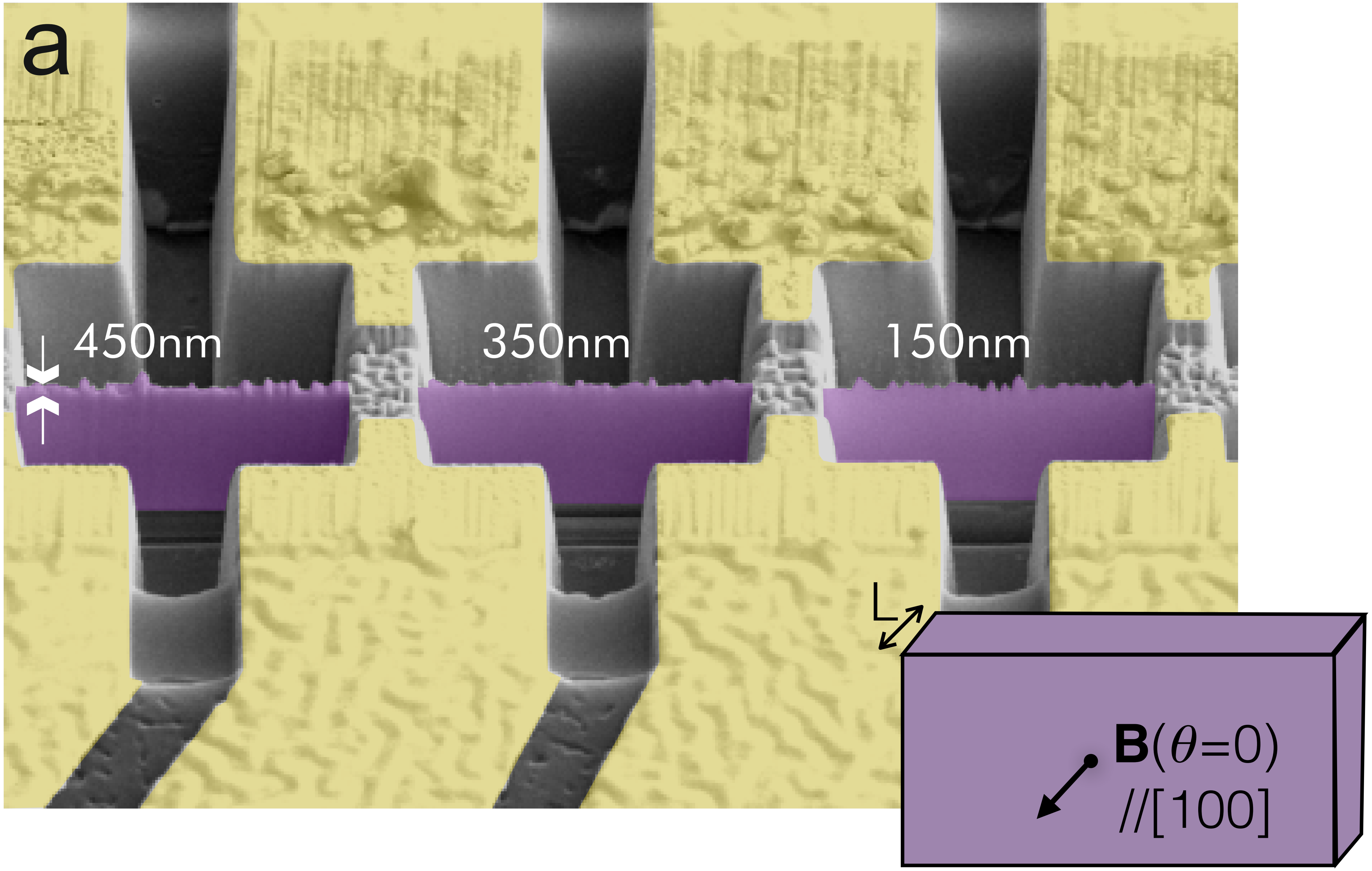


=

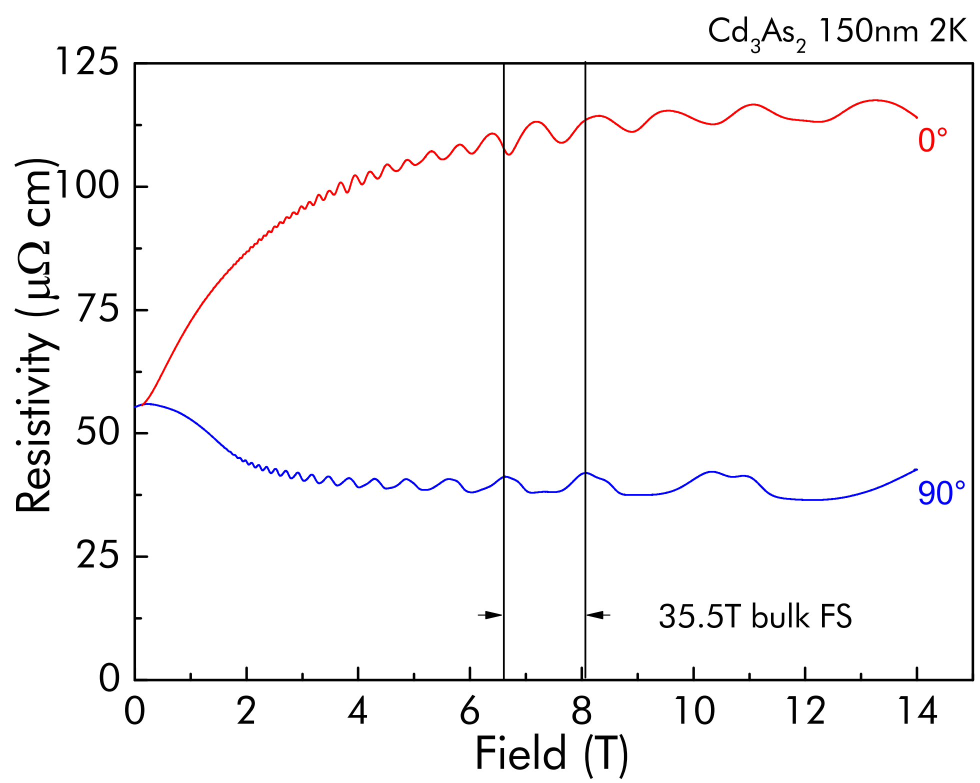


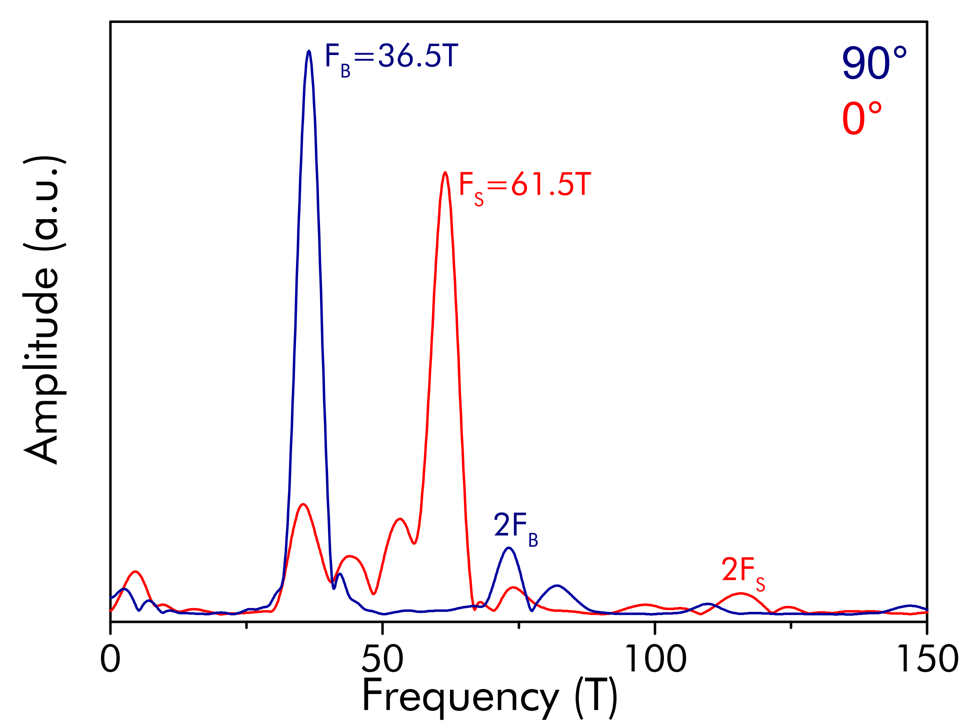
Thickness-dependent quantum oscillatory study in Cd_3As_2

Philip J. W. Moll, JGA et al. arXiv:1505.02817

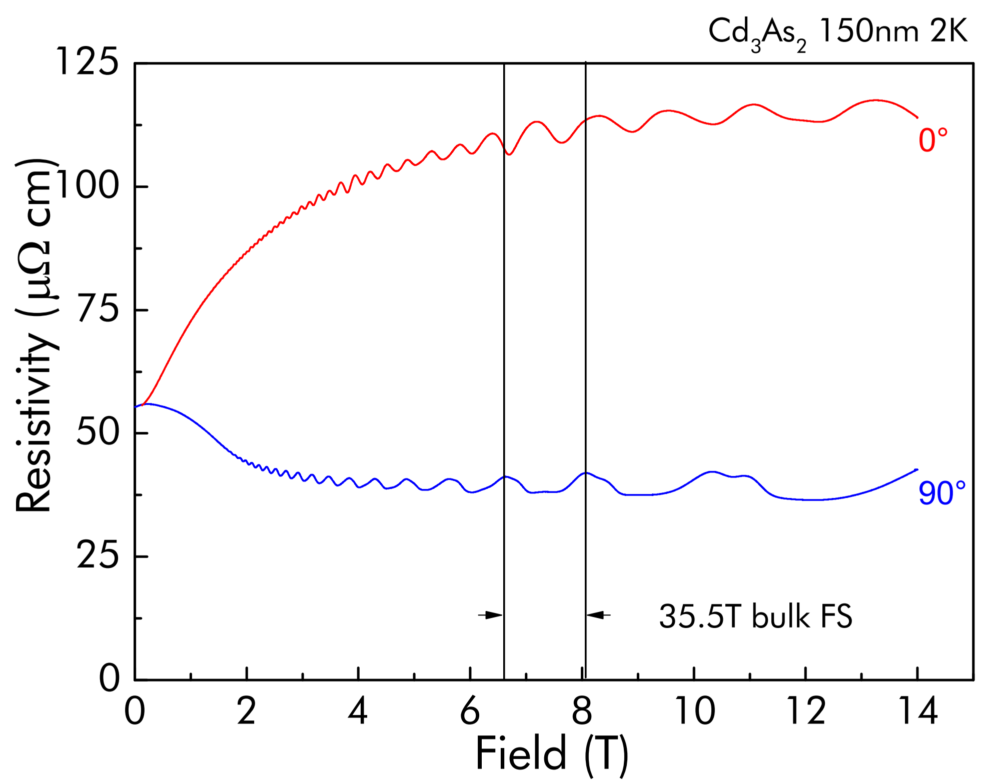


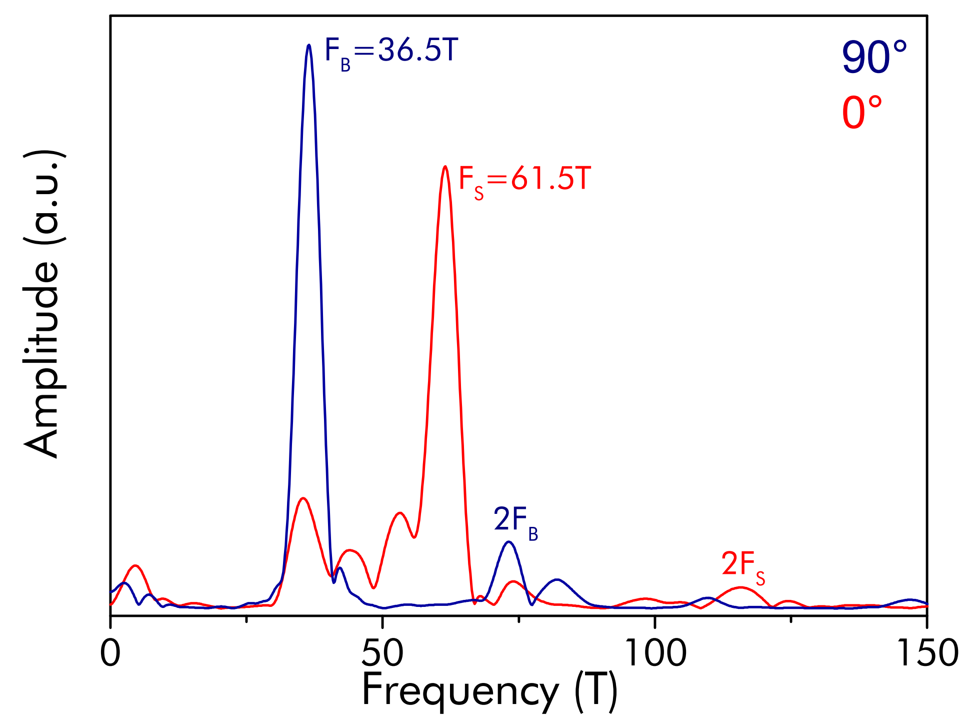
New quantum oscillatory frequency at ~60T (k_0~$0.08Å^{-1}$)





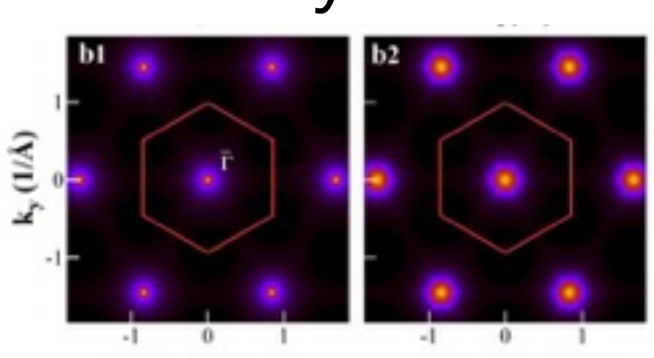
New quantum oscillatory frequency at ~60T (k_0~$0.08Å^{-1}$)



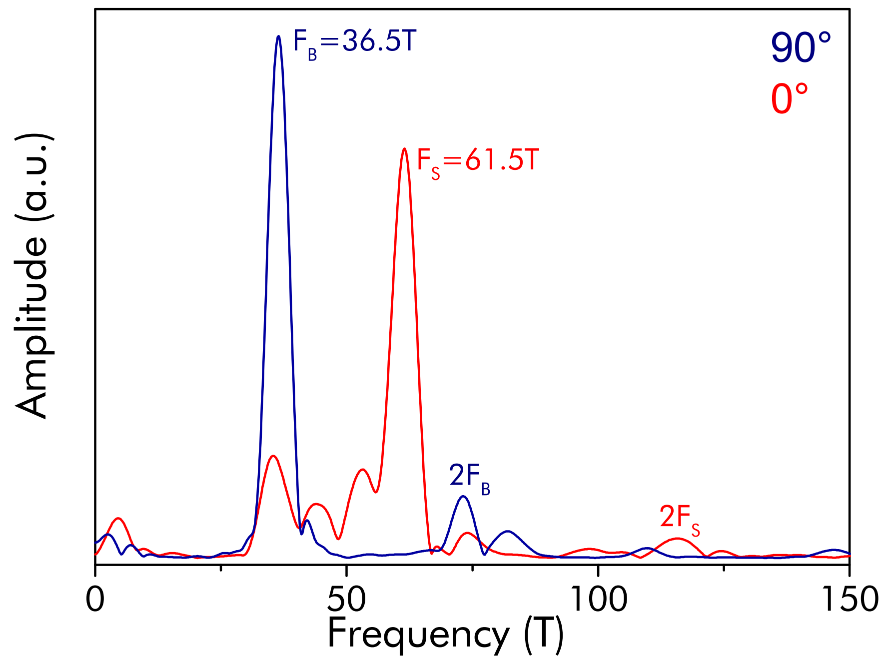


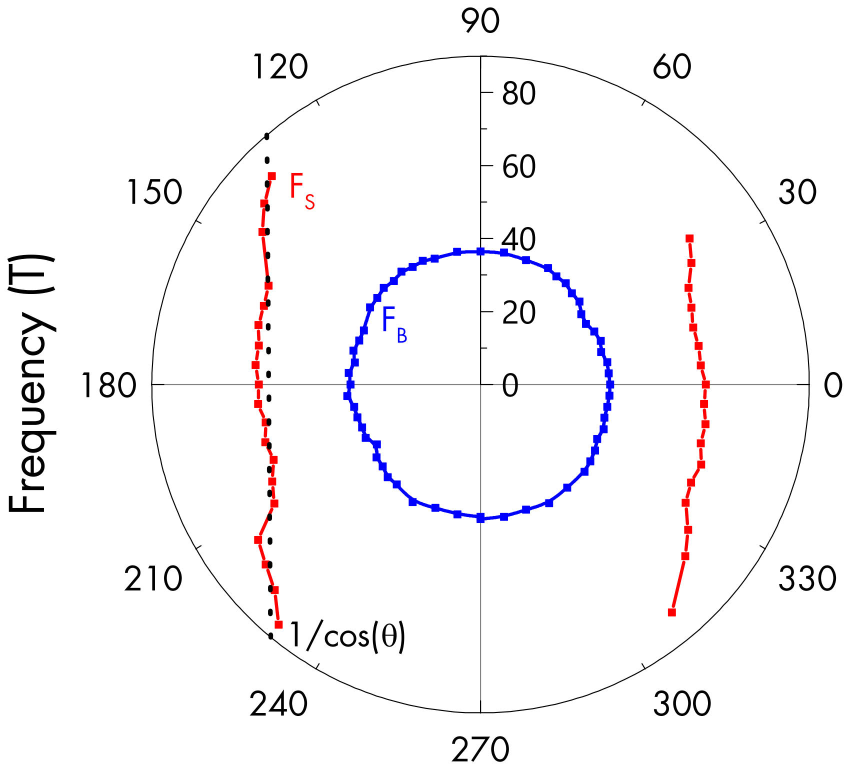
Note this is approximately the k_0 measured by ARPES

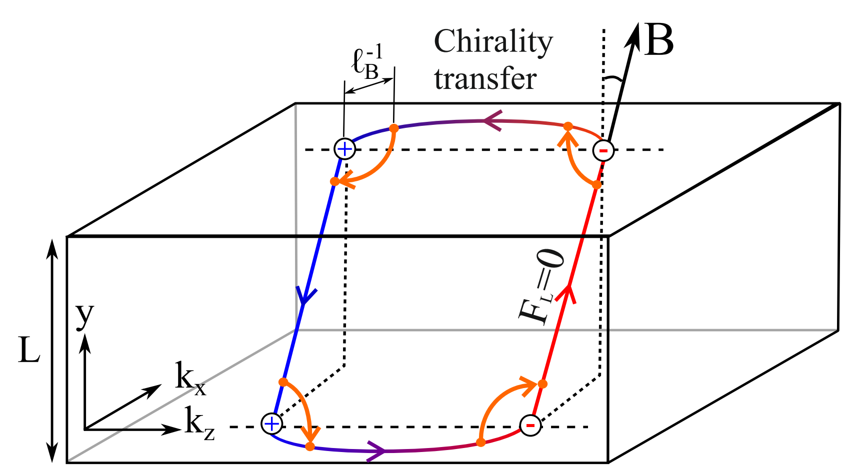
Yi et al. *Sci. Rep.* **4**, 6106 (2014)



Oscillations are 2D

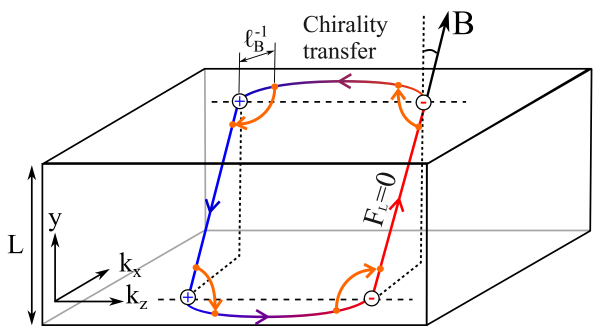






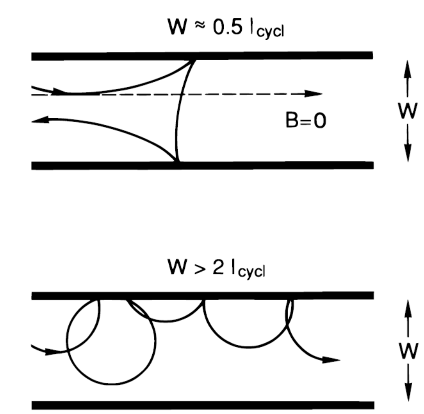
Philip J. W. Moll, JGA et al. arXiv:1505.02817

And grows exponentially with thinness

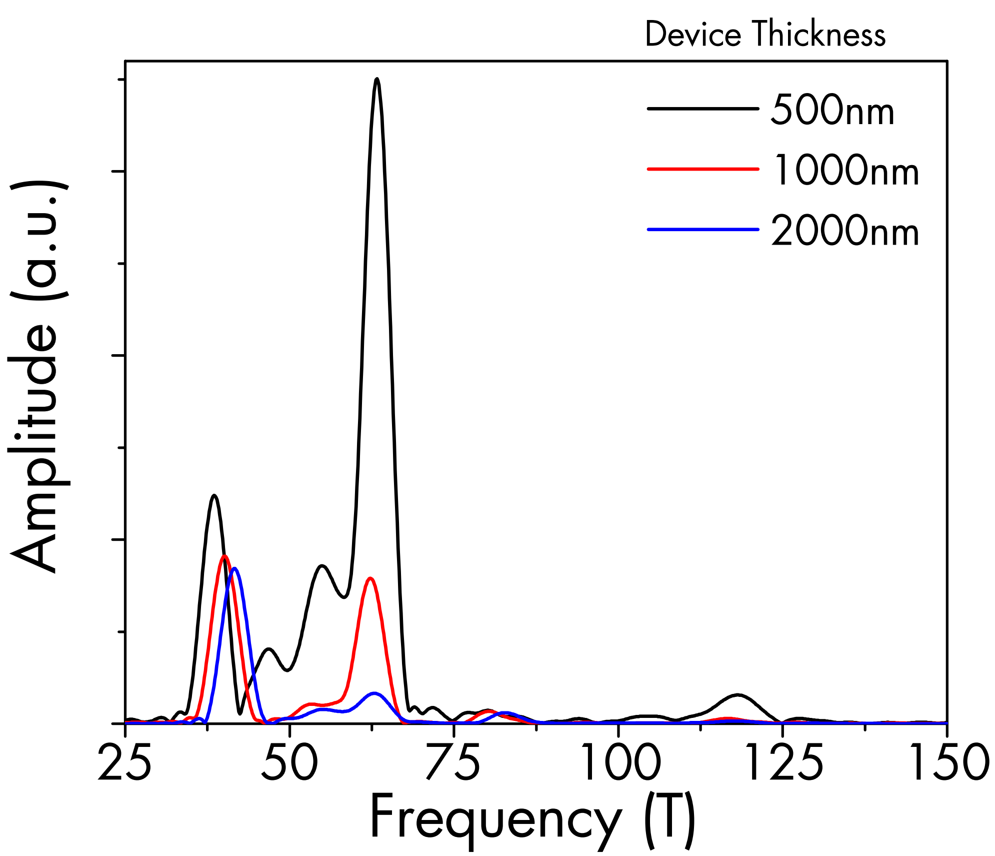


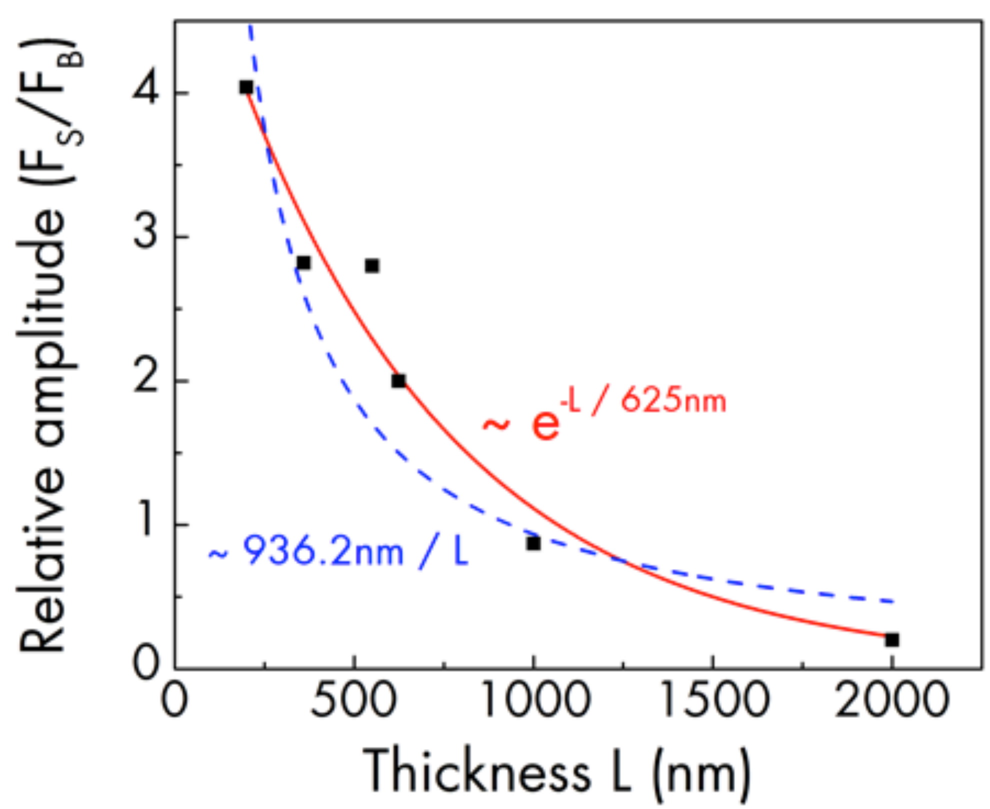
$$A_{\mathrm{SdH}} \sim e^{-1/\omega_c \tau}$$

$$\text{“}\frac{1}{\omega_c \tau}\text{”} \approx \frac{k_0}{e v_s B} \tau_s^{-1} + \frac{L}{v_B} \tau_B^{-1}$$



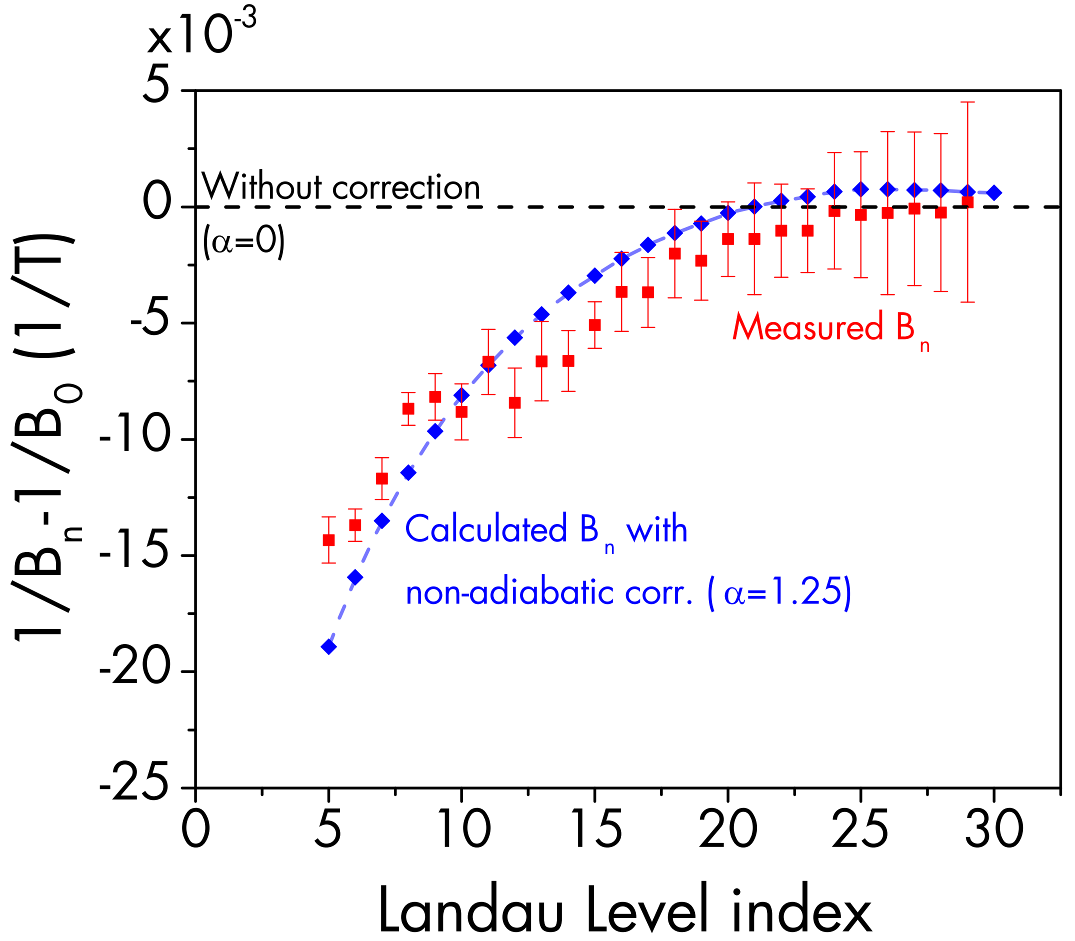
$$\ell_{\mathrm{MFP}} \approx 1.2 \mu m$$

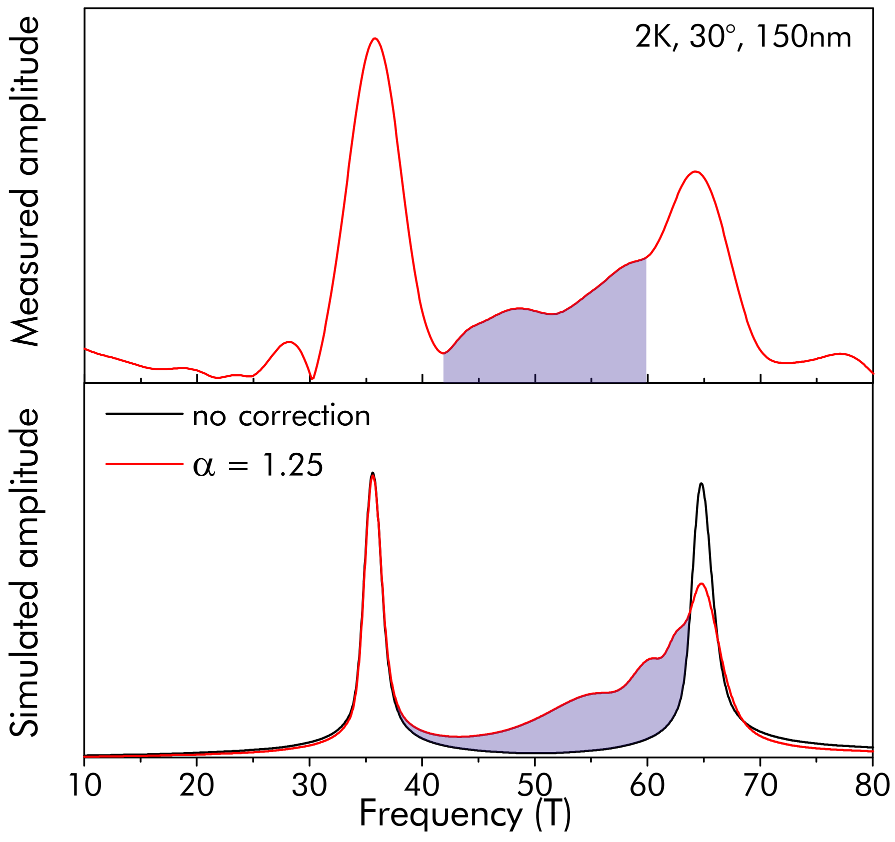




Philip J. W. Moll, JGA et al. arXiv:1505.02817

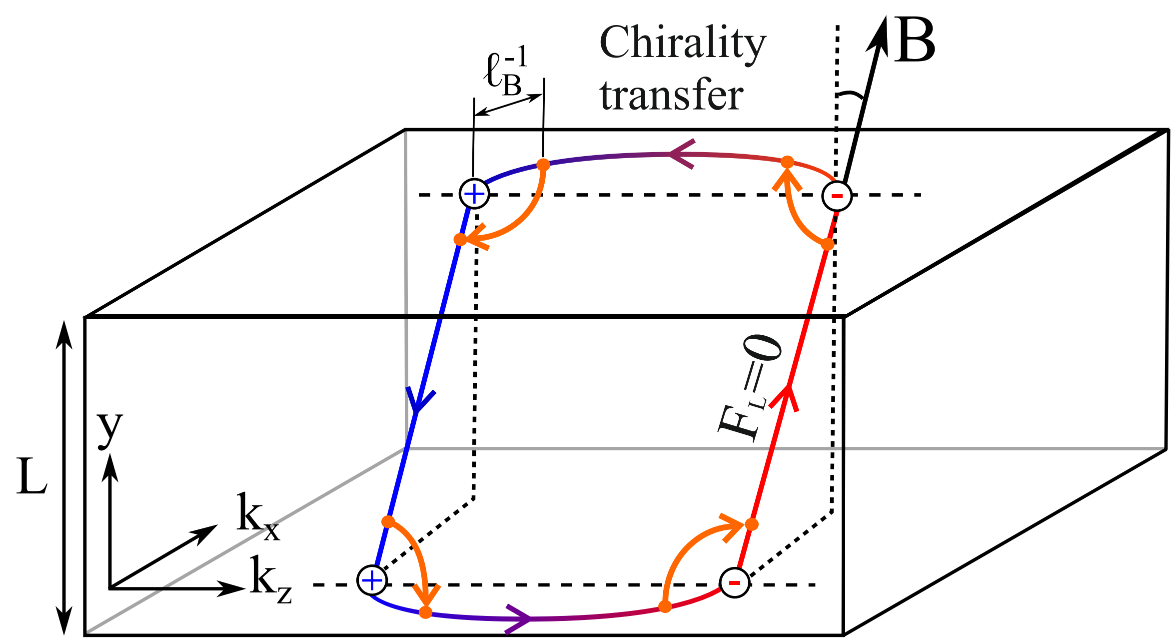
Some other, unexpected details





Philip J. W. Moll, JGA et al. arXiv:1505.02817

Non-adiabatic corrections in Weyl orbits?



$$\delta F_s(B) \approx -4\alpha \frac{\ell_B}{k_{\rm arc}}$$

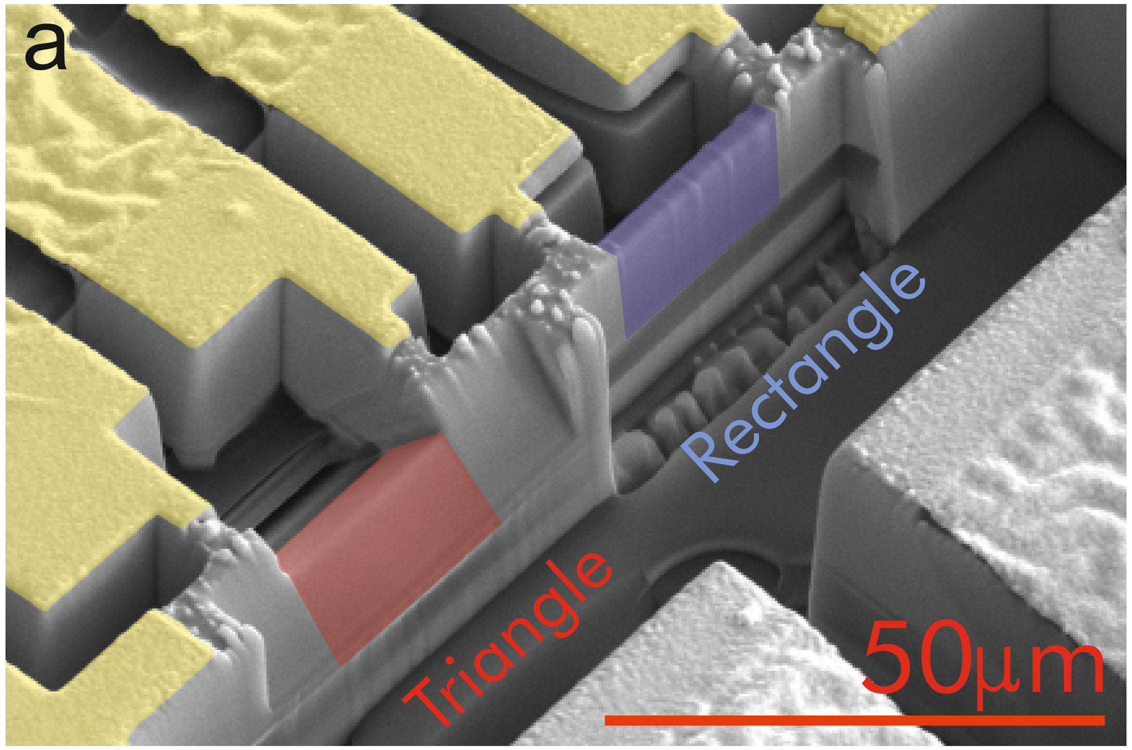
Andrew C. Potter, I.Kimchi, A. Vishwanath, Nature Communications (2014)

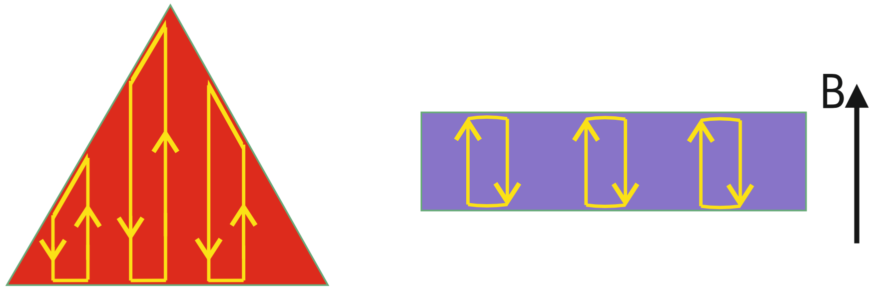
We get α~1.2

Philip J. W. Moll, JGA et al. arXiv:1505.02817

Does the phase of the oscillations depend on thickness?

- Detailed thickness dependence prohibitively difficult at the moment (frequency is too high and would require 1nm thickness dependence)
- So we came up with something different - a triangular geometry.
- The orbit is averaging over all length scales, causing the QOs to destructively interfere.





$$\frac{1}{B_n} = \frac{2\pi n}{f_{1/B}} - \frac{e}{k_{\mathrm{arc}}} L$$

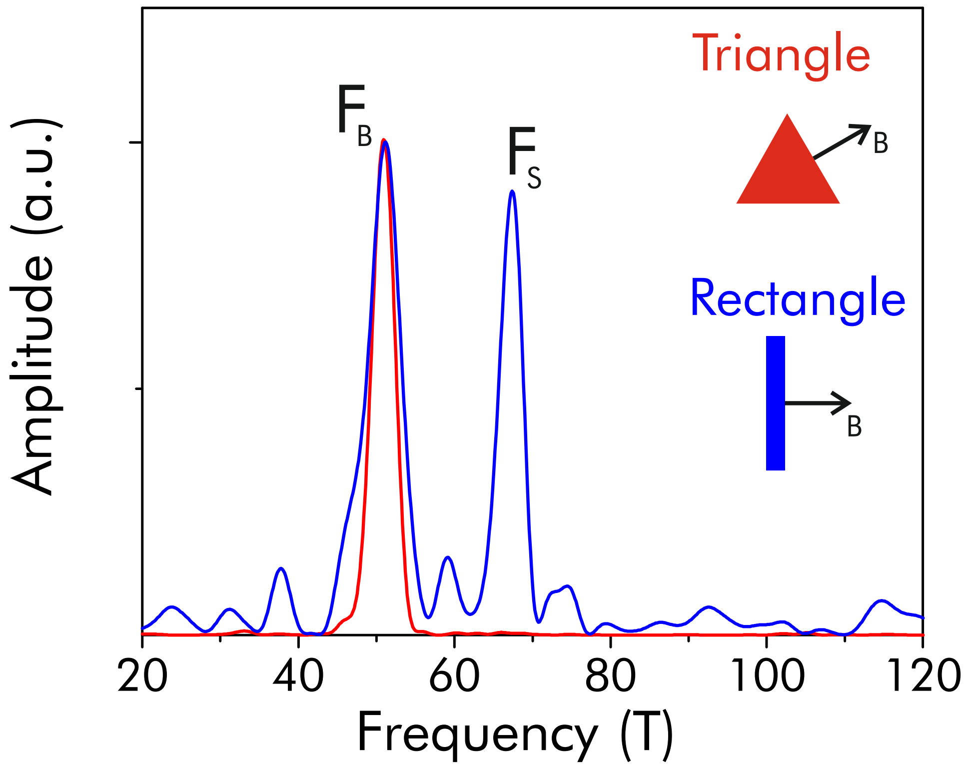
Philip J. W. Moll, JGA et al. arXiv:1505.02817

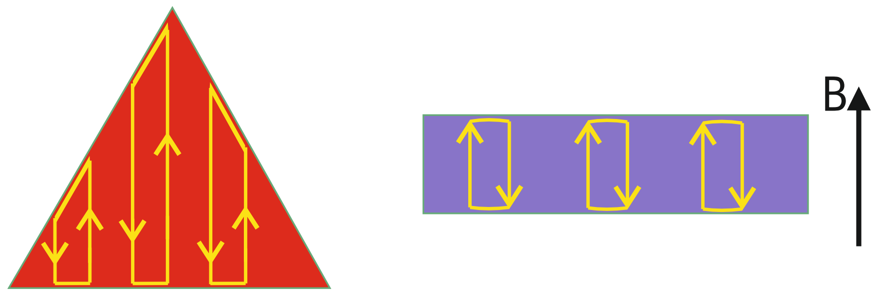
Does the phase of the oscillations depend on thickness?

- Detailed thickness dependence prohibitively difficult at the moment (frequency is too high and would require 1nm thickness dependence)

- So we came up with something different - a triangular geometry.

- The orbit is averaging over all length scales, causing the QOs to destructively interfere.



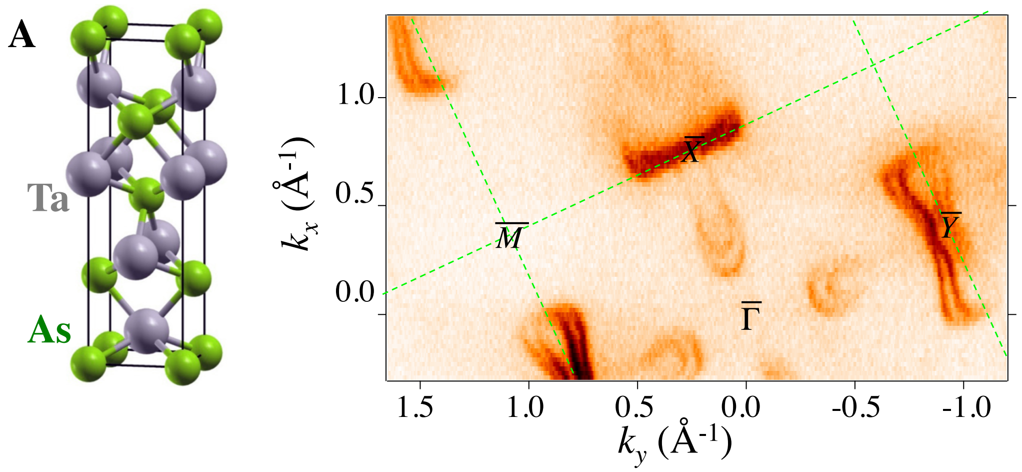


$$\frac{1}{B_n} = \frac{2\pi n}{f_{1/B}} - \frac{e}{k_{\text{arc}}} L$$

Philip J. W. Moll, JGA et al. arXiv:1505.02817

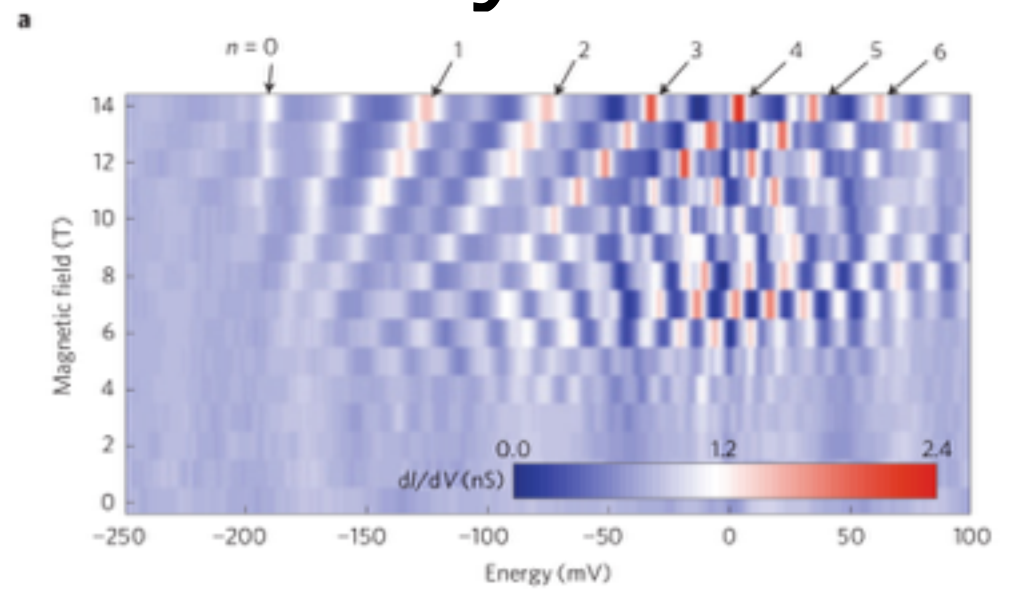
Observation	Trivial	Weyl
2D QOs	Y	Y
Frequency ~56T	Coincidence	Y
Amplitude exponential with L	Close	Y
Onset at $L=2l$	Coincidence	Y
Parallel surface required	N	Y
Field dependent phase.	Unphysical	Y
Saturation field B*	N	N (not yet)

Take a walk on the Weyl side



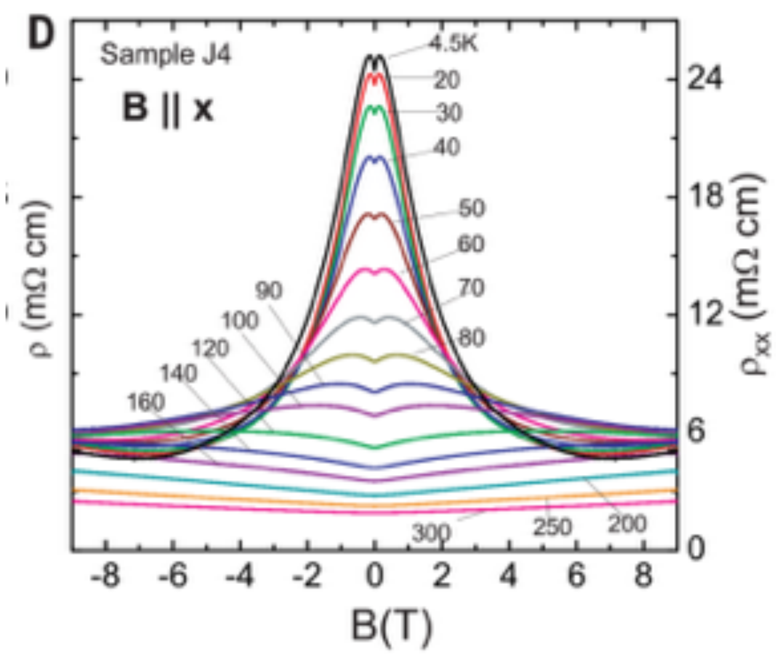
ARPES on TaAs

(Hasan group, Nat. Comm. '15)



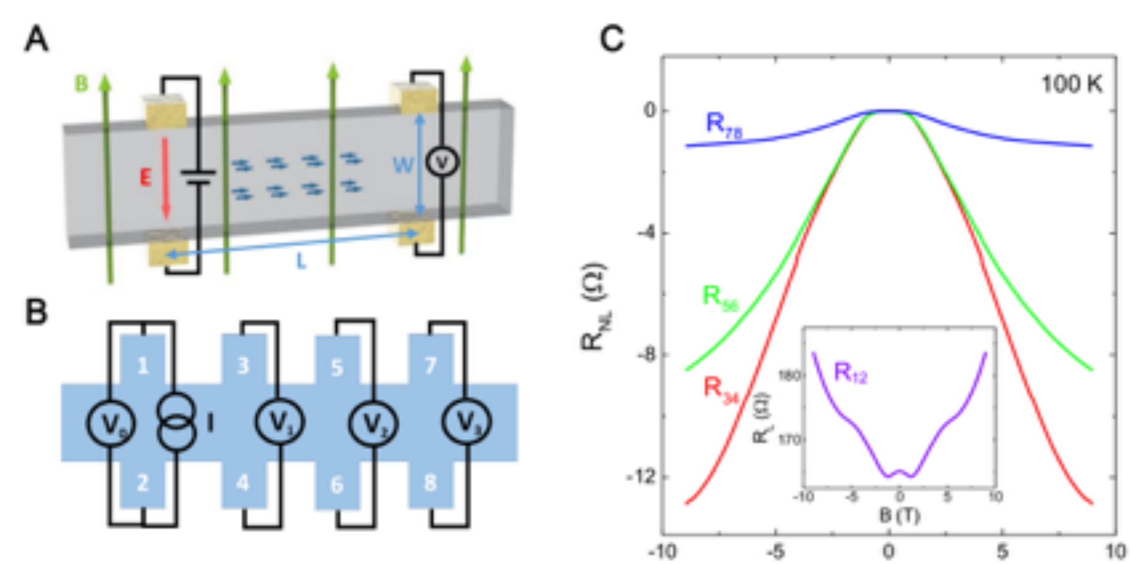
STM on Cd_3As_2

(Yazdani group, Nat. Mat. '14)



Negative MR on Na_3Bi

(Ong group, Science '15)

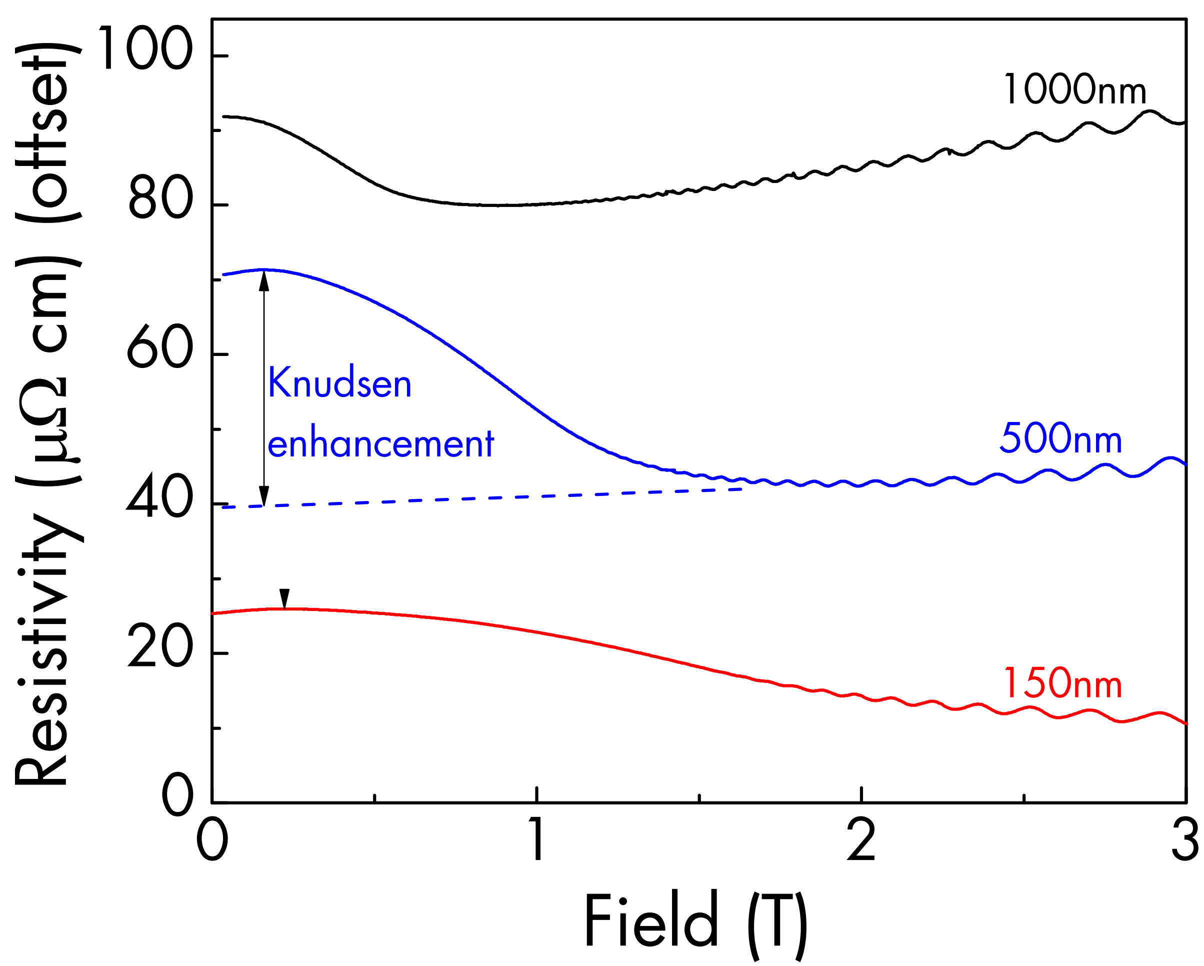


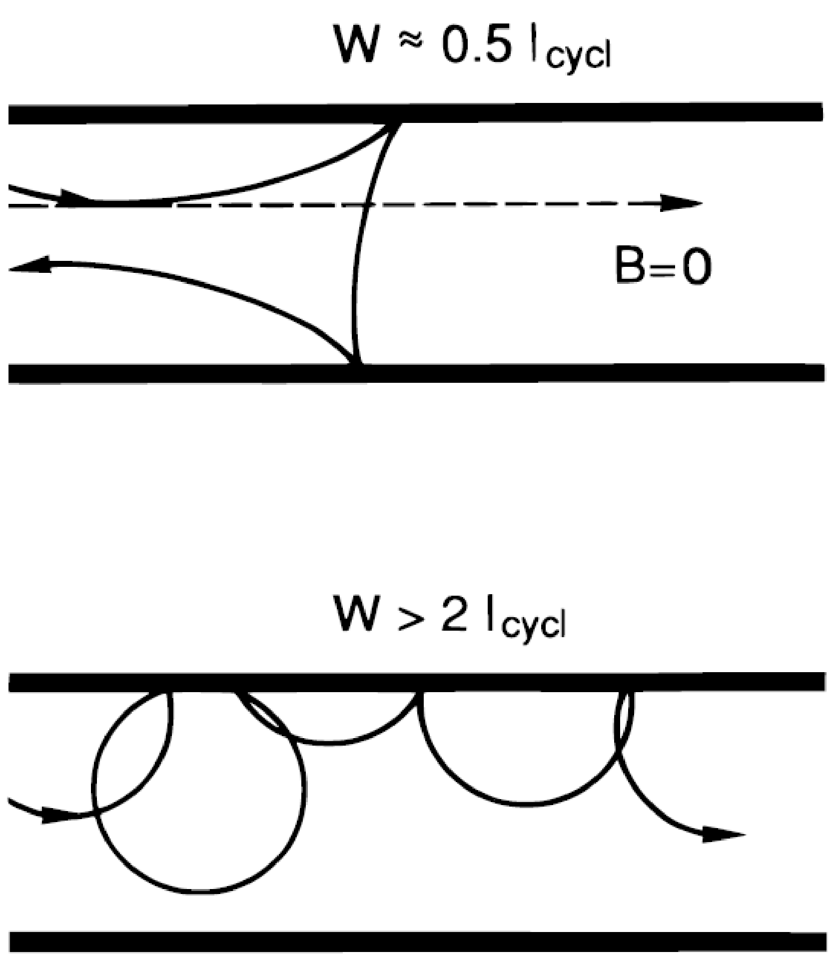
Non-local transport

(Xiu group, Fudan '15)

Amplitude onsets at *L~2l* (Knudsen effect)

(*B* Parallel to surface only)





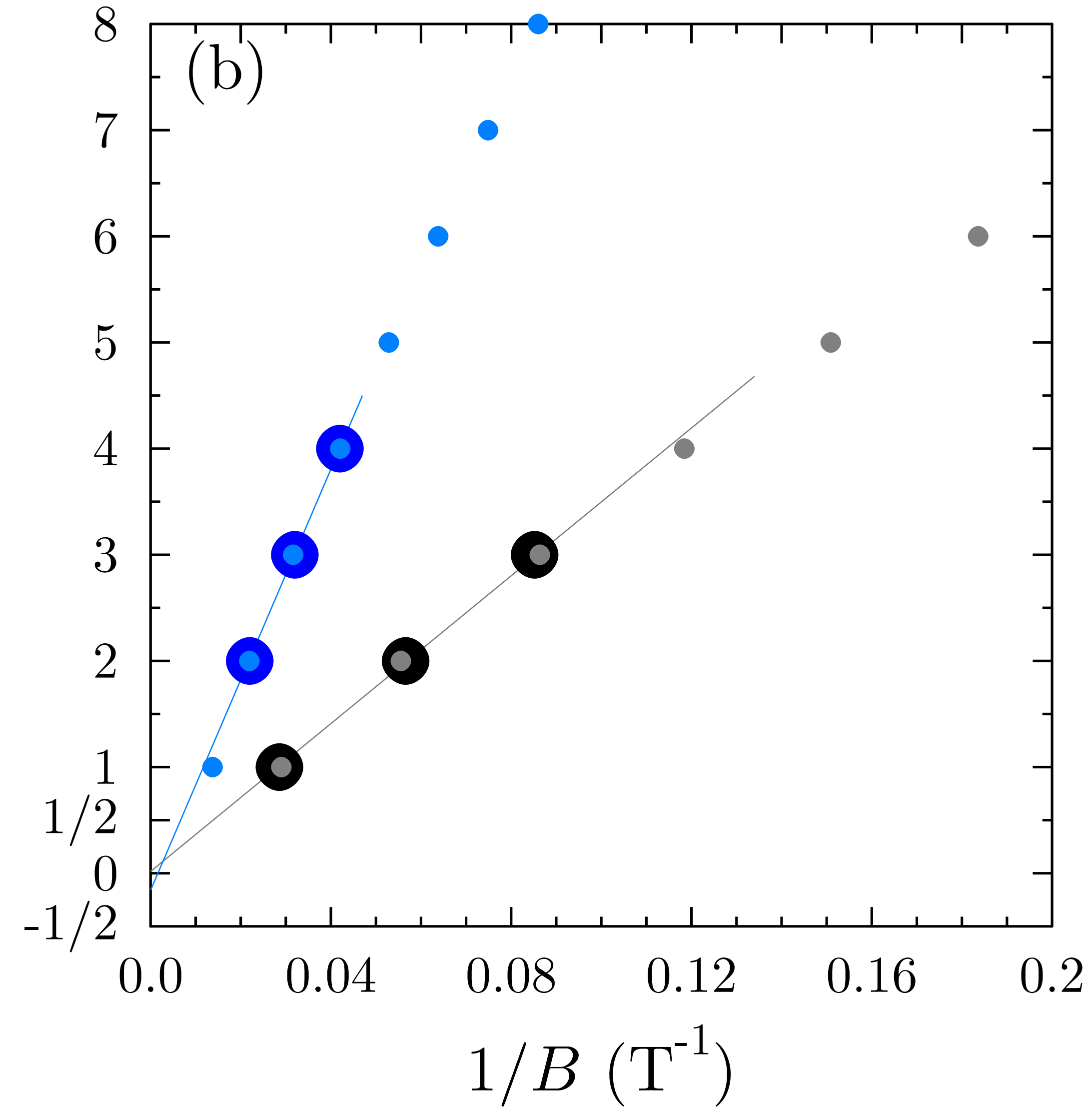
$\ell_{\mathrm{MFP}} \approx 1.2\mu m$

Philip J. W. Moll, JGA et al. arXiv:1505.02817

Mass corrections in TI surface states?

- Seen in Rashba systems (BiTeI) and TIs
- But….

    1. the effect seems to go in the wrong direction
    2. The g-factor required is 300 (10x anything measured in these compounds)



Analytis et al Nature Physics 2010

Transport and Focused Ion Beam microstructuring

FIB devices

A new 2D quantum oscillation

What about trivial surface states?

Exponential dependence on thickness
Onset at L=2lambda

Non-adiabatic corrections

Triangular vs parallel plate device