

Chiral transport of hot carriers in graphene in the quantum Hall regime

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Hicks Chair in Quantum Materials

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Classical Hall physics circa 1880



Wikipedia

Edwin Hall showed the transverse resistivity ρ_H of a thin metallic plate **varies linearly**

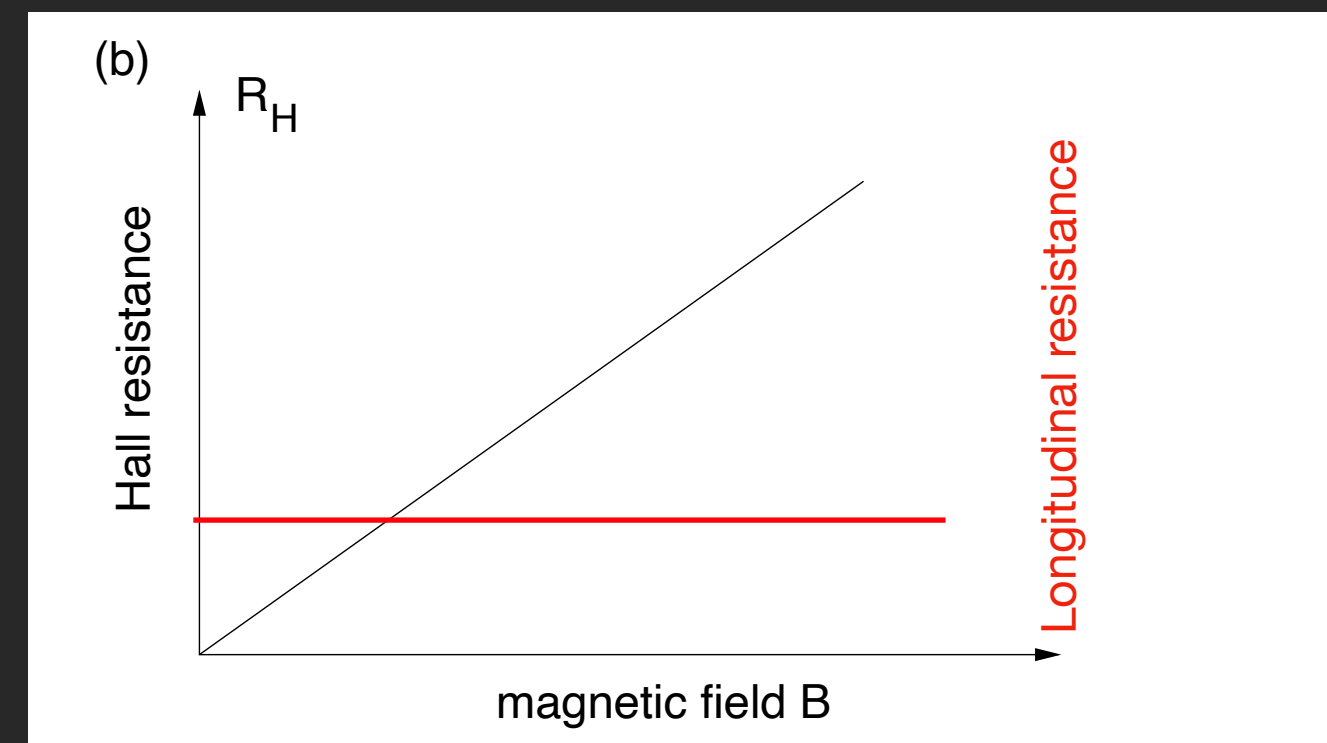
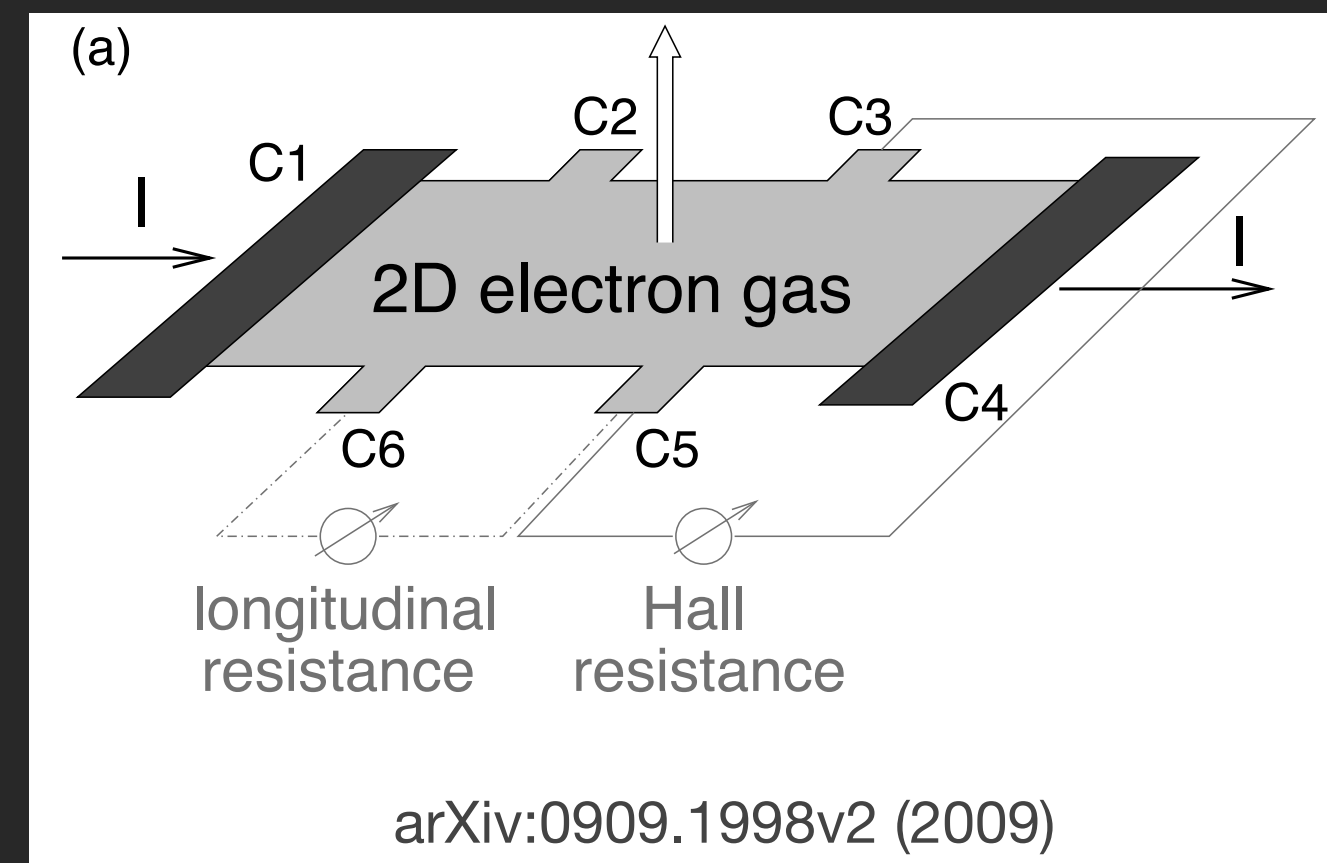
with the strength B of the perpendicular magnetic field

$$\rho_H = \frac{B}{qn_{el}}$$

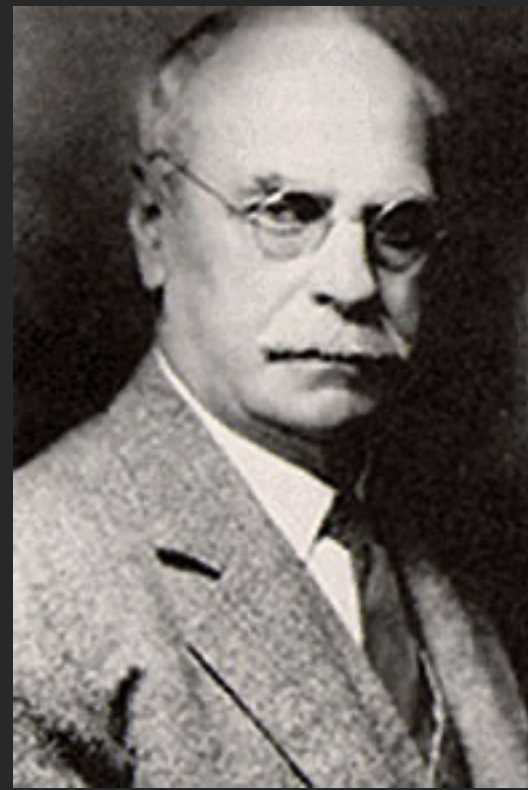
q is the charge ($-e$ for electrons), n_{el} is the 2D carrier density.

The longitudinal resistivity is independent of B .

Just the Lorentz force changing the trajectory of a charged particle.



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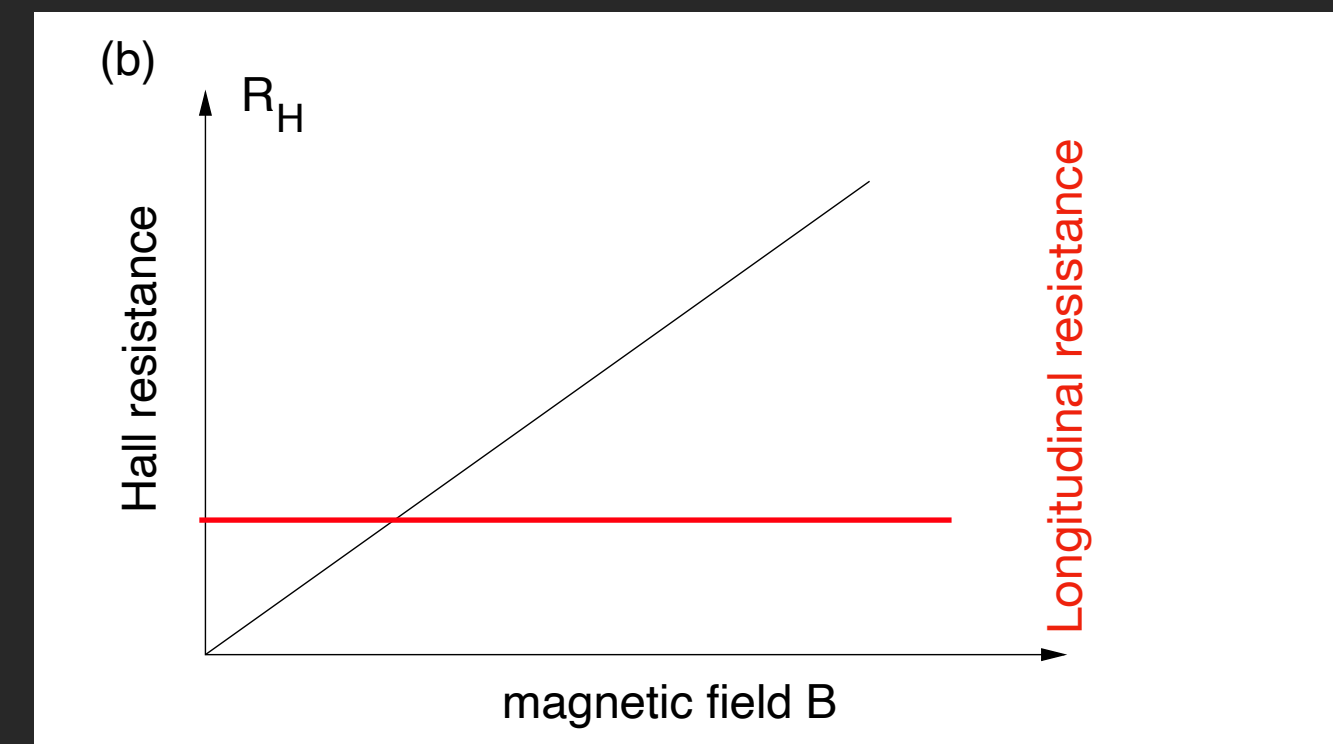
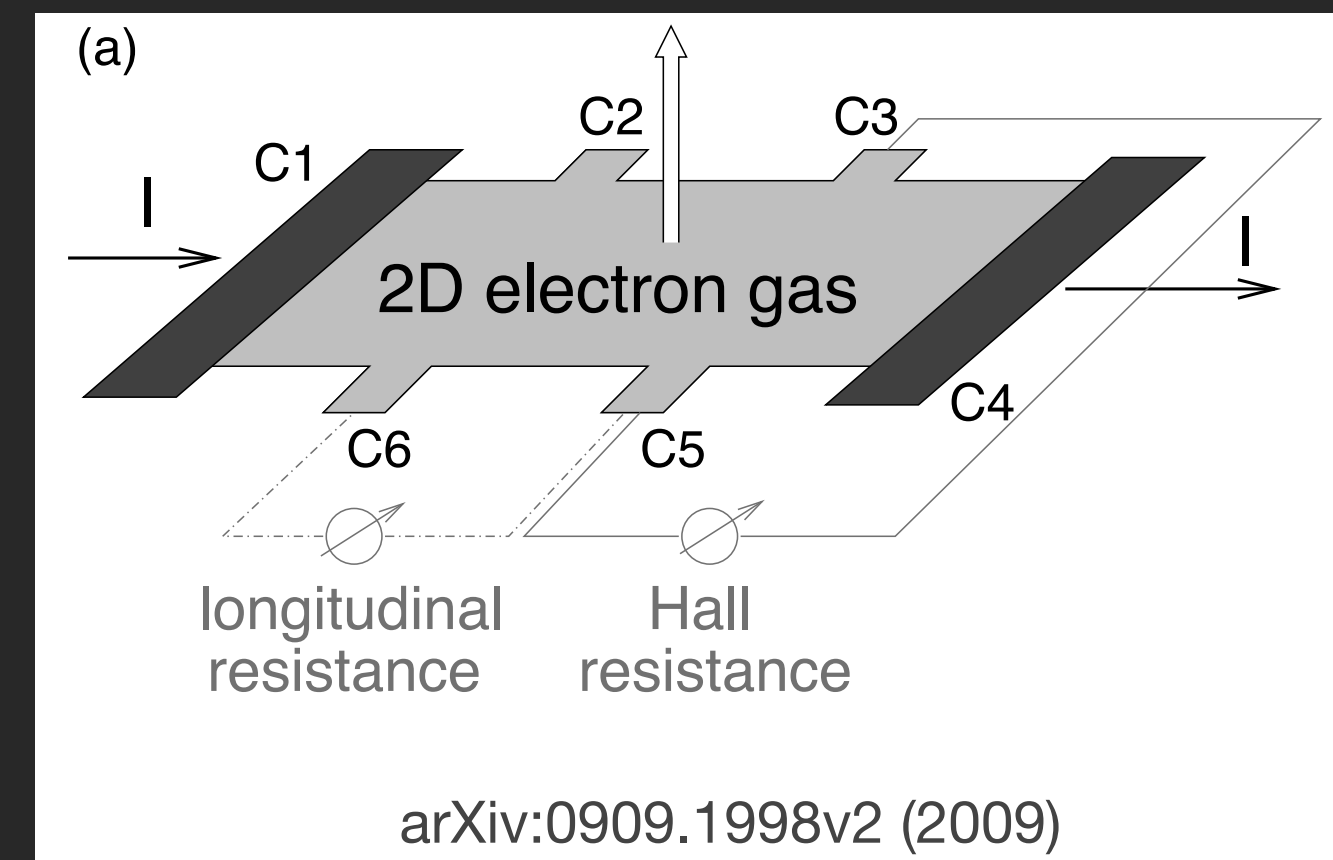
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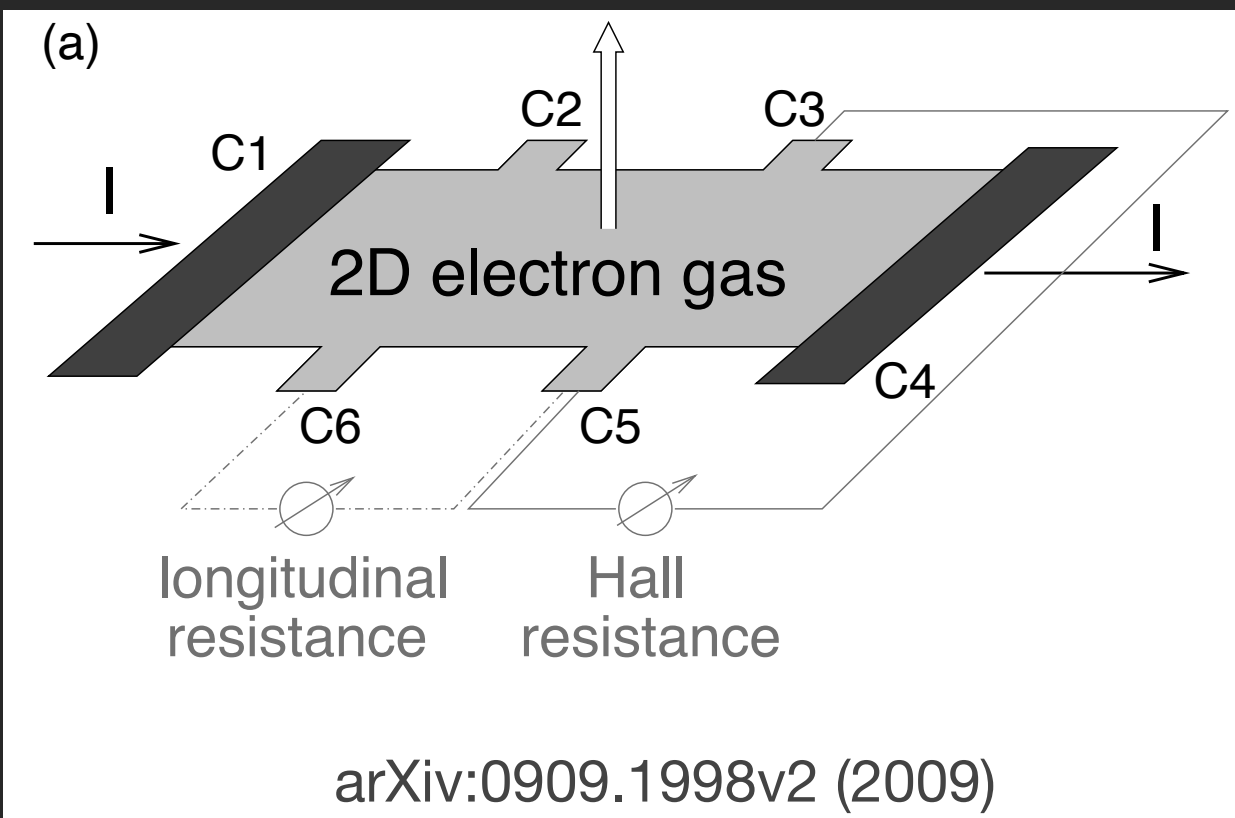
In the lab, we measure **Resistance** not **resistivity**, and $R = (L/A)\rho$ [l = length, A = cross section]

For a quasi 2D system, $R_H L = A$. $\rho = R$ making it robust with geometry change.

Another feature: In a semiconductor the carrier density and carrier mobility can found independent of each other.



Part 1: Integer Quantum Hall



In a 2D system quantisation of allowed electron/hole energies can present

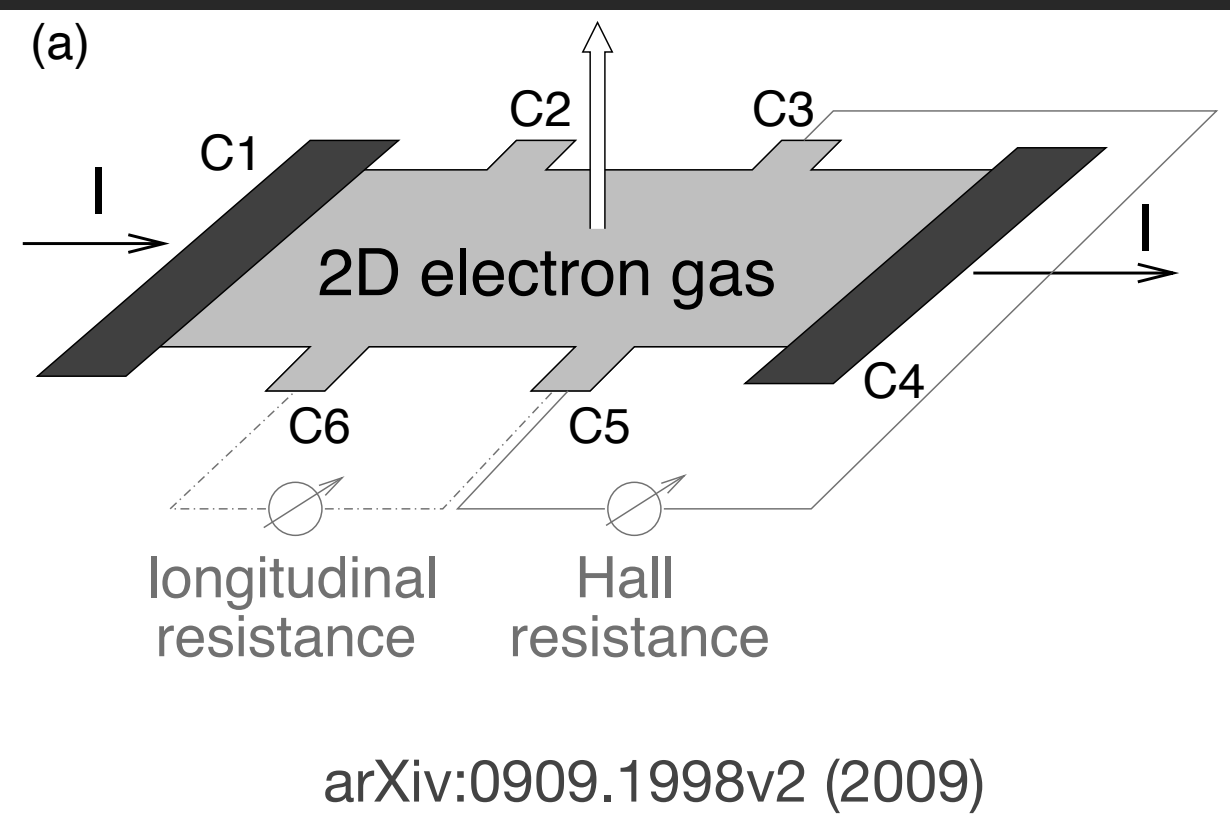
Parabolic bands: $\epsilon_n = \hbar\omega_C(n + 1/2)$,

n is an integer, cyclotron freq, $\omega_C = \frac{-qB}{m}$

iff charged-carrier scattering, τ^{-1} is weak, i.e., $\omega_C\tau > 1$

Quantisation of Density of States, $\rho(\epsilon) = \sum_n g_n \delta(\epsilon - \epsilon_n)$ g_n - degeneracy

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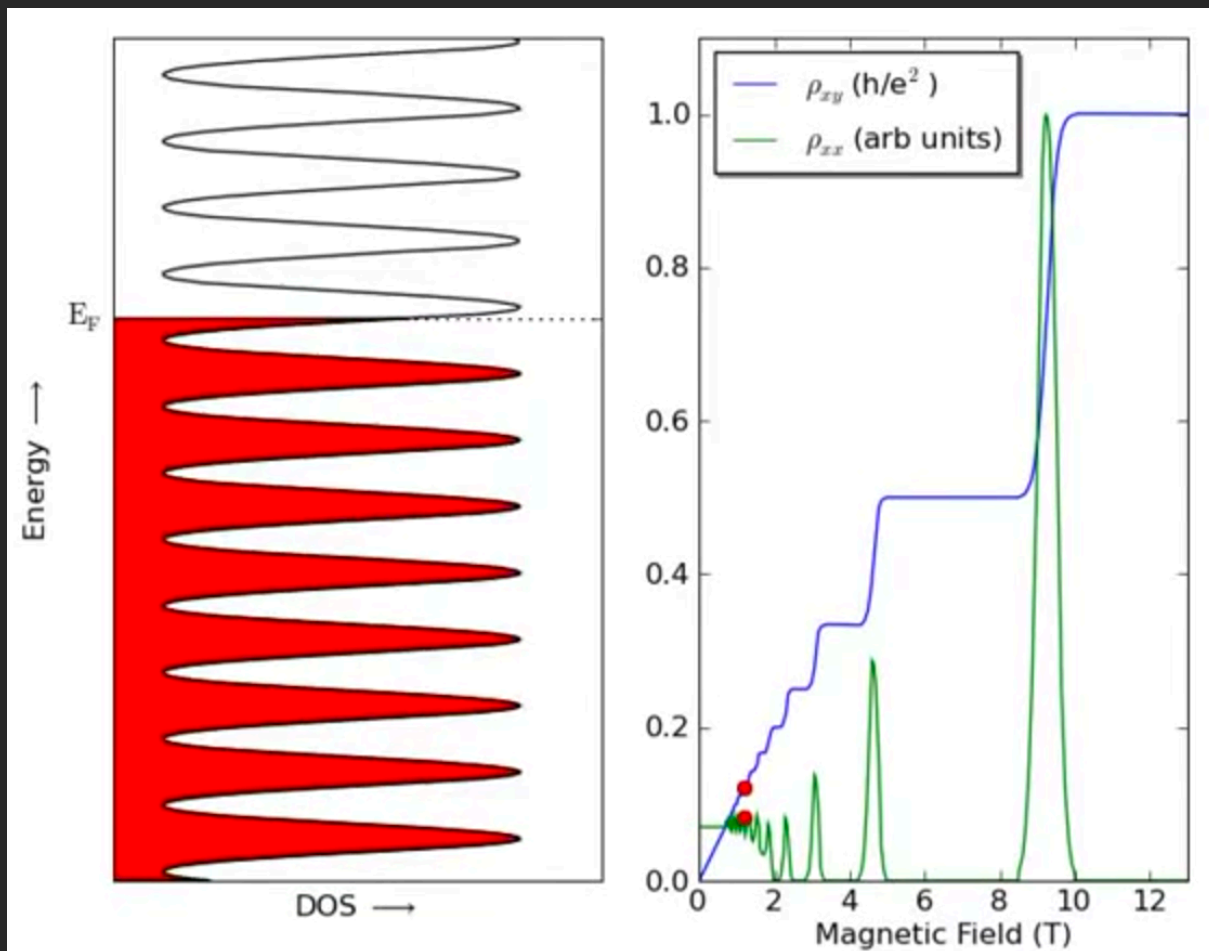
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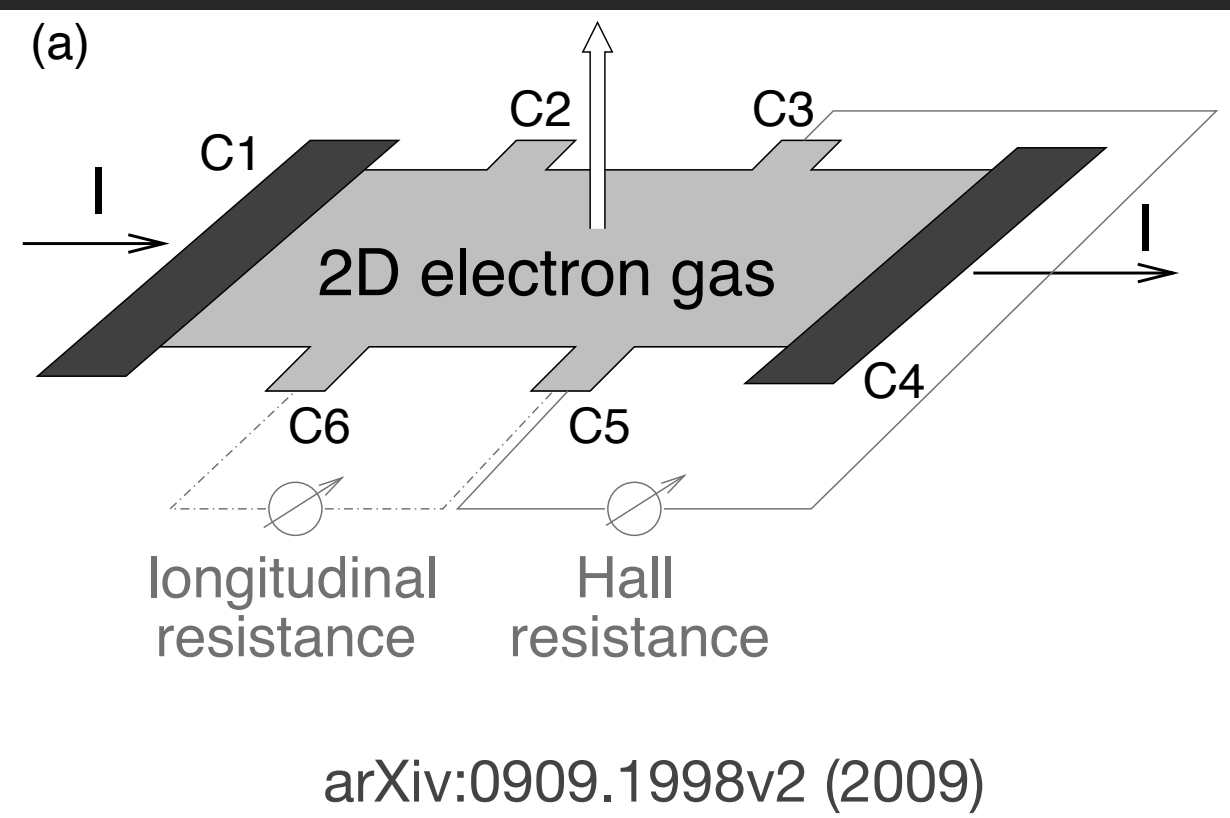
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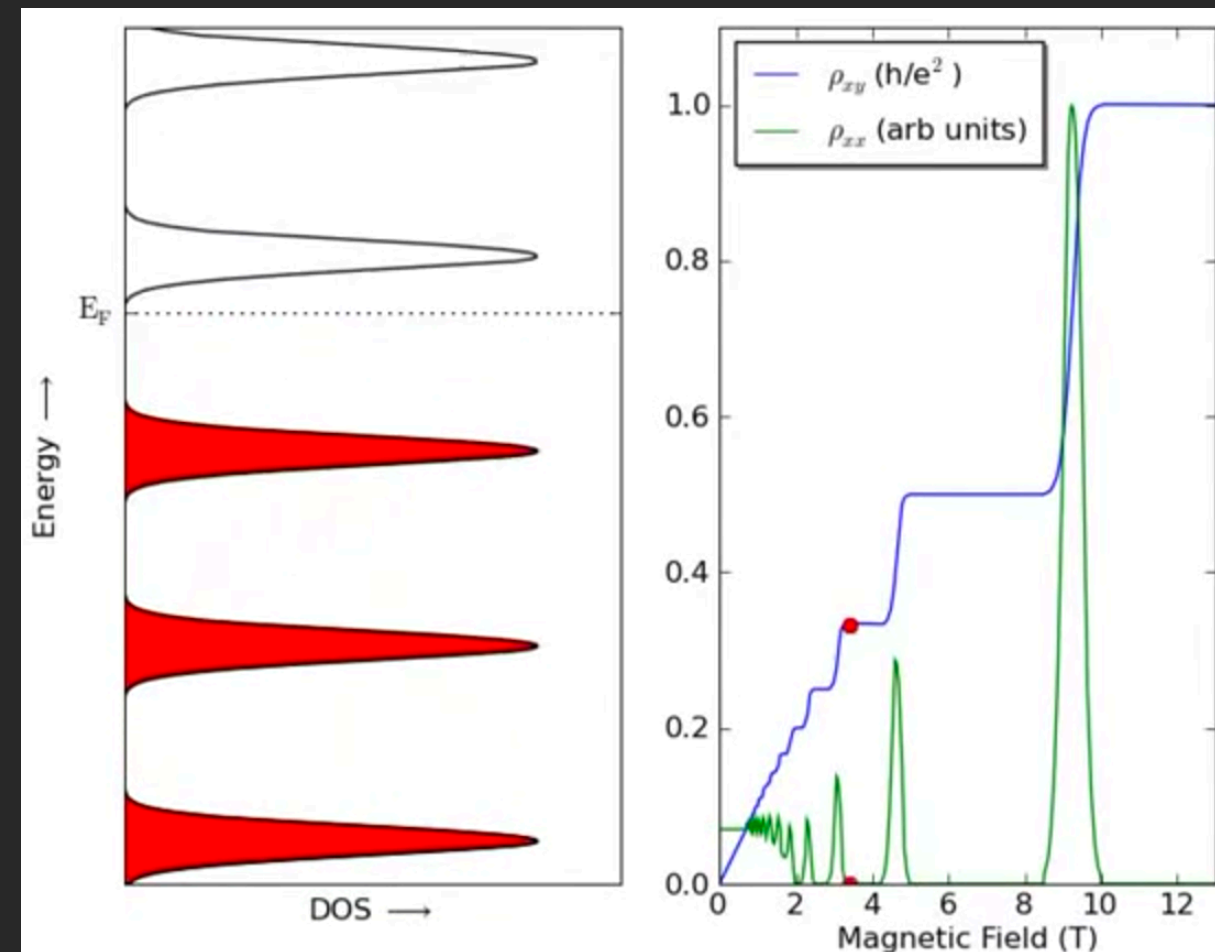
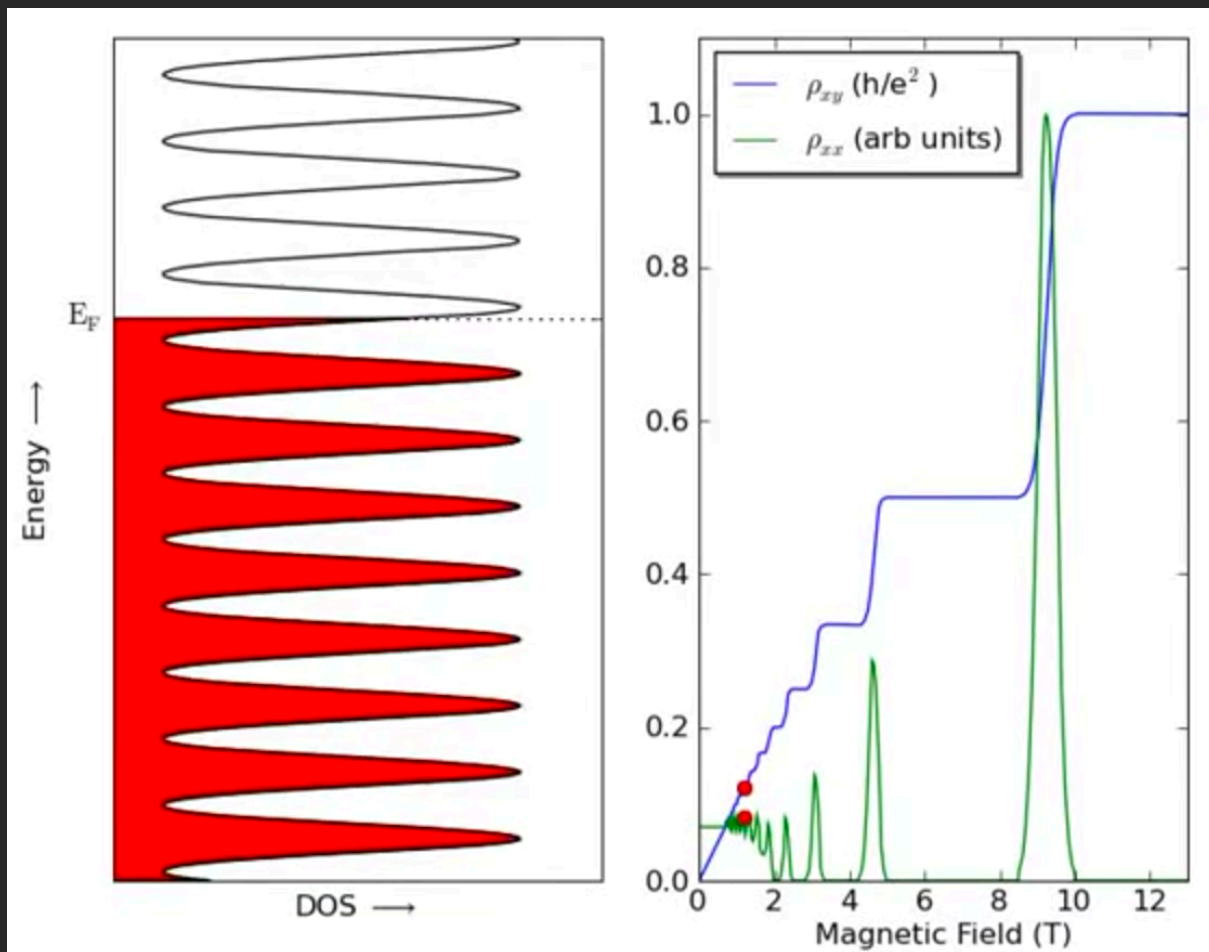
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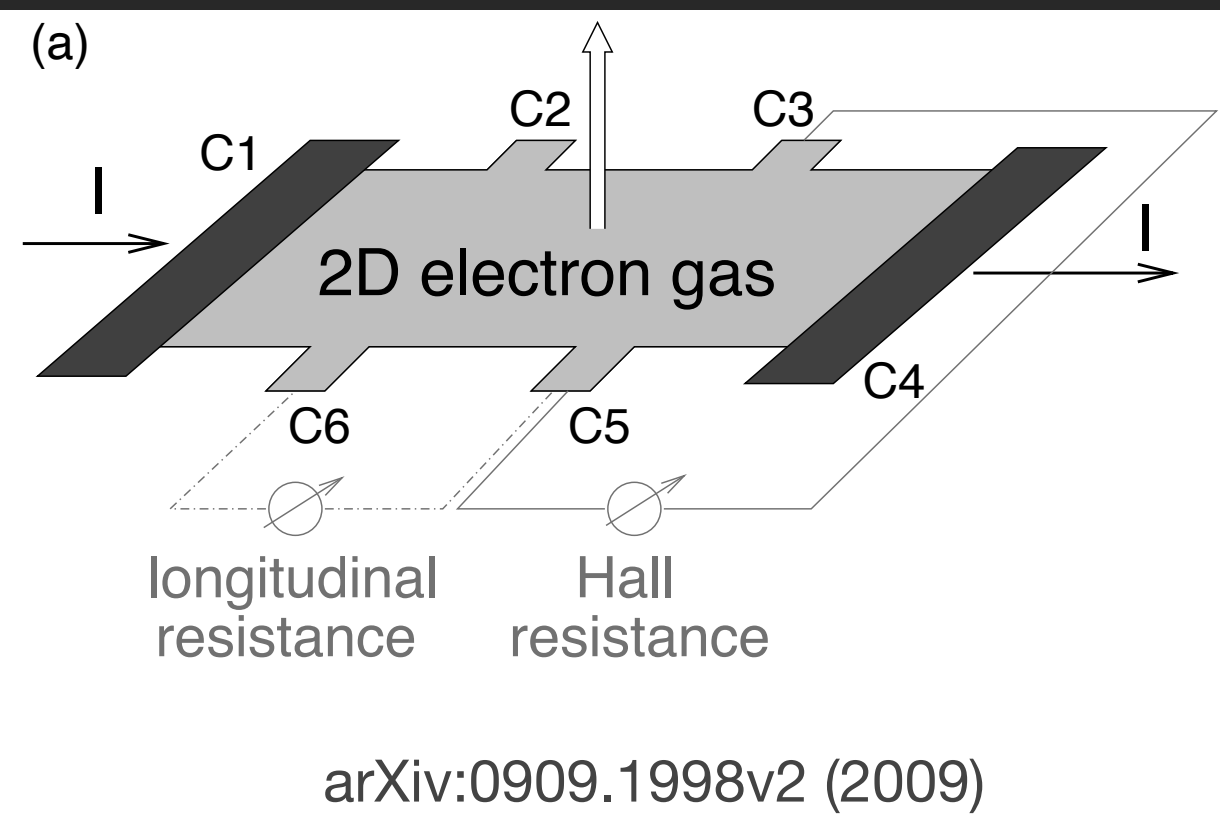
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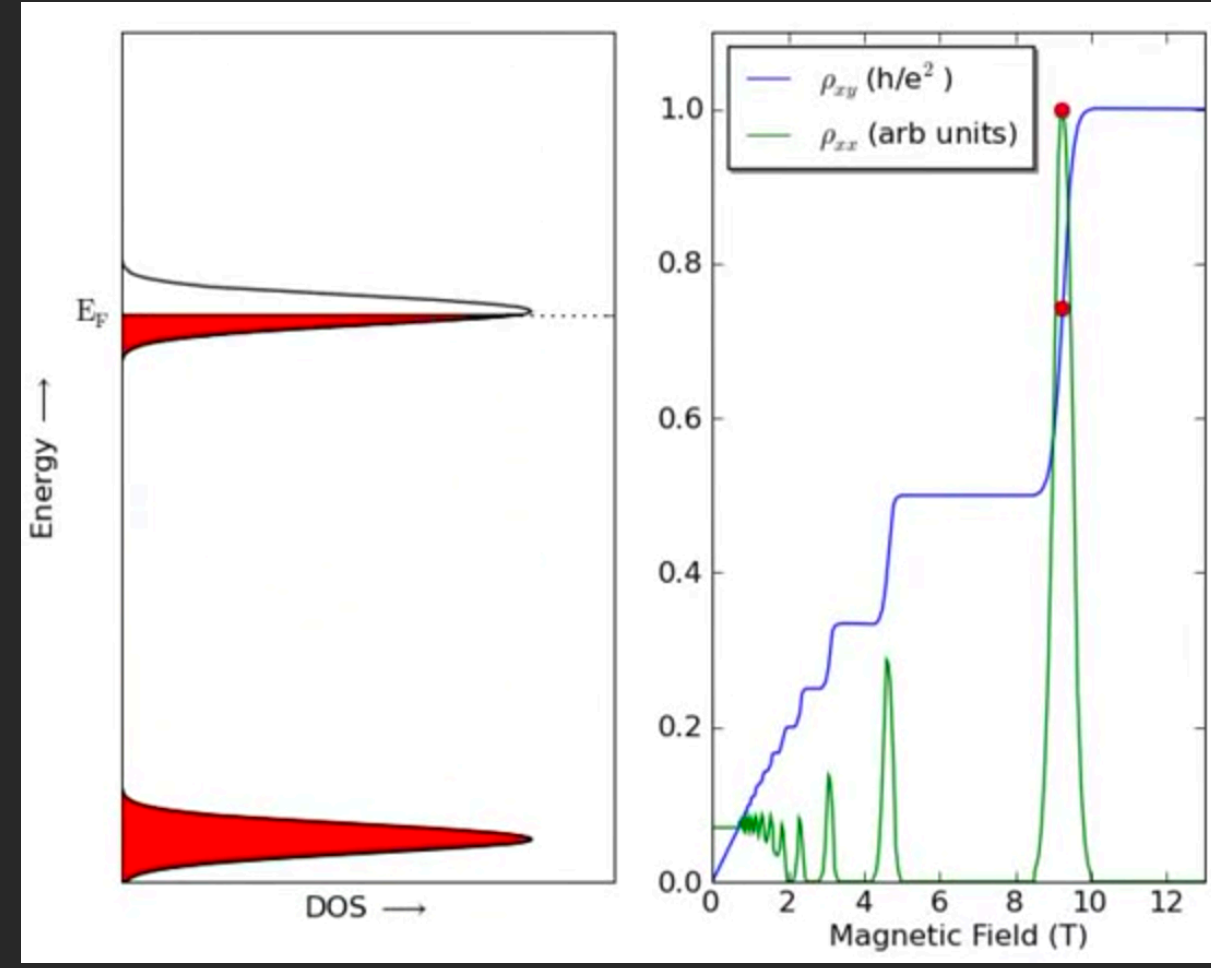
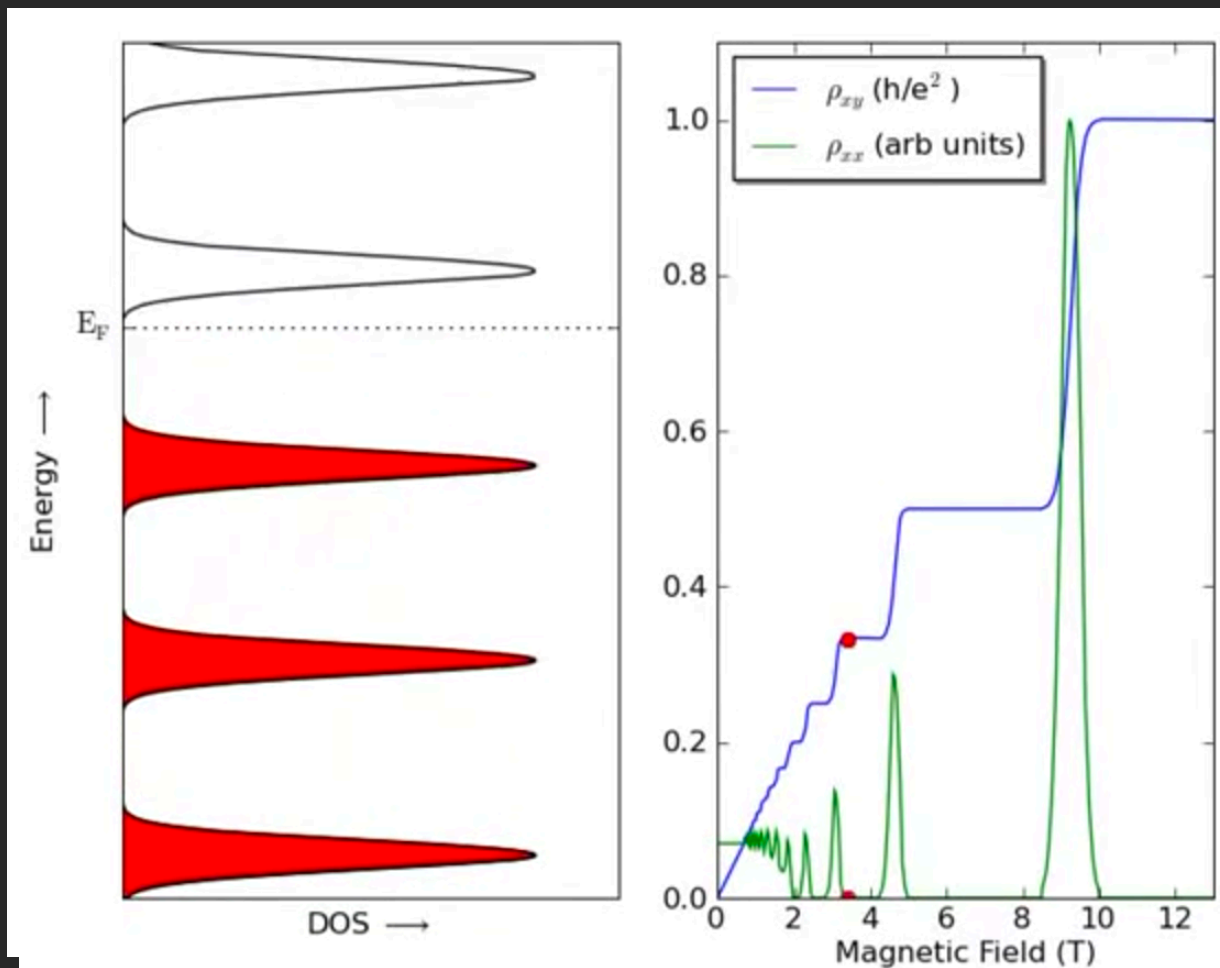
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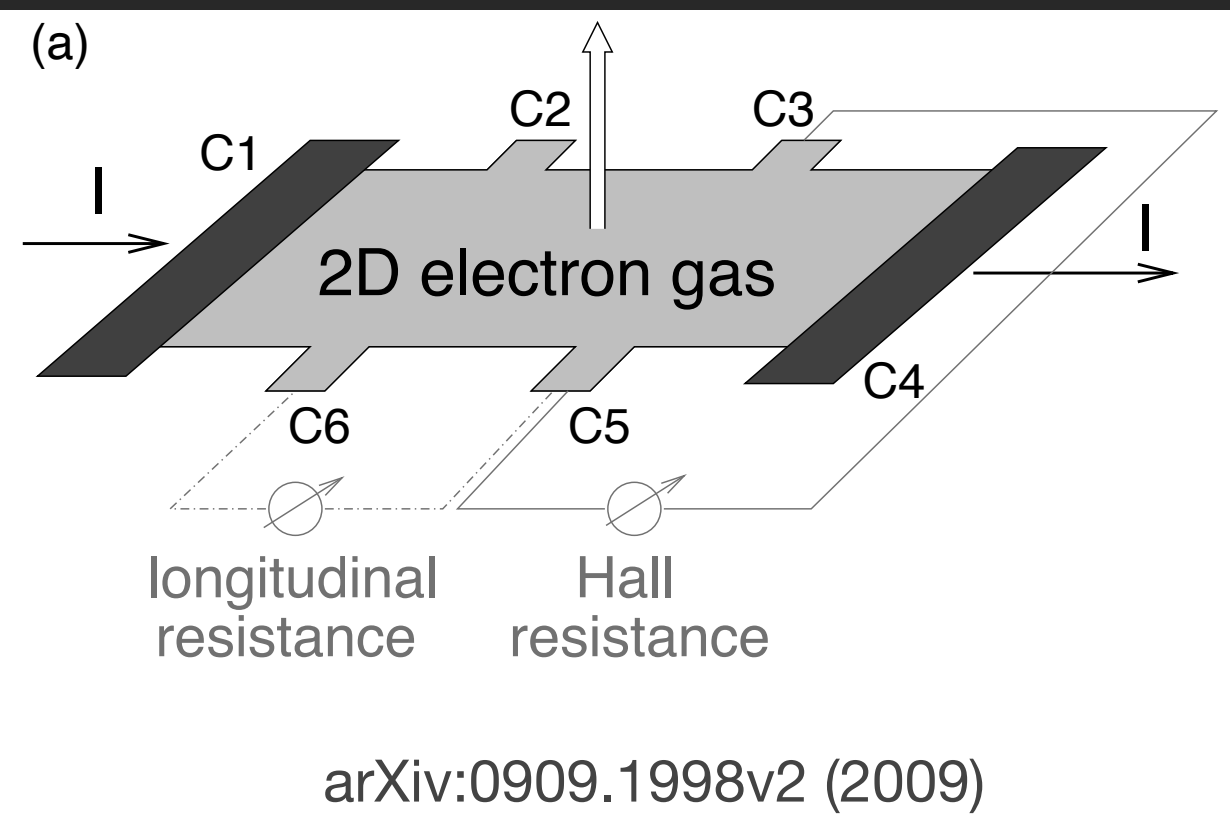
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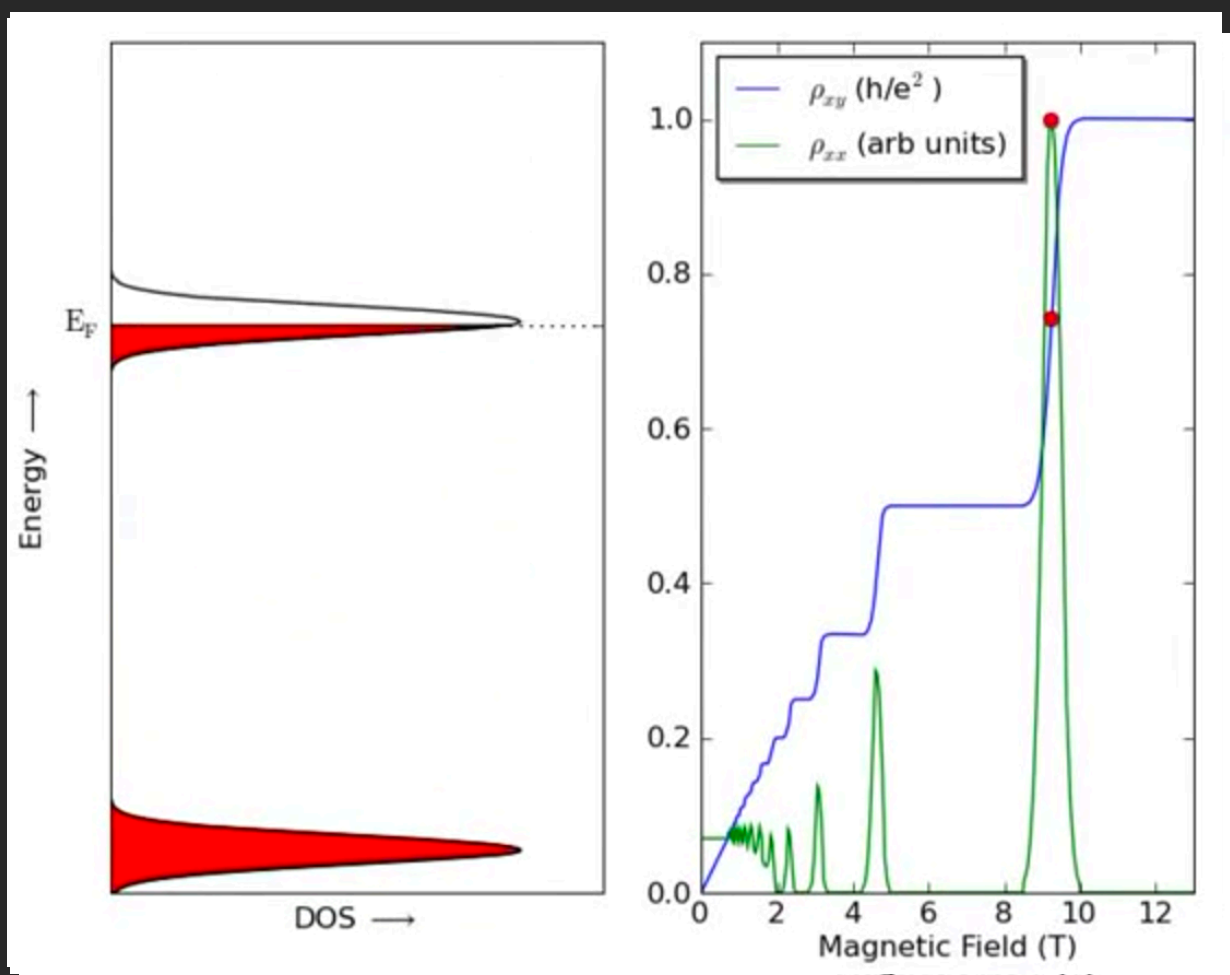
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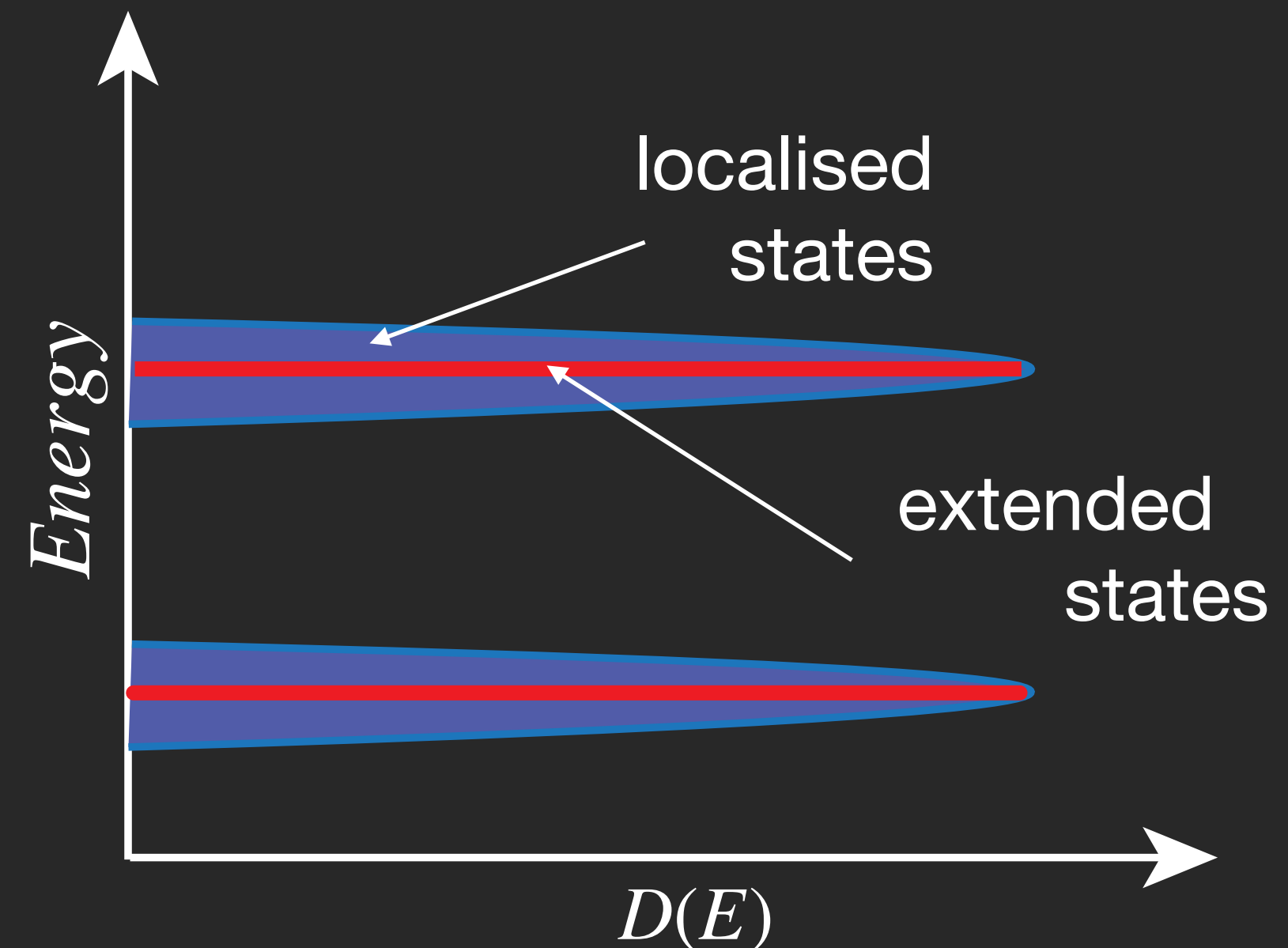
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Peaks in the DOS are broadened by impurity scattering

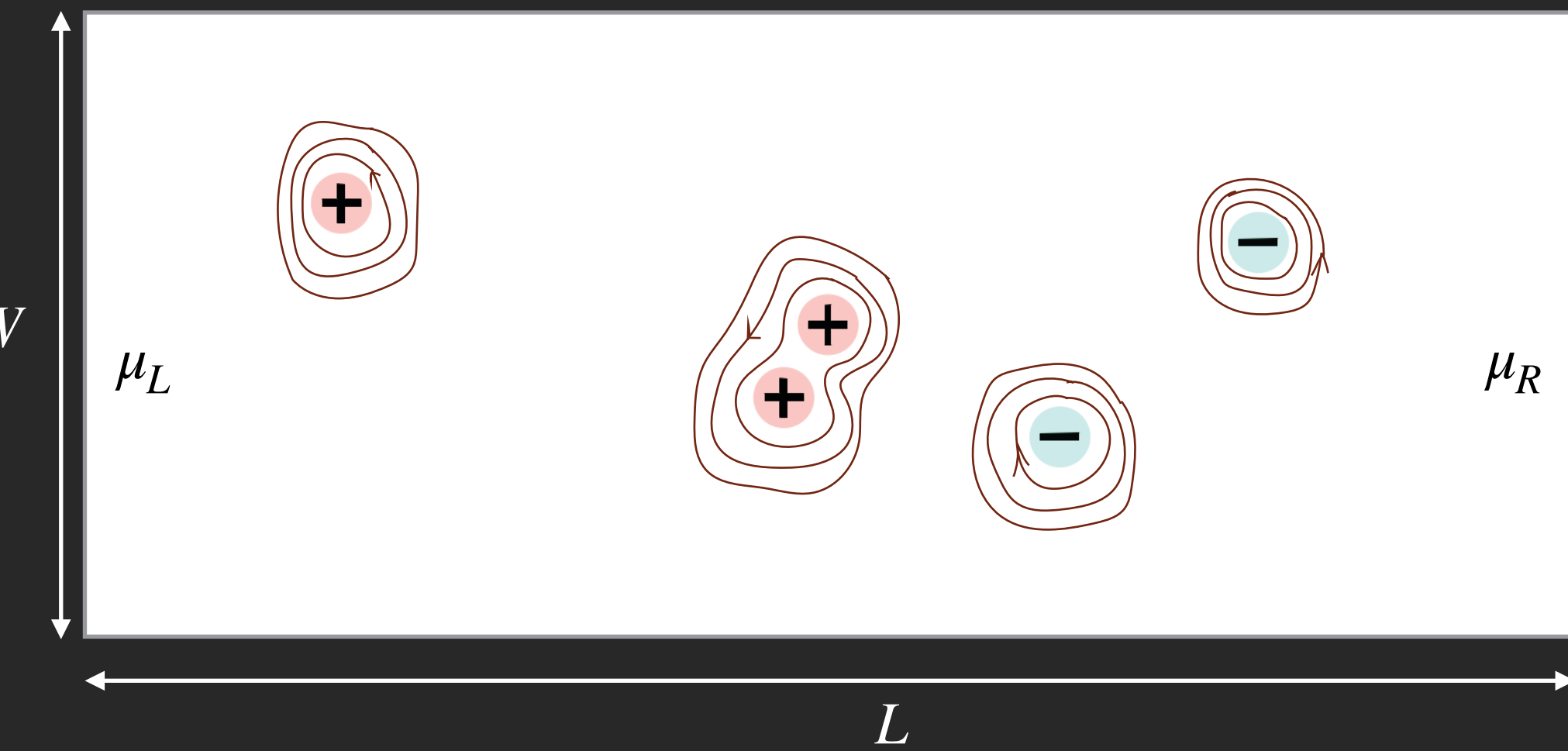


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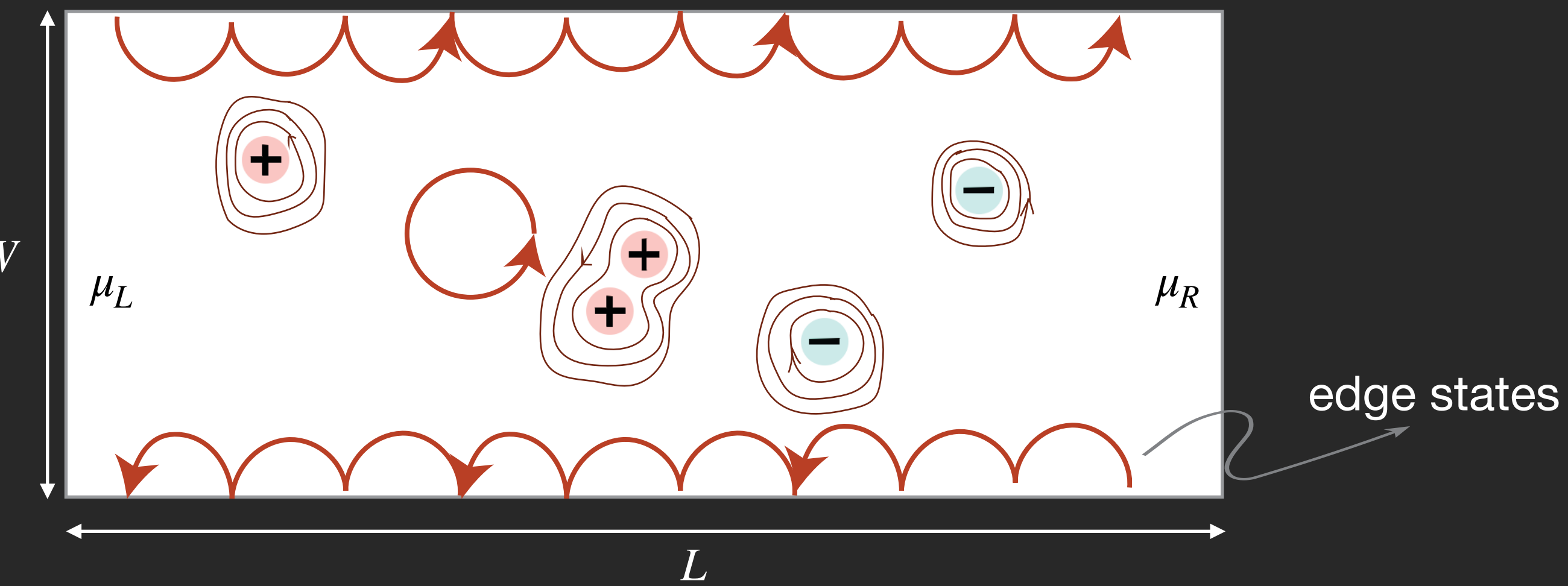
Integer Quantum Hall

- With low temperature, and low impurity scattering the DOS become quantised.



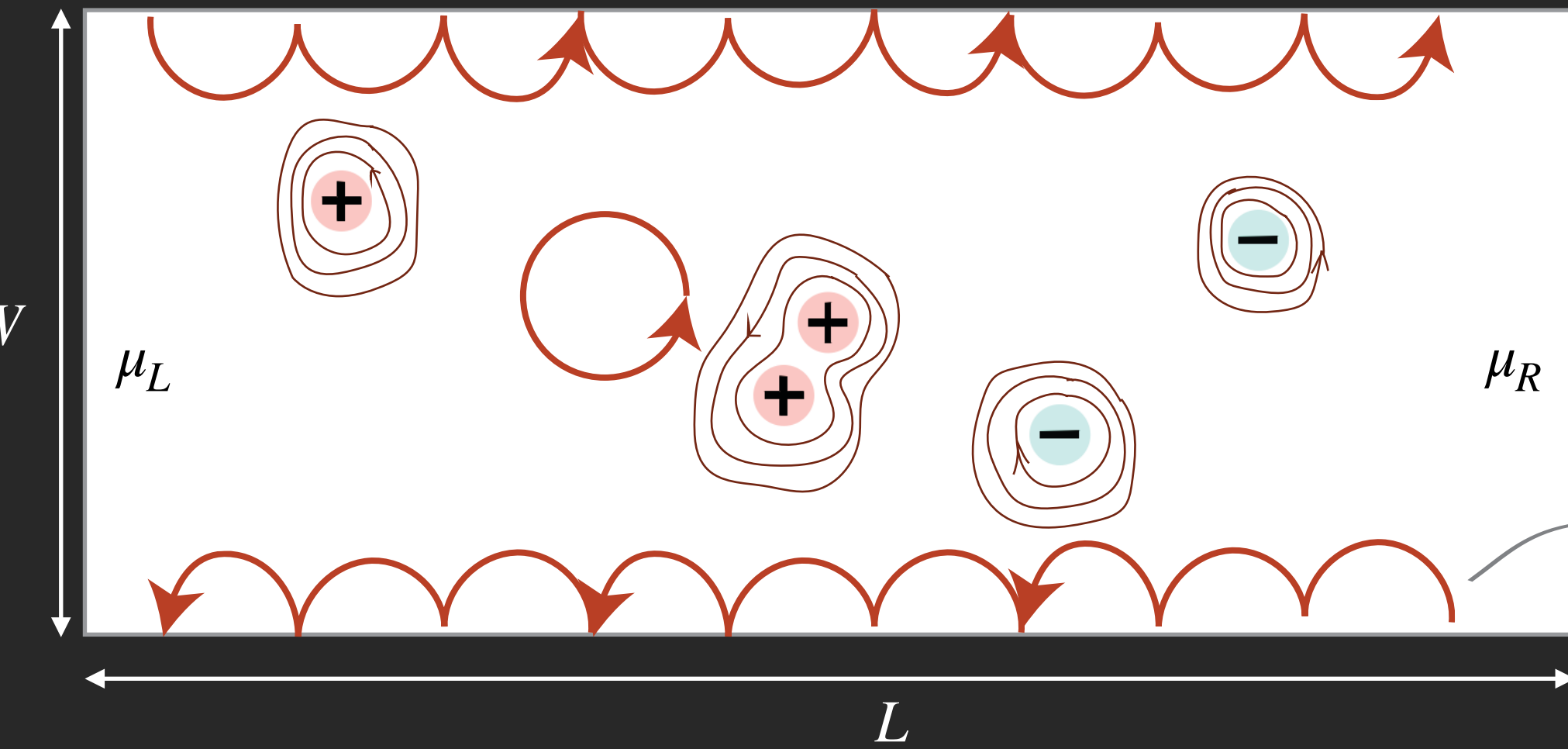
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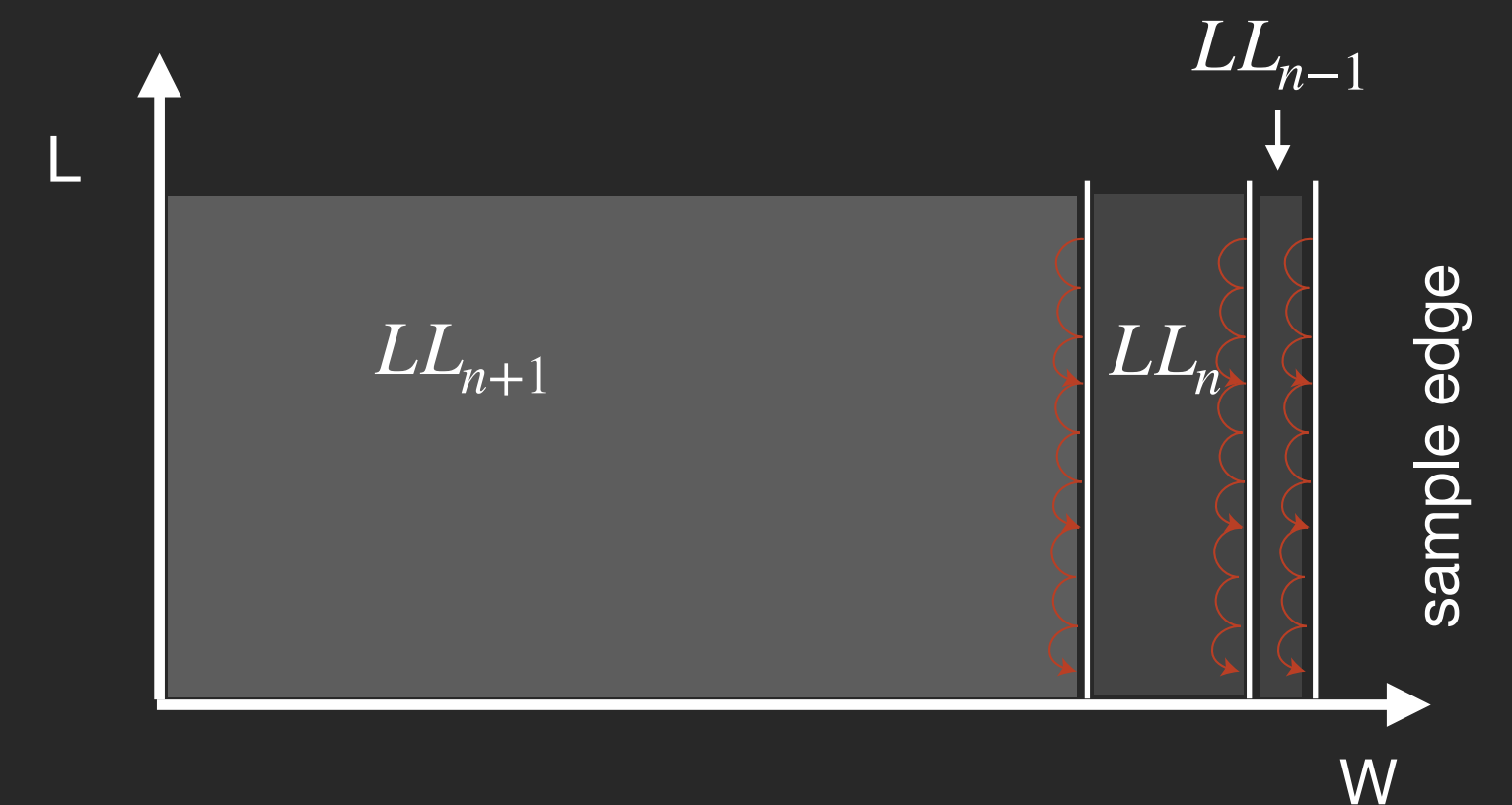
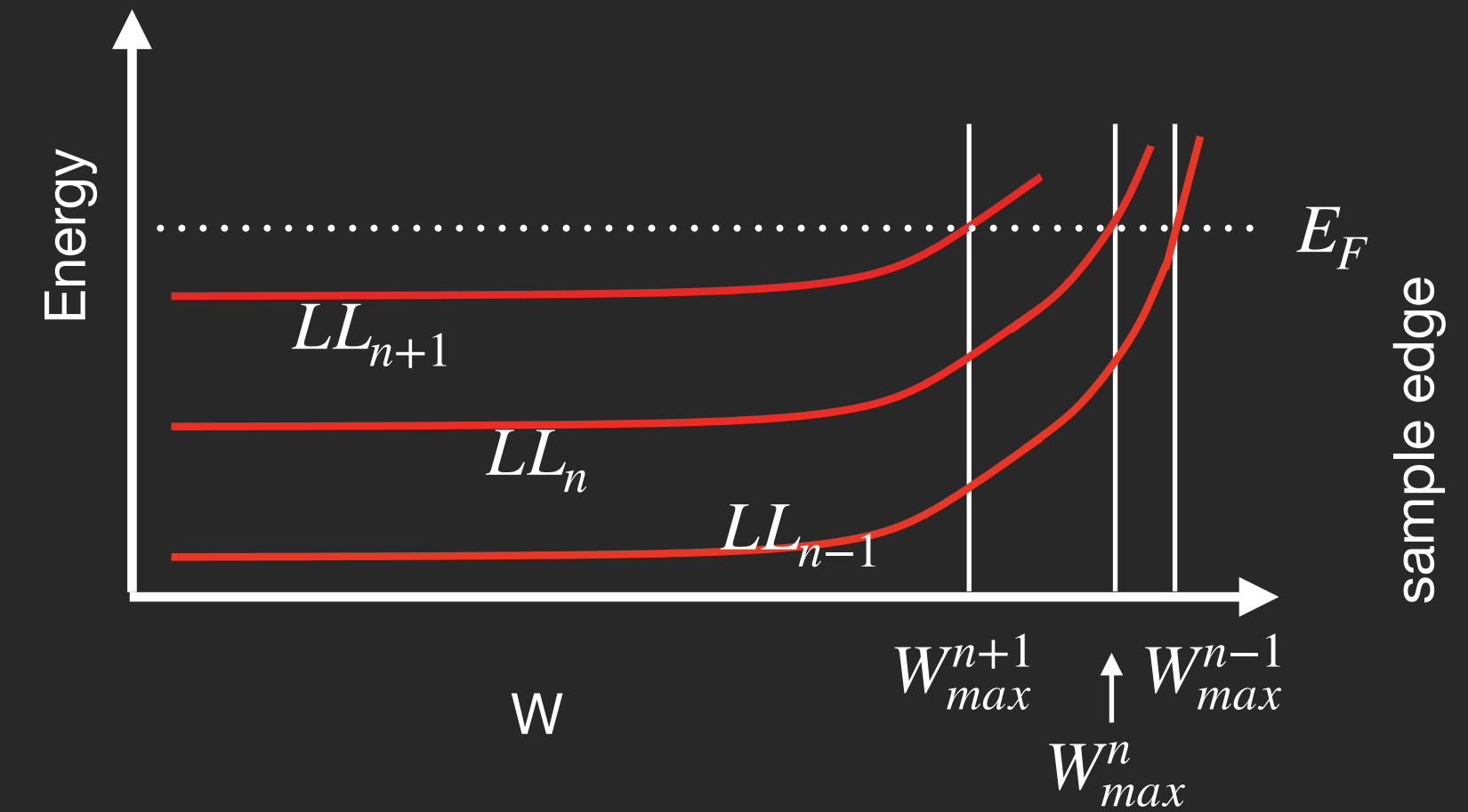
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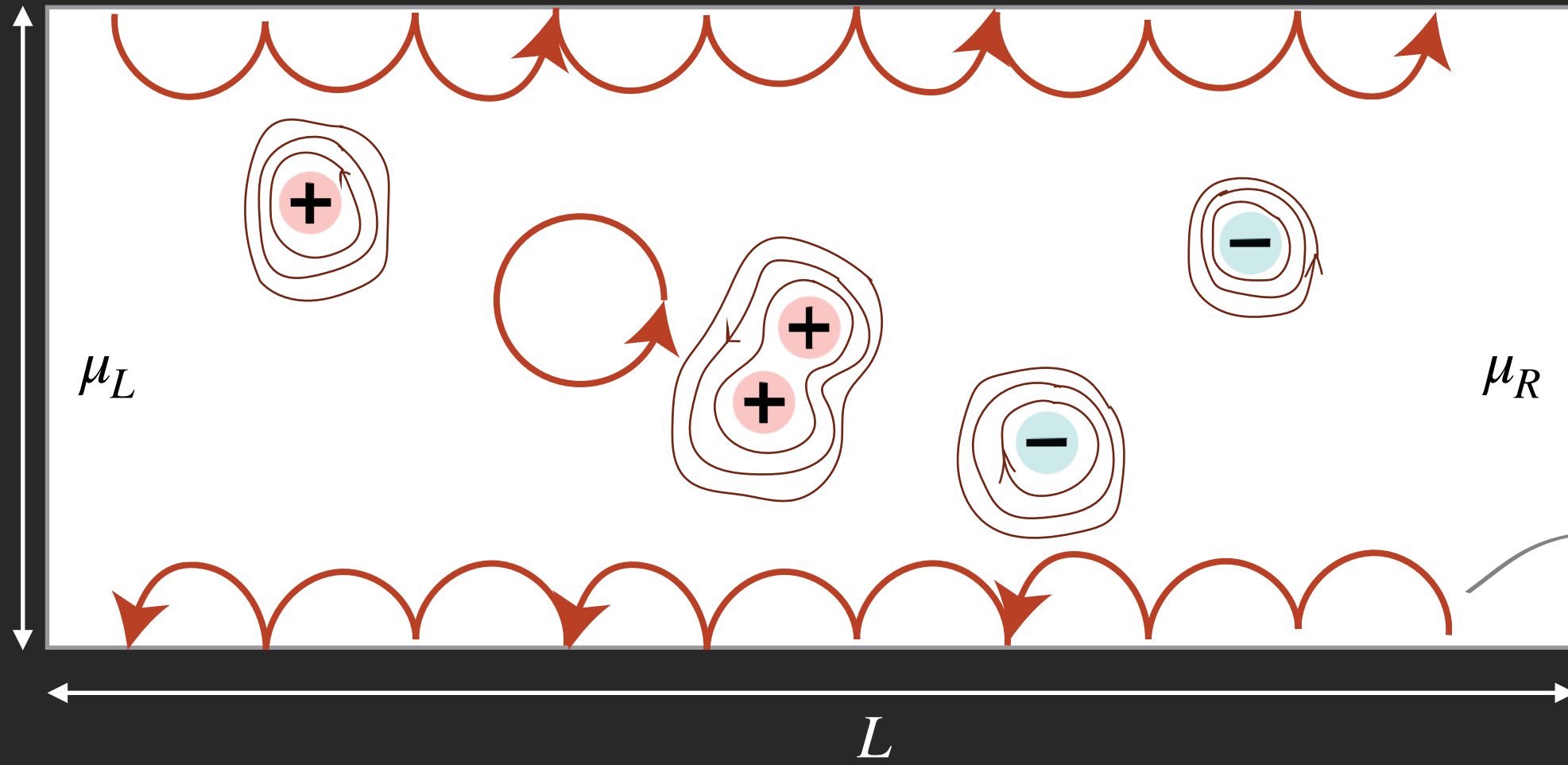
edge states

Edge-states due to sample-vacuum potential, and B-field.



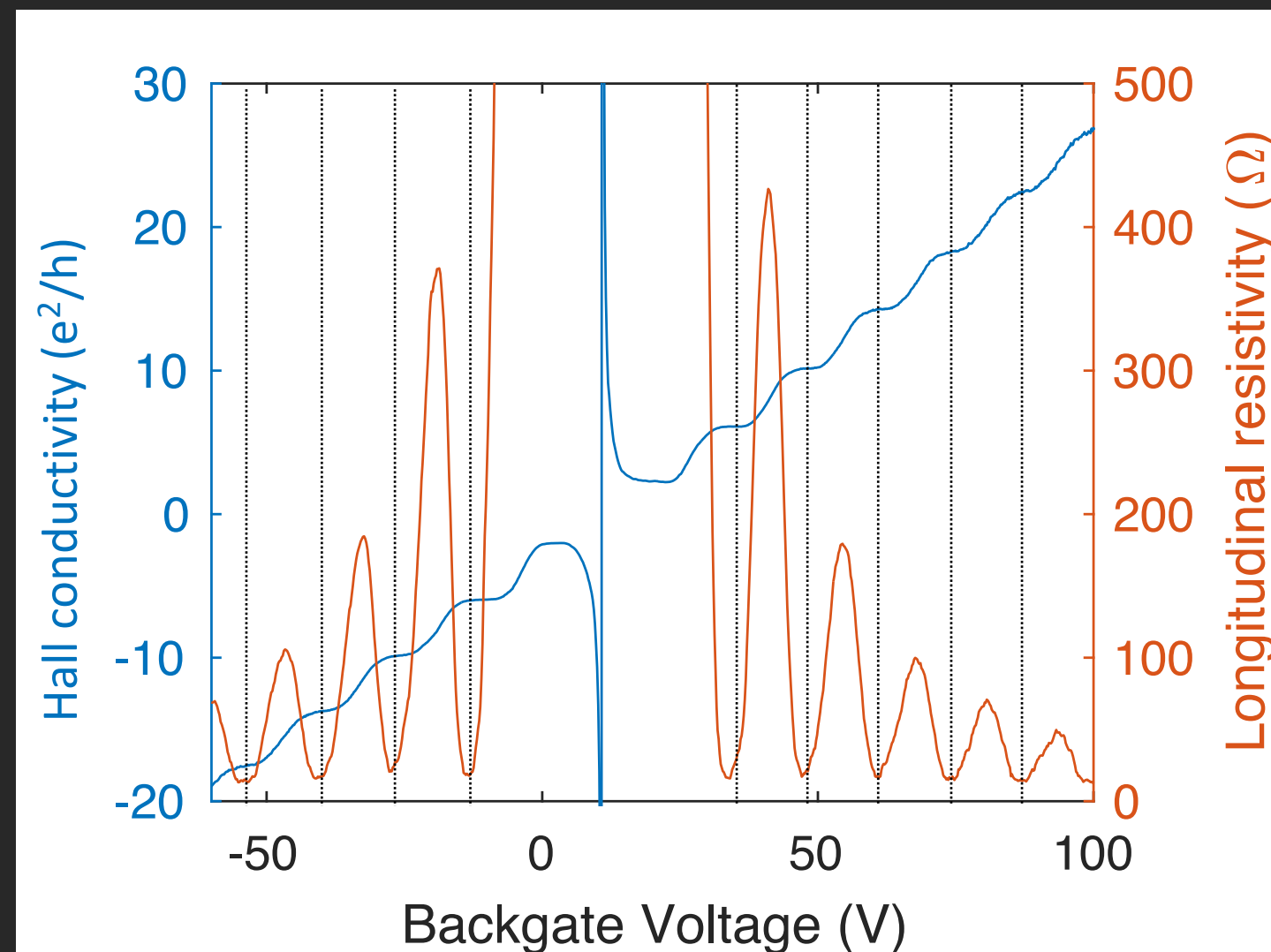
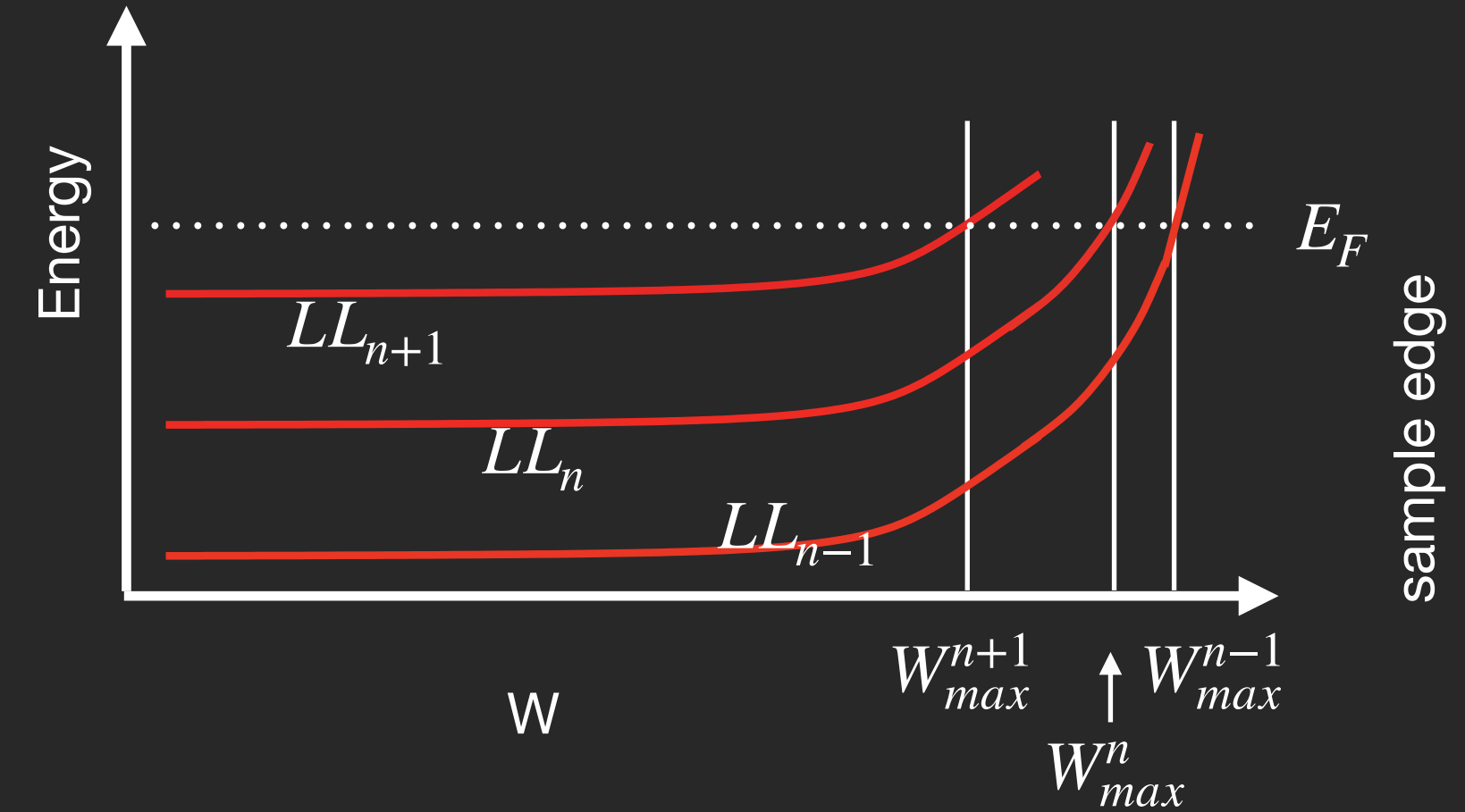
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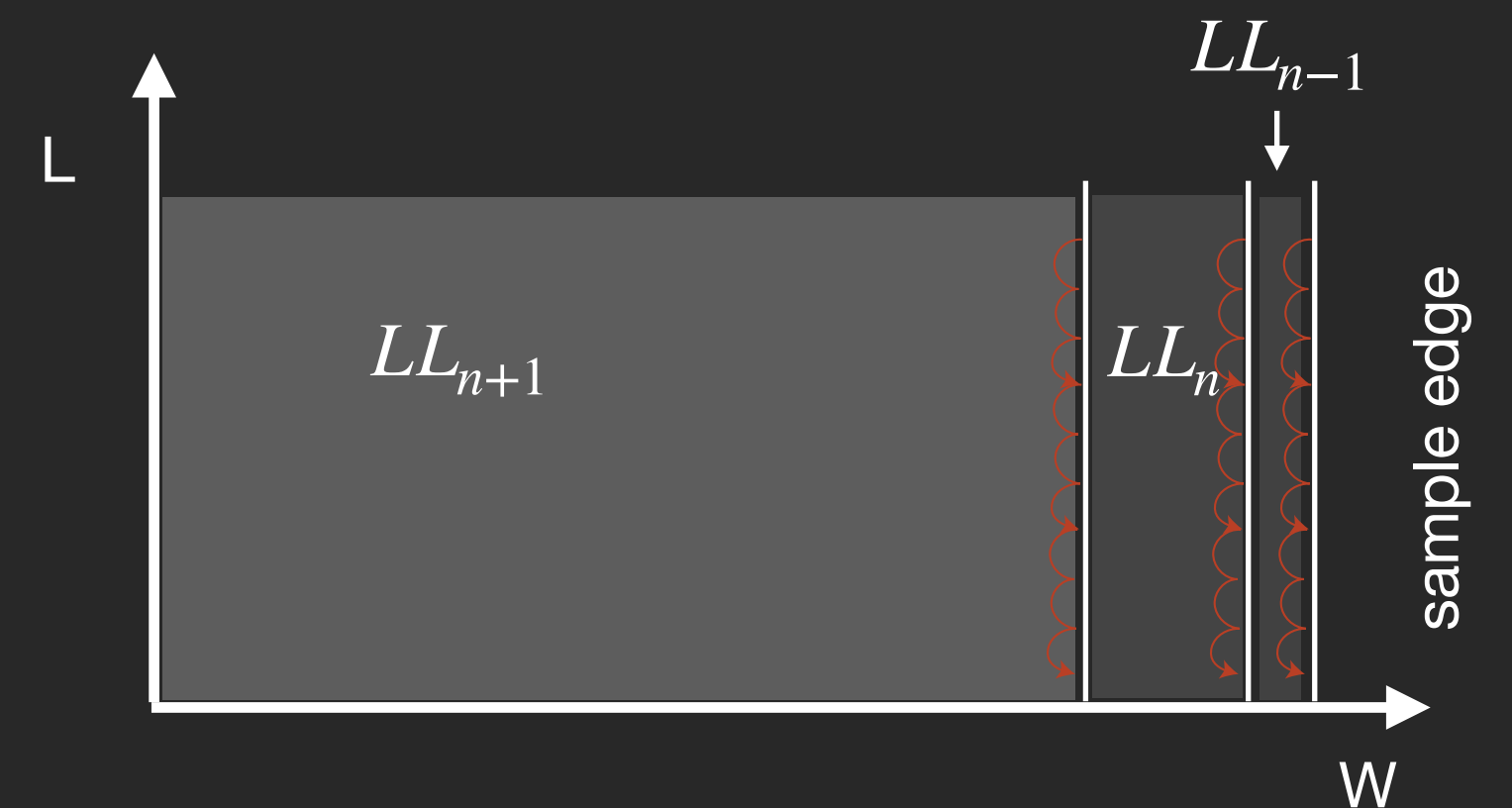


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Edge-states due to sample-vacuum potential, and B-field.



- The R_H plateaus are due to quantised DOS,
- The R_H slope is due to edge states.

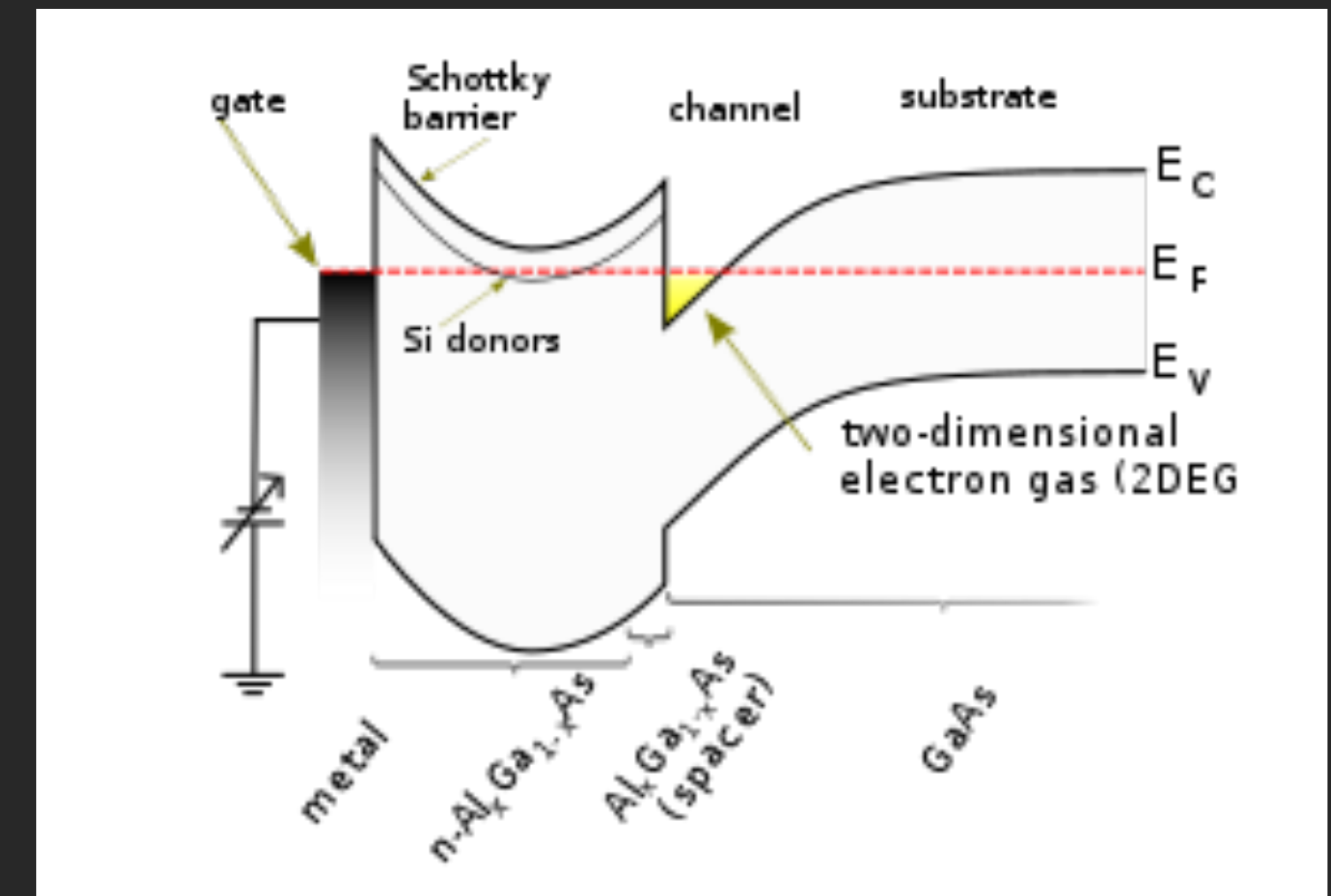


The III-V 2D Electron Gas (2DEG)

Initial integer quantum Hall (Stuttgart) - MOS structure

$\omega_c \tau > 1$ defines a critical magnetic field, $B_c \approx \frac{m}{e\tau} = \mu^{-1}$

With current III-V materials $\mu > 10^7 \text{cm}^2/\text{Vs}$ $B_c \approx 1 \text{mT}$



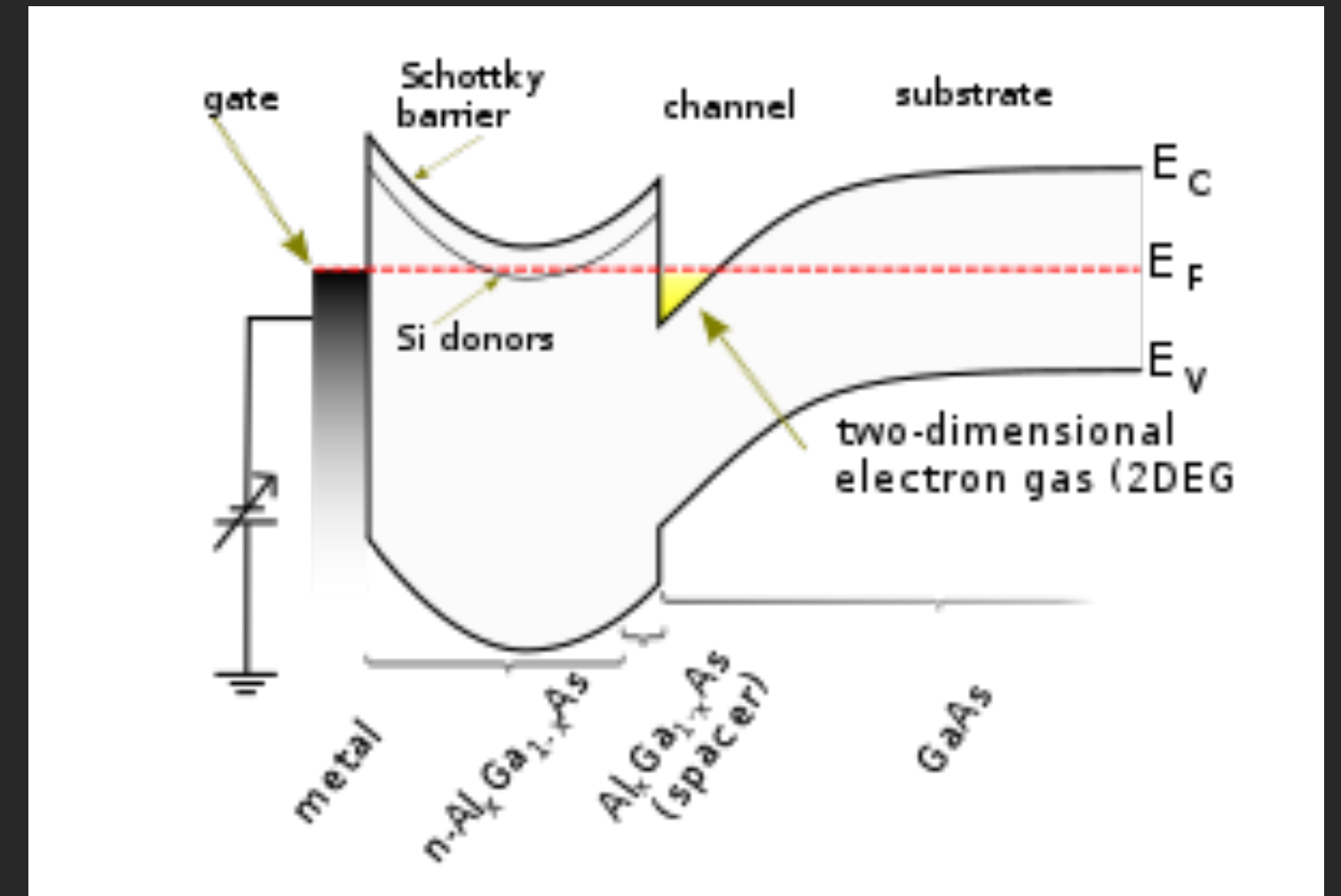
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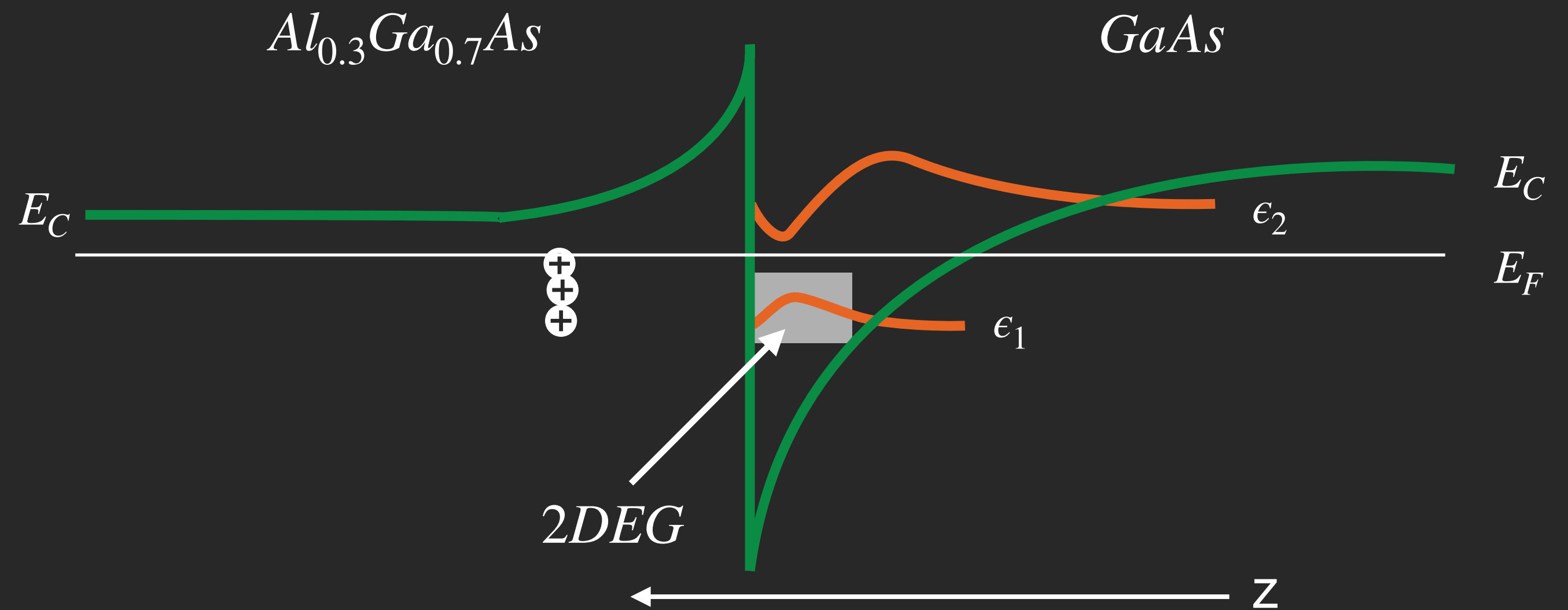
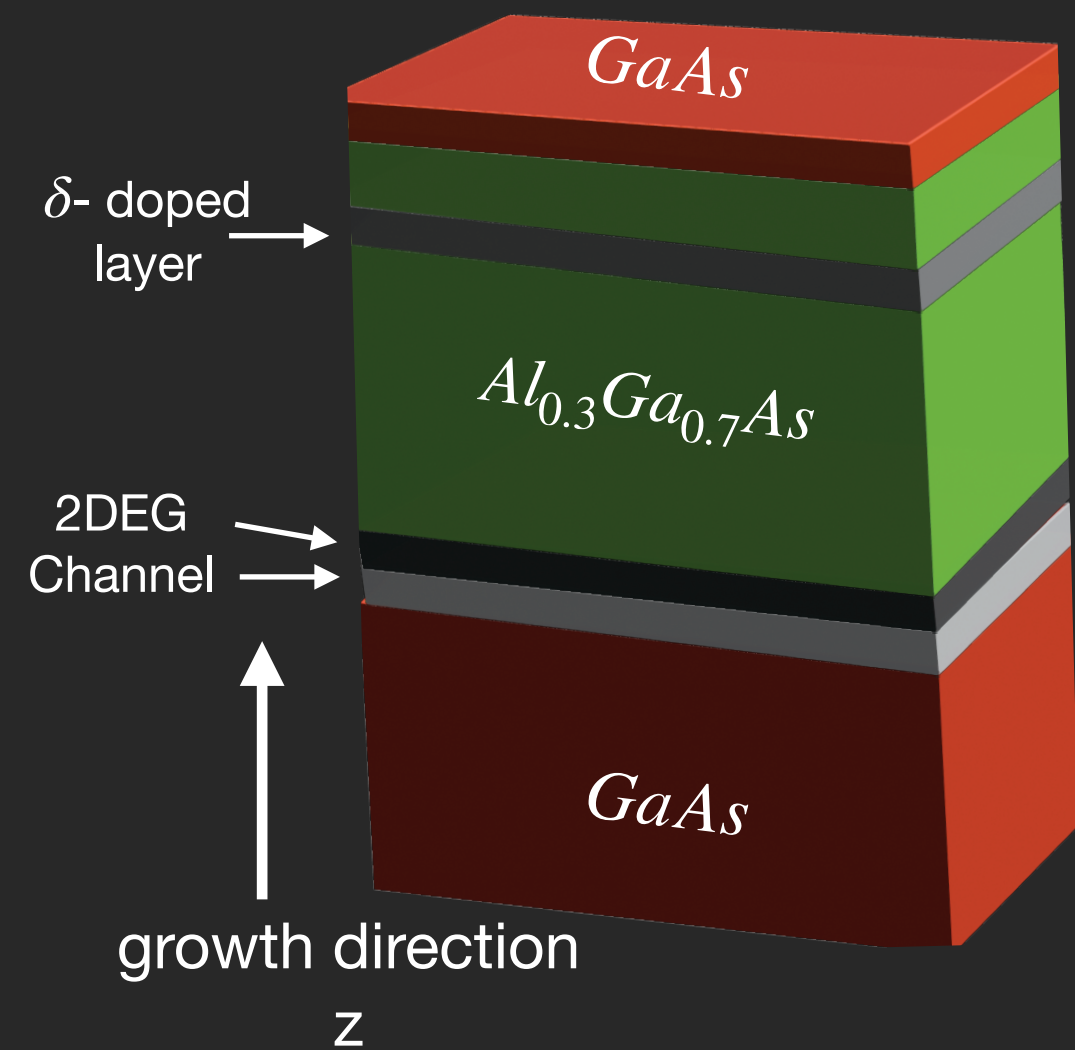
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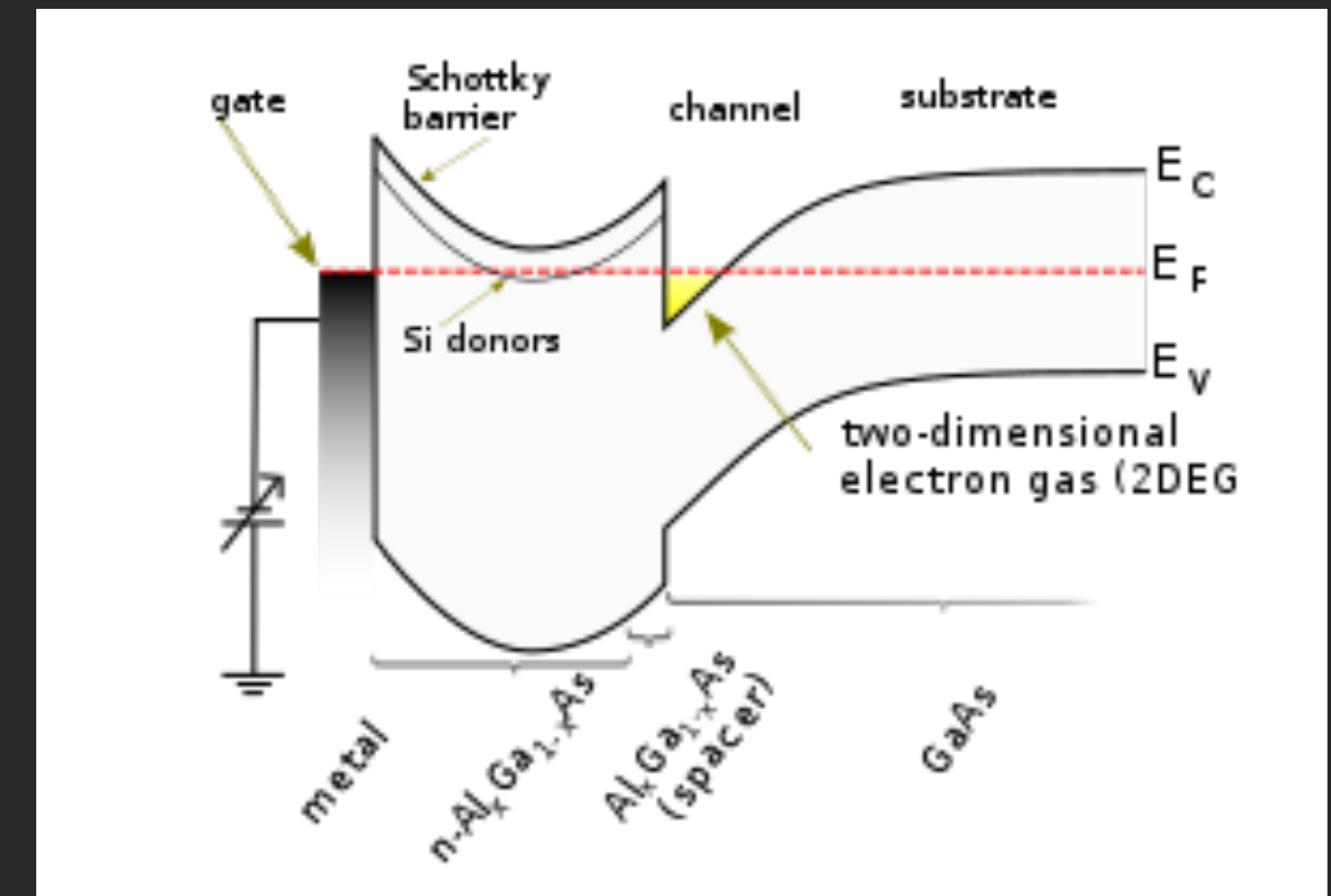


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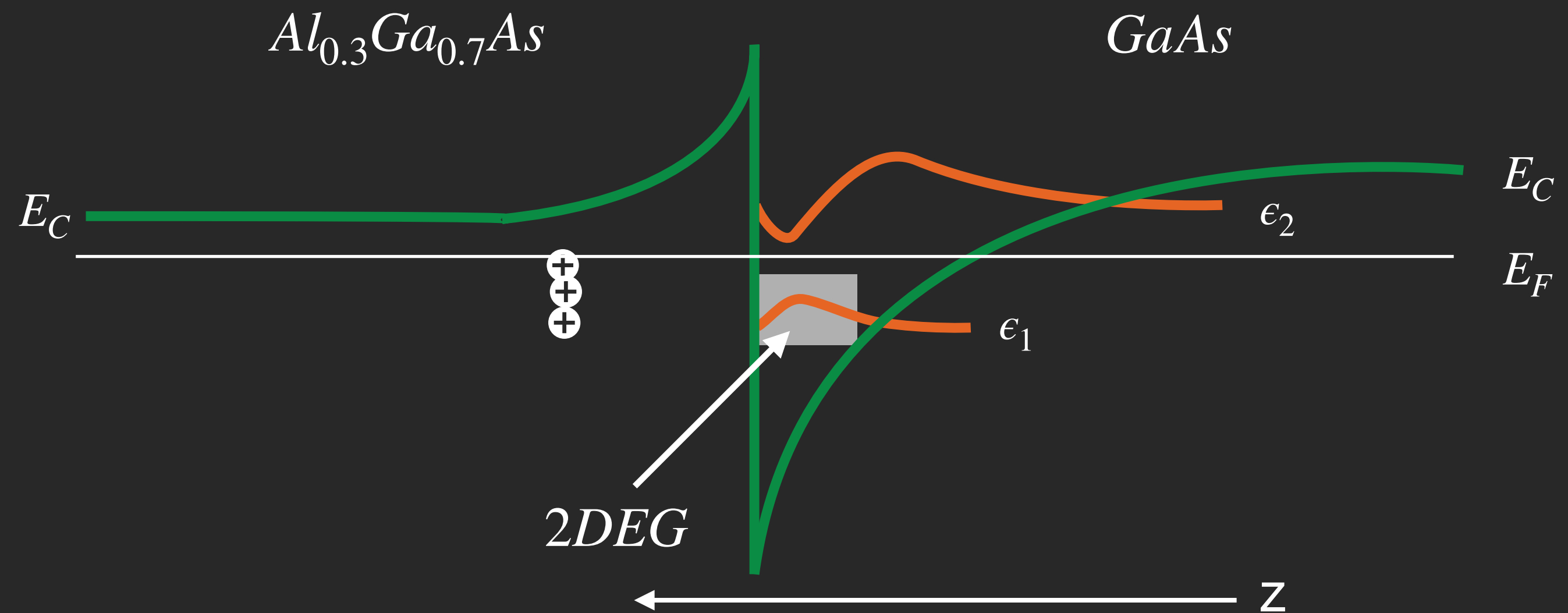
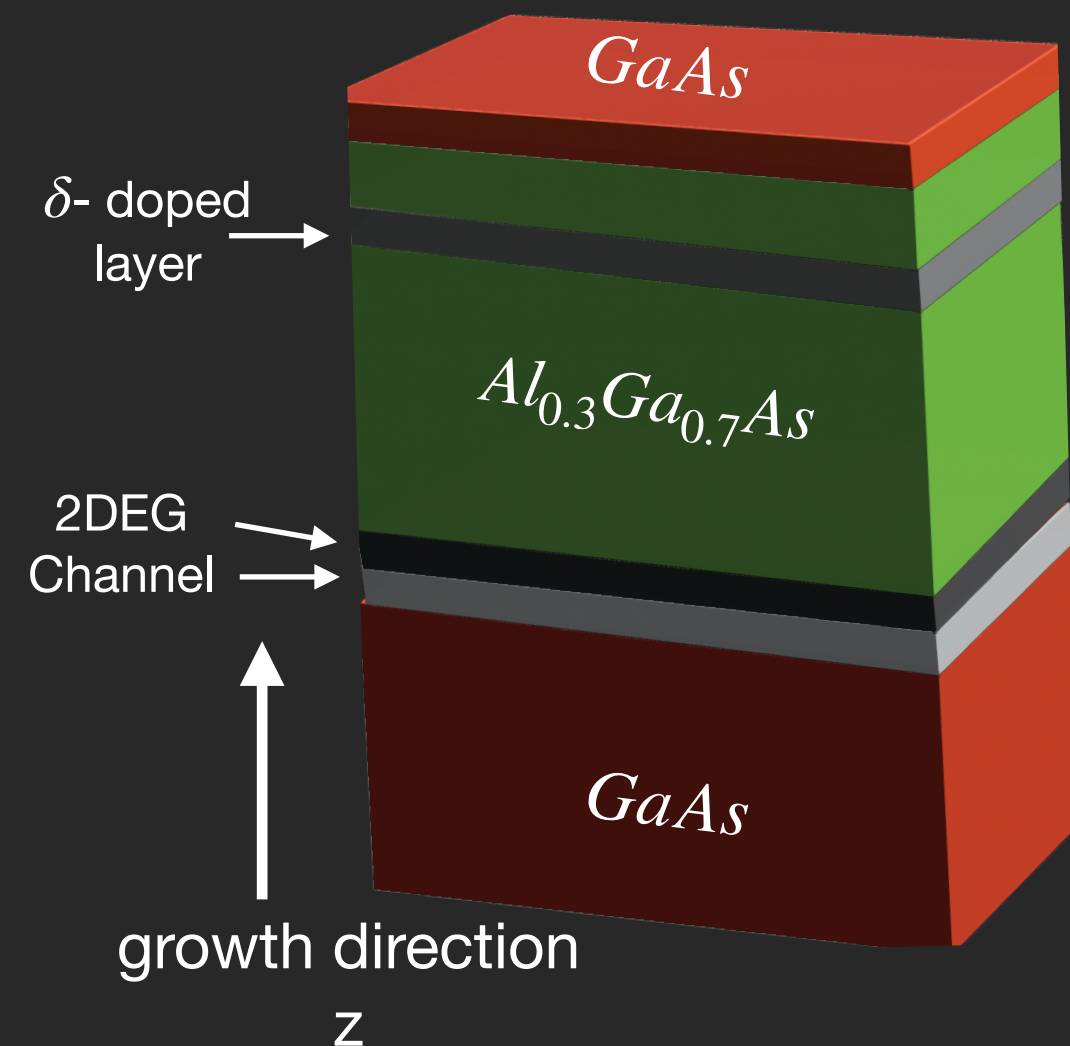
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Let's look at a different way to make a 2-dimensional electron gas: Using Graphene

Part II: Graphene

Allotropes of carbon

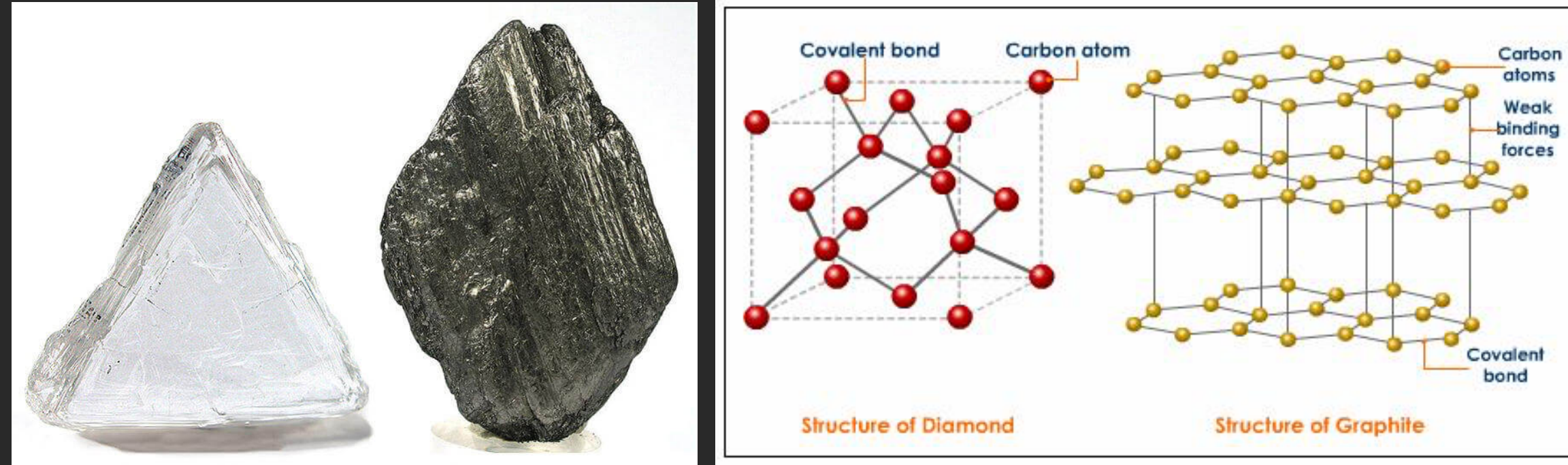


all images: www.wikipedia.com

- Two “natural” forms of carbon—they are 3 dimensional:
Diamond and **Graphite** have been well known for ages.
One is one of the hardest of materials, the other, one of the softest.

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Allotropes of carbon

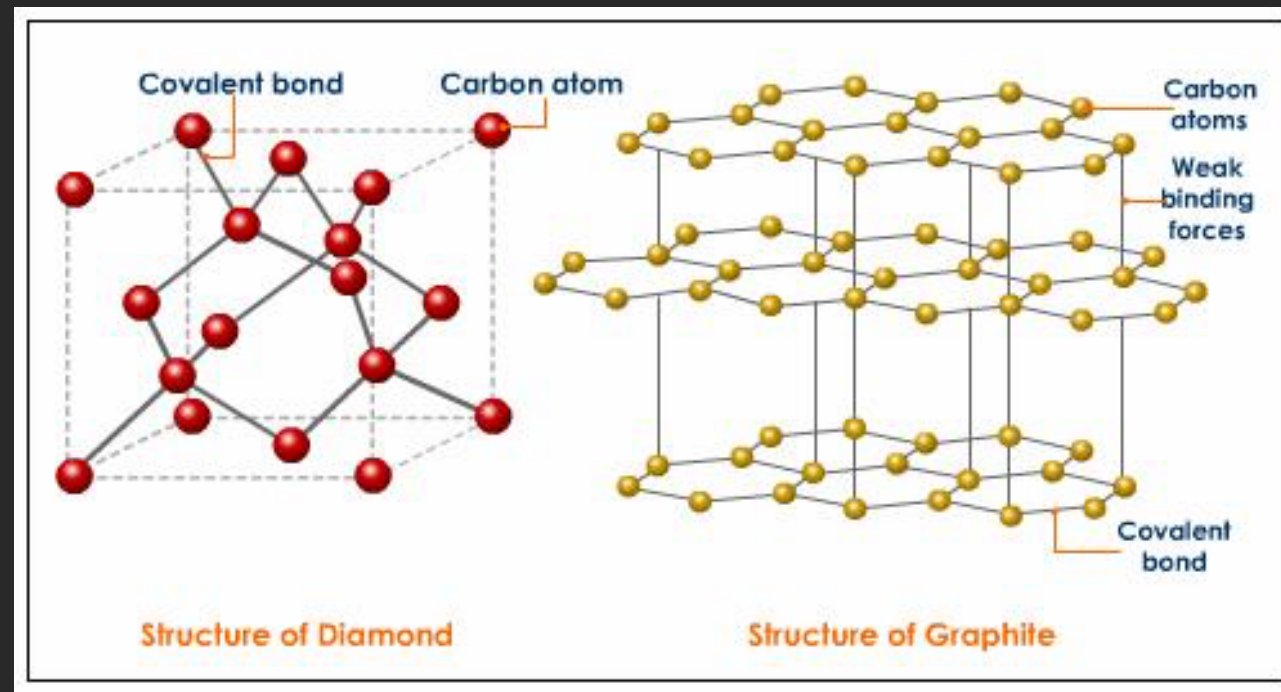


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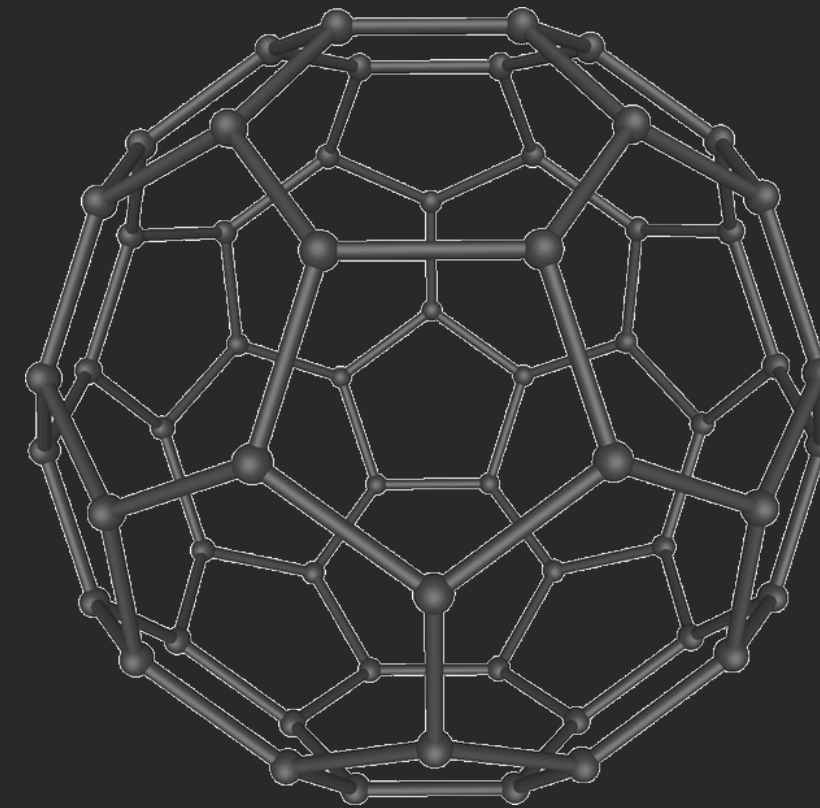
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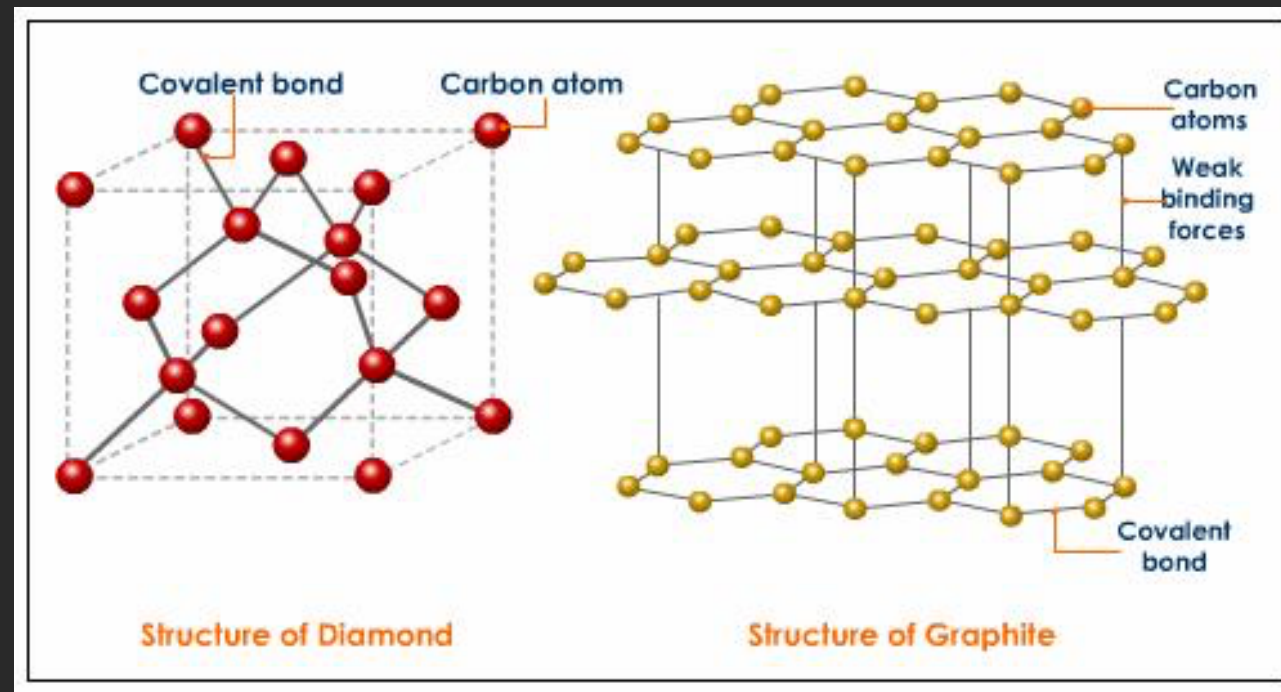
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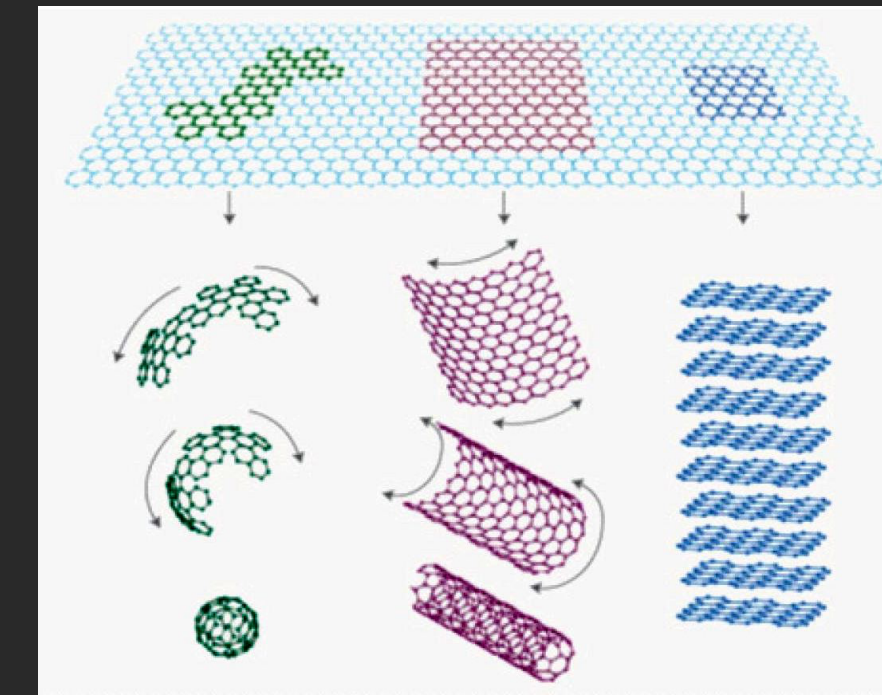
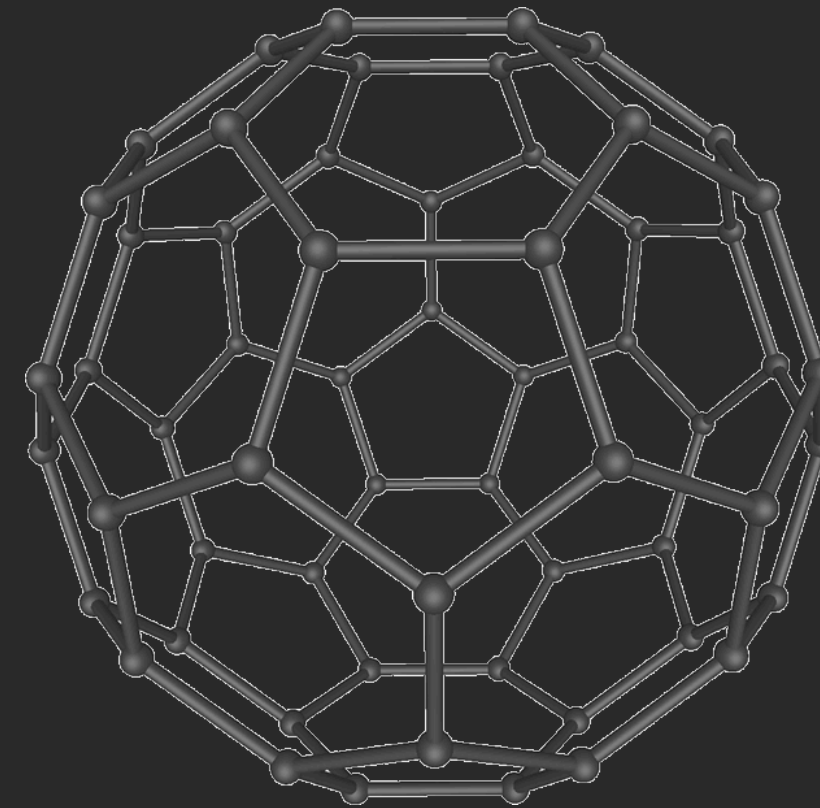
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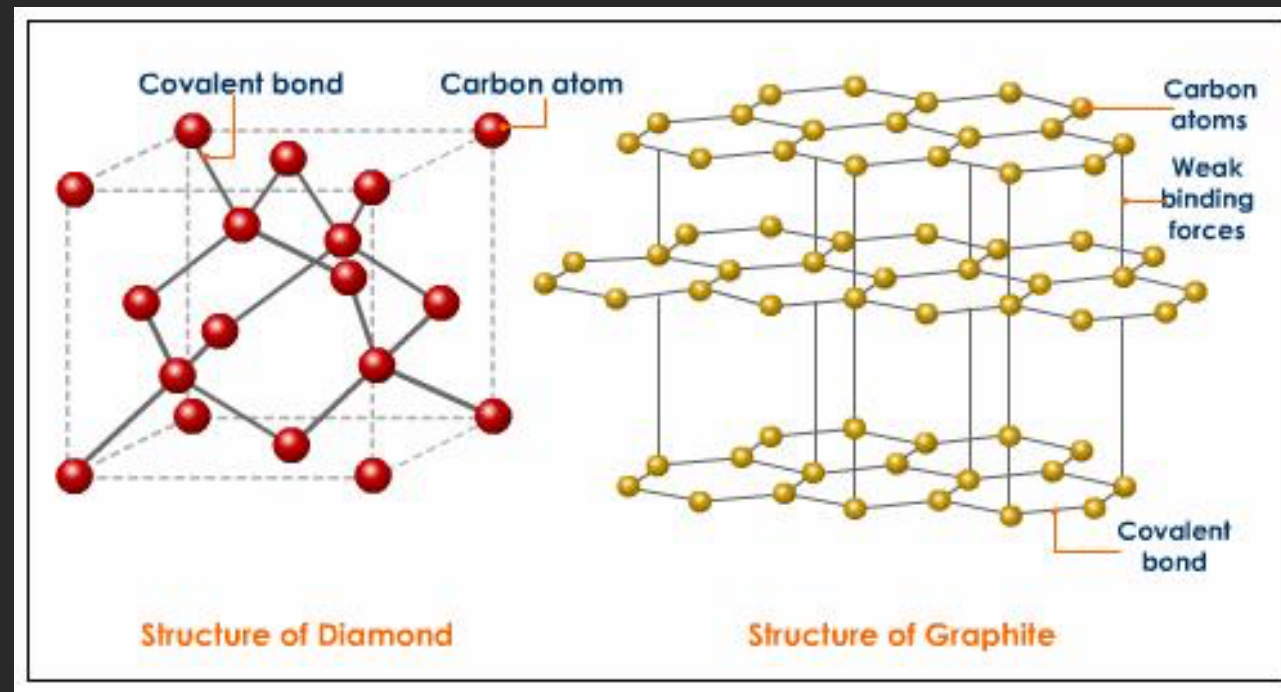
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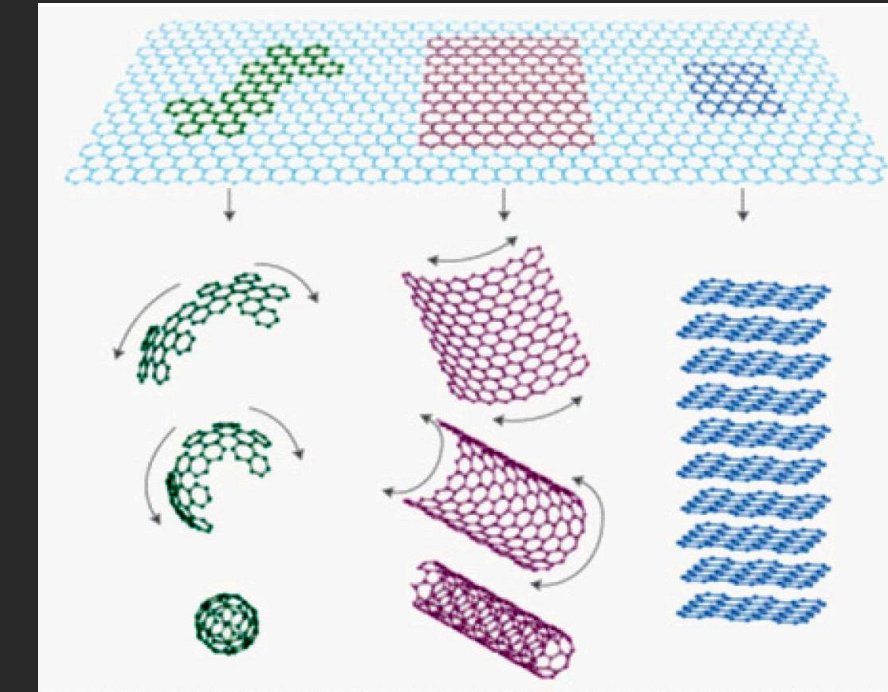
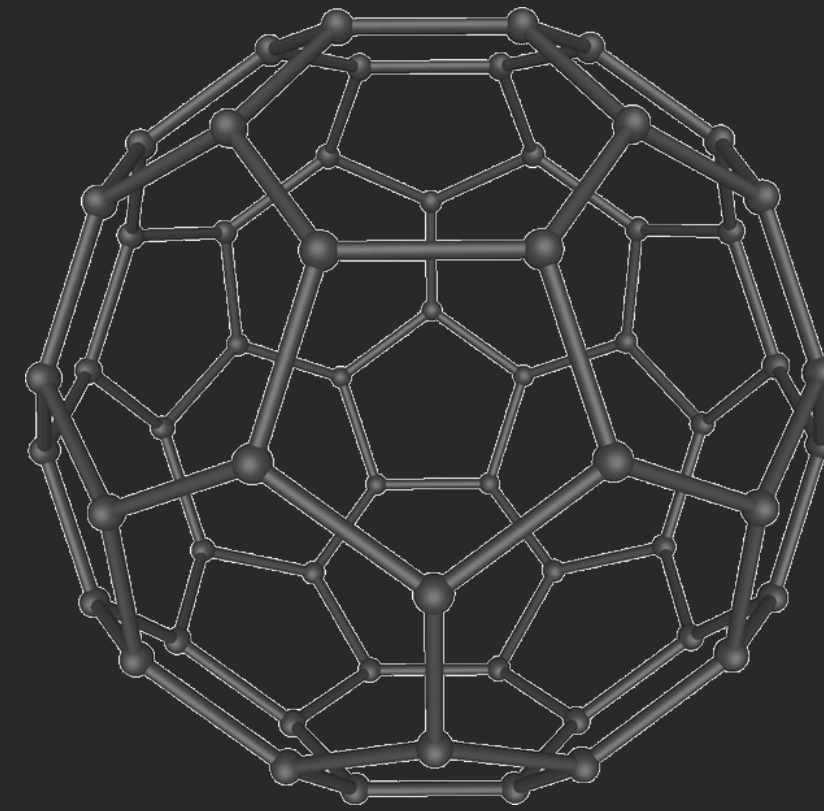
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- Expect for diamond (sp³), all allotropes are based on a 2D layer, we know as graphene.

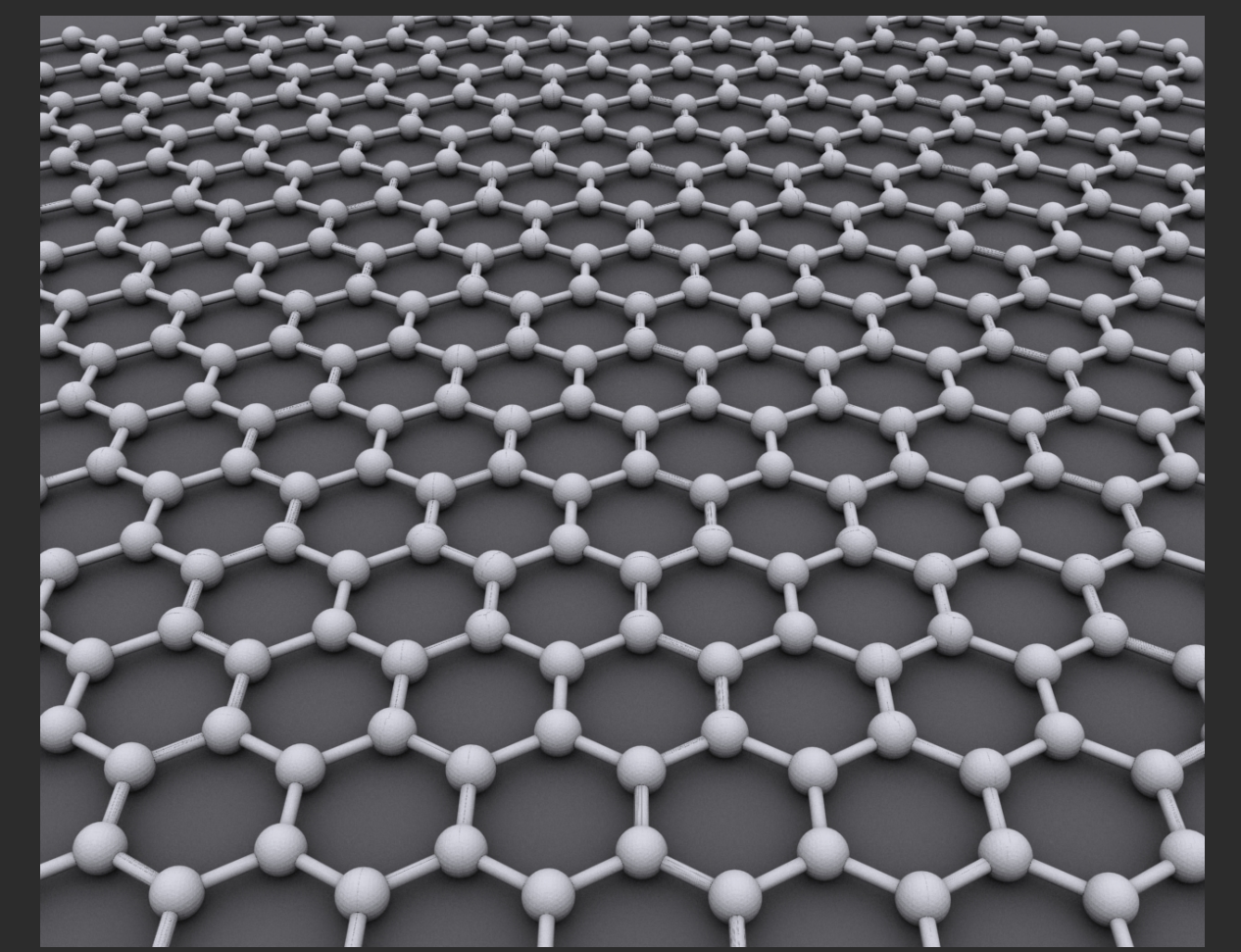
The curious case of graphene

An isolated single 2D layer, graphene, was not seen until 2004.

K. S. Novoselov, *et al.*, *Science*. **306**, 666 (2004).

Most believed 2D layers were not stable against thermal vibration.

sp^2 hybridise, plane bonding. Remaining p -orbital out of plane



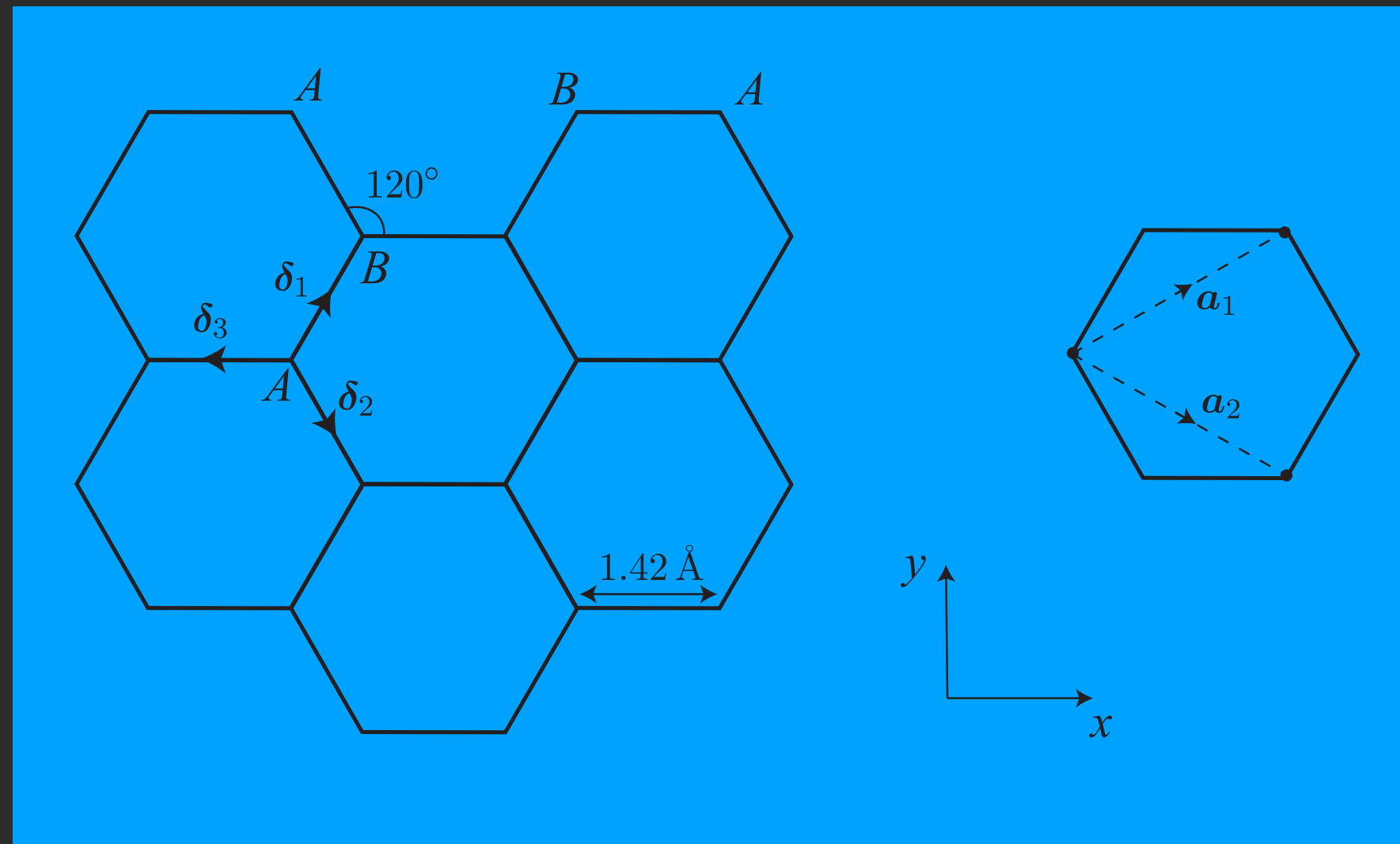
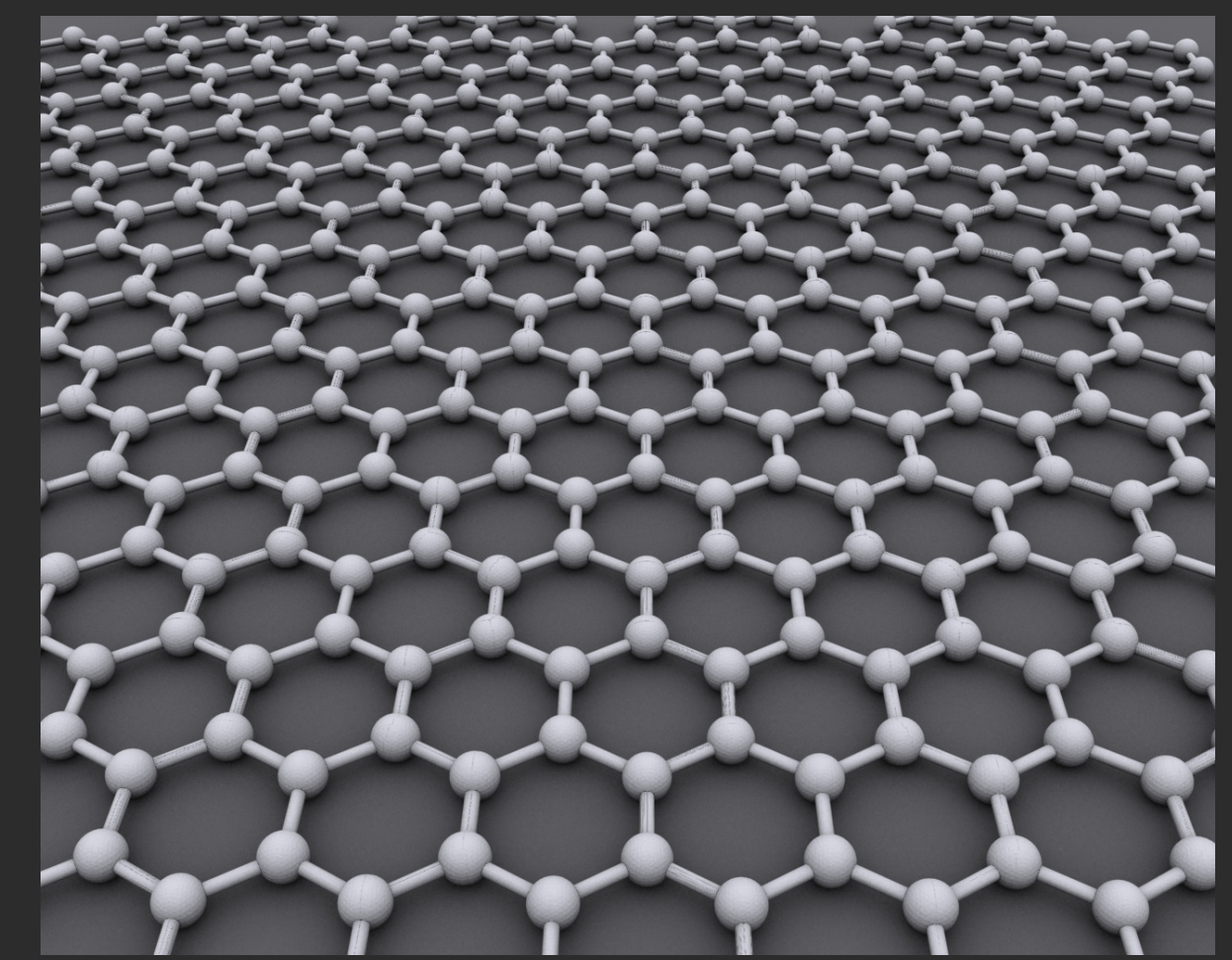
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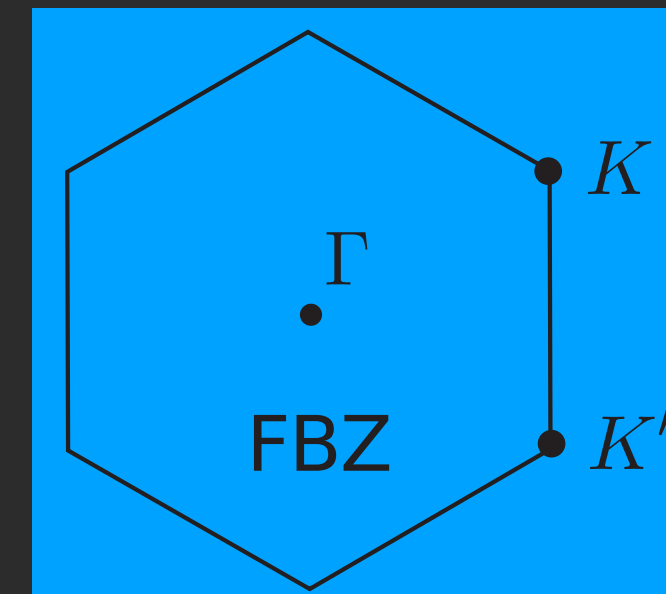
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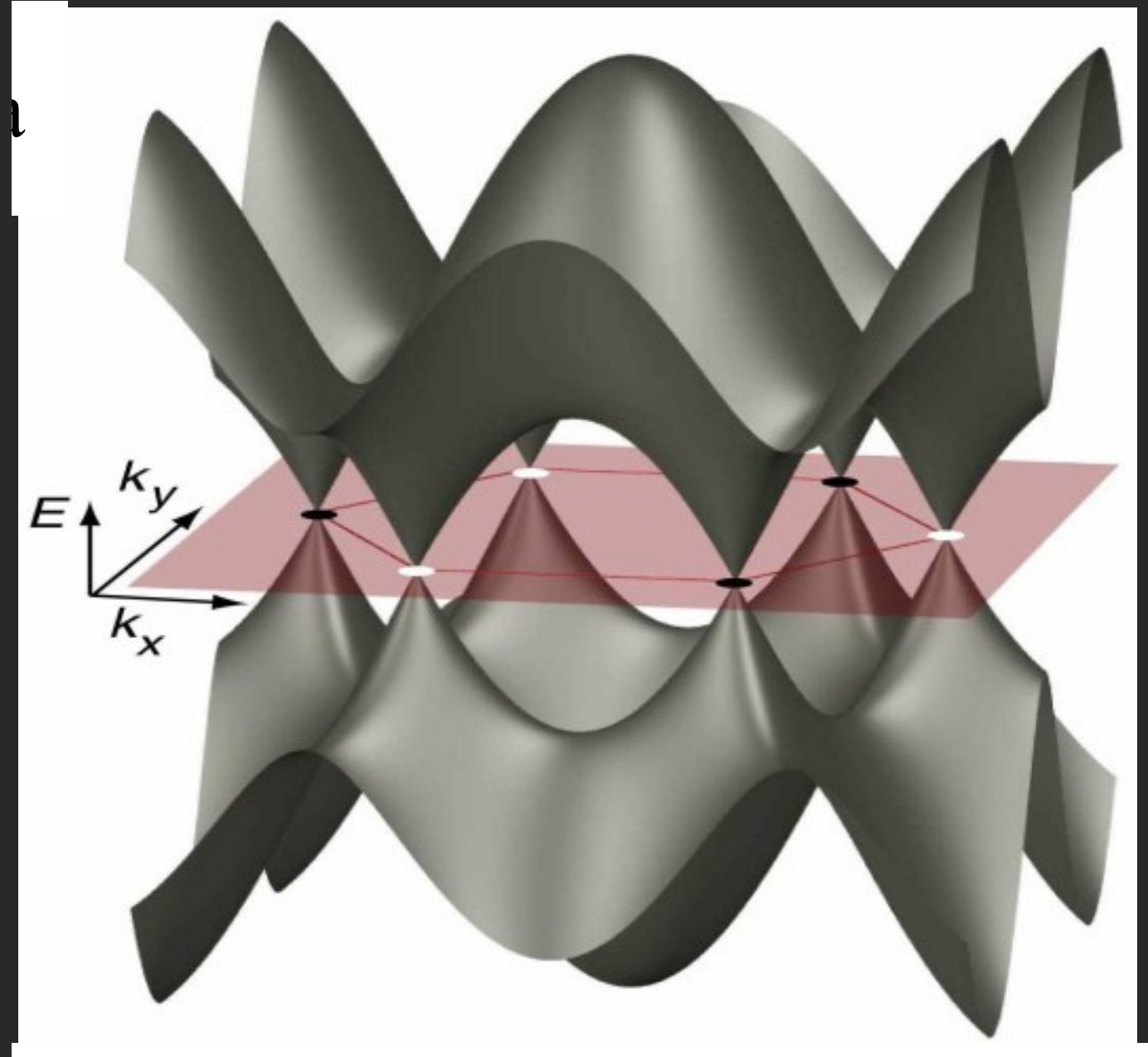
Two non-equivalent sublattices, A and B, are mirror symmetric.



Reciprocal lattice — First Brillouin zone (FBZ),
Each sublattice point, K and K' in FBZ are :

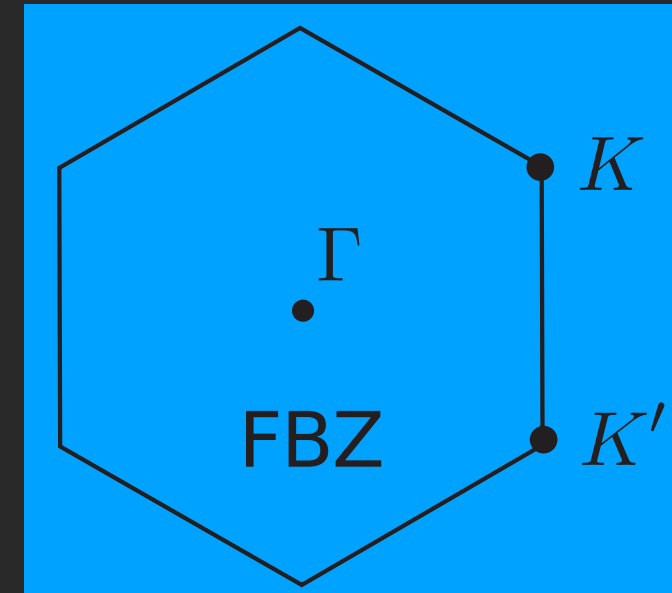
$$K = \frac{2\pi}{3a} \left(1, \frac{1}{\sqrt{3}}\right), \quad K' = \frac{2\pi}{3a} \left(1, -\frac{1}{\sqrt{3}}\right)$$

Graphene band structure



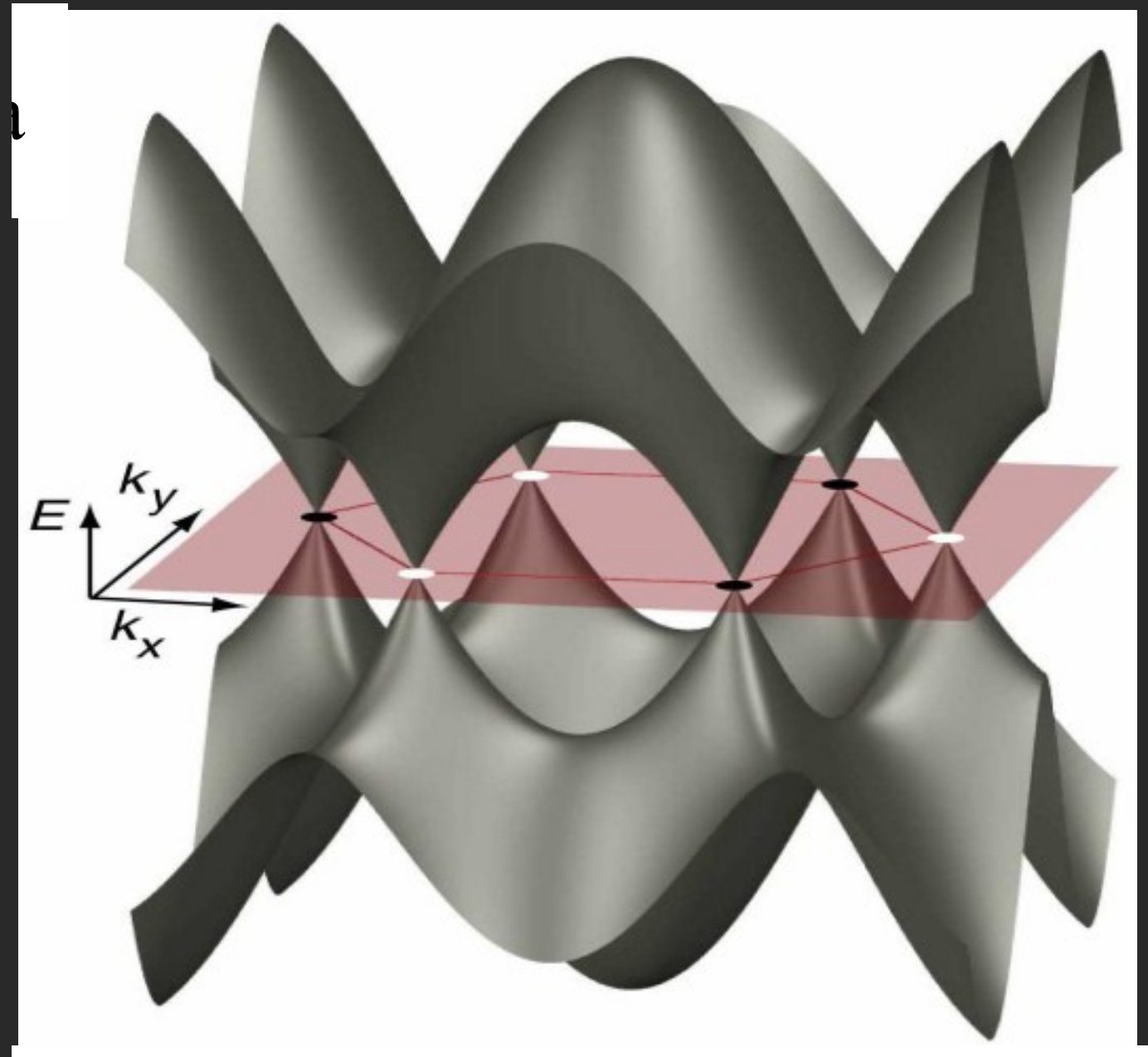
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C.W.J. Bennaker, Rev.Mod.Phys., 80, 1337 (2008)



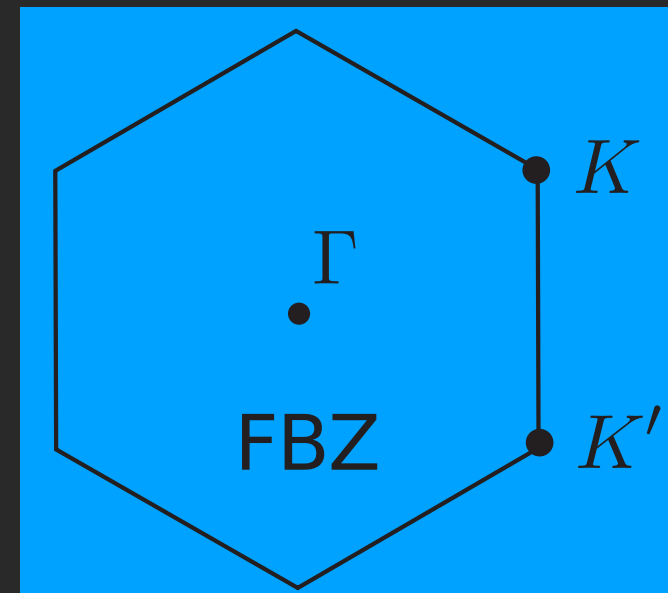
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- With no doping, no DOS at E_f .
- A perfect semi-metal.

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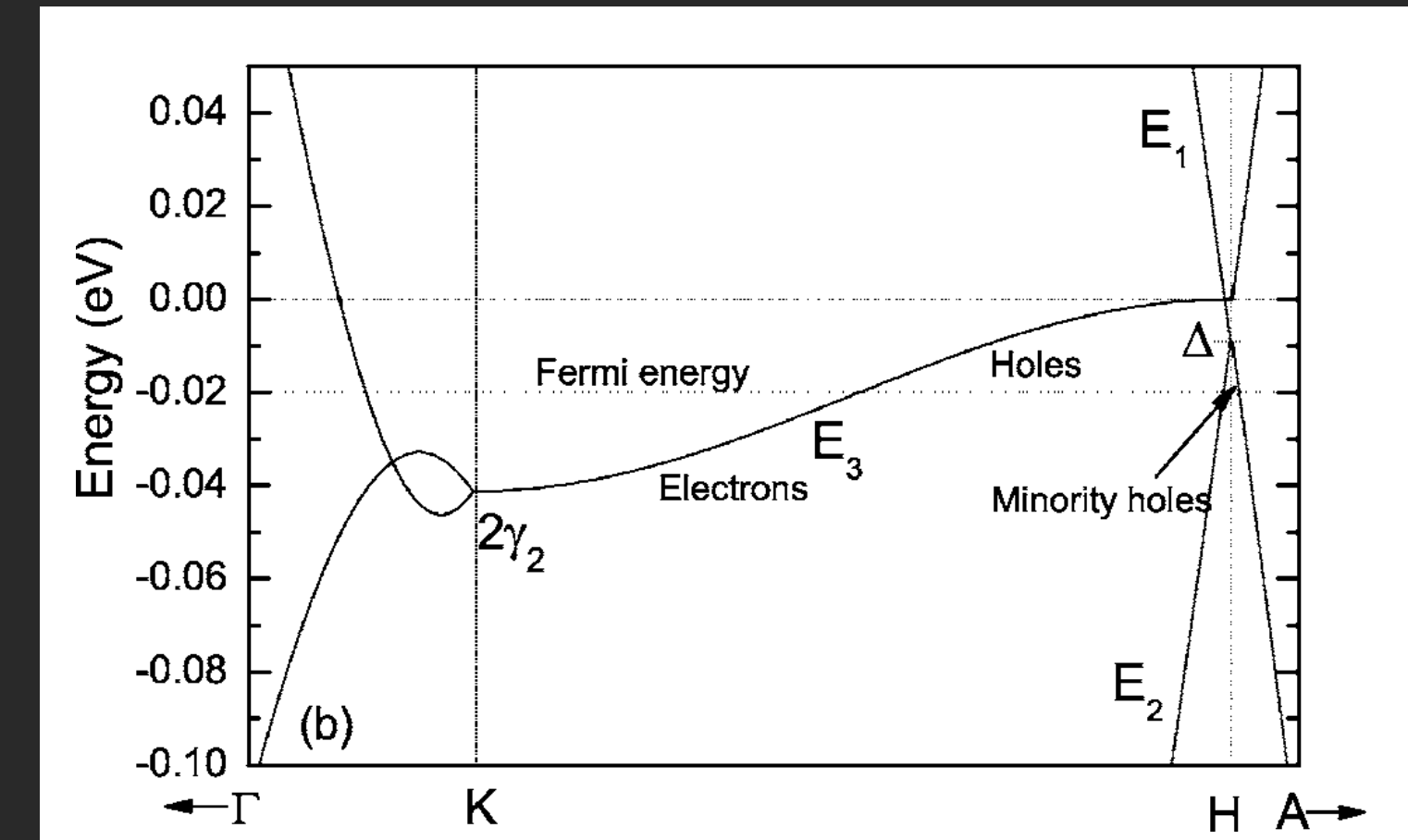
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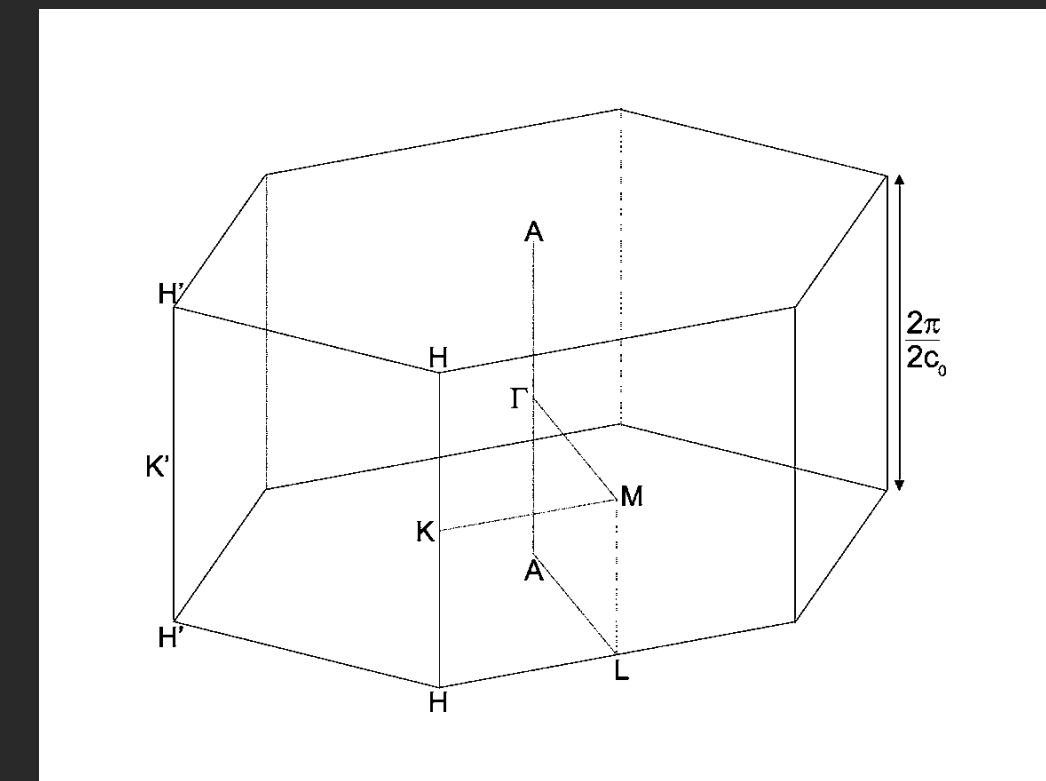


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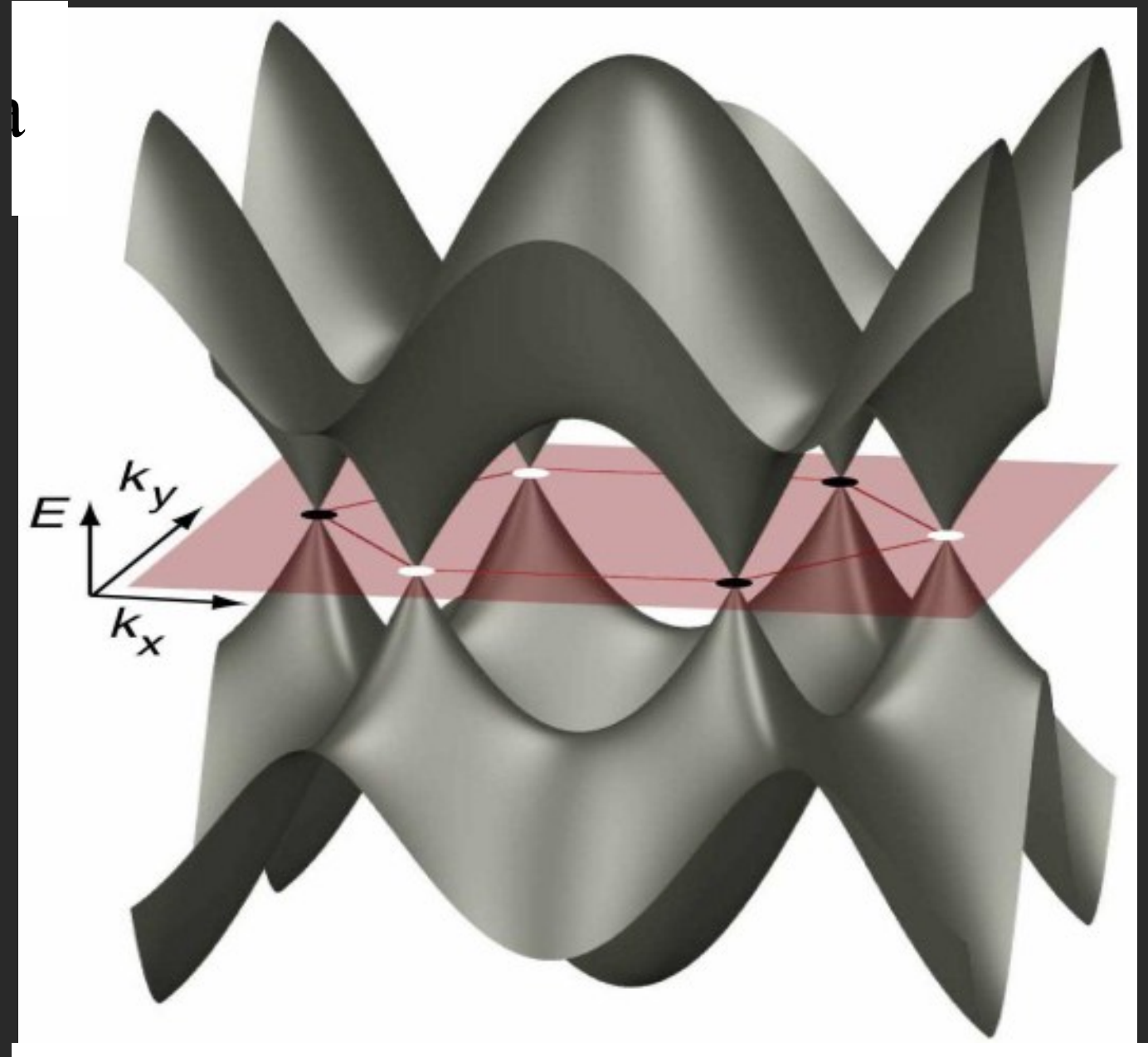
Reference: Graphite (bulk)



Partoens & Peeters, PRB, 74, 075404 2006

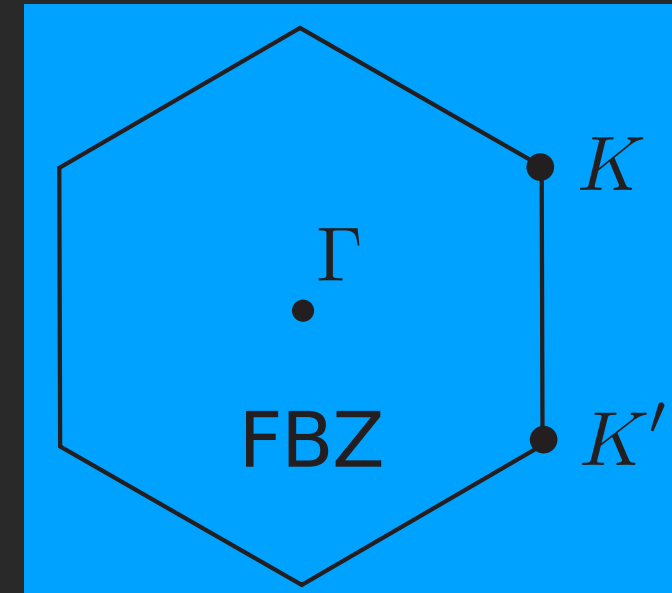


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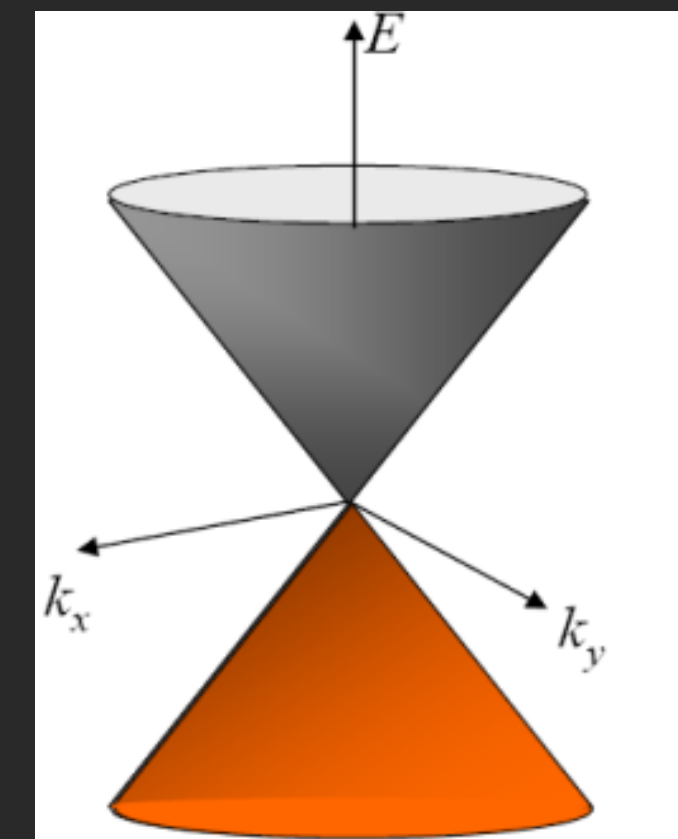
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$$\epsilon(q) \approx \hbar v_F(q_x - iq_y) + 0 (1 + q/K)^2; \quad v_F = \frac{3ta}{2} \approx 10^6 \text{ m/sec}$$

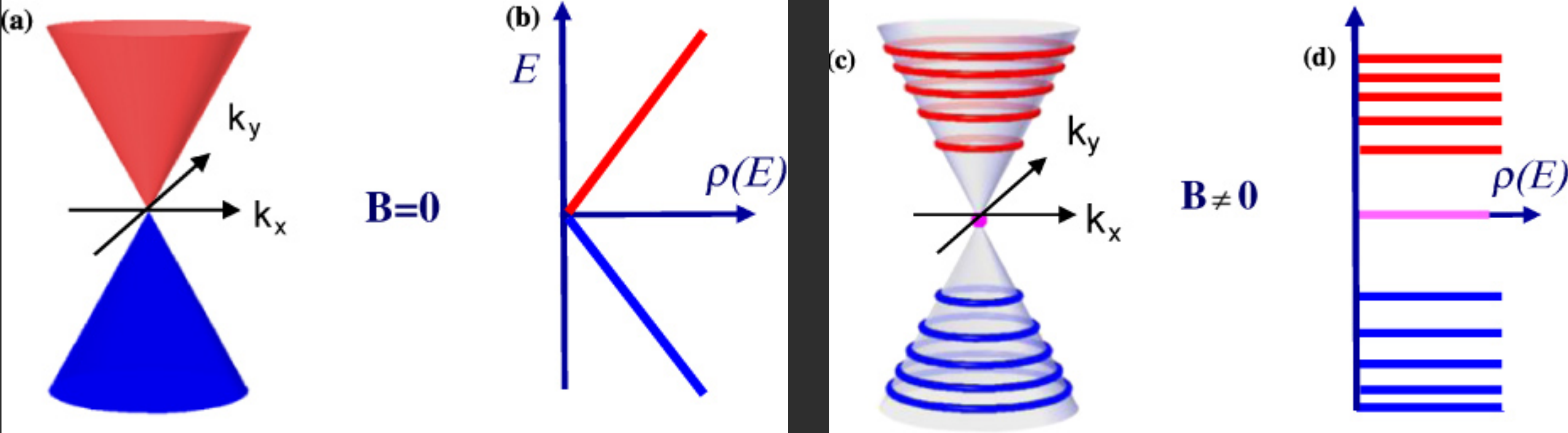
- Then the dispersion (wrt q) near K is constant
- No band curvature.



Part III: Integer quantum Hall physics in graphene

- Add a perpendicular magnetic field, B
- Eigenenergies:

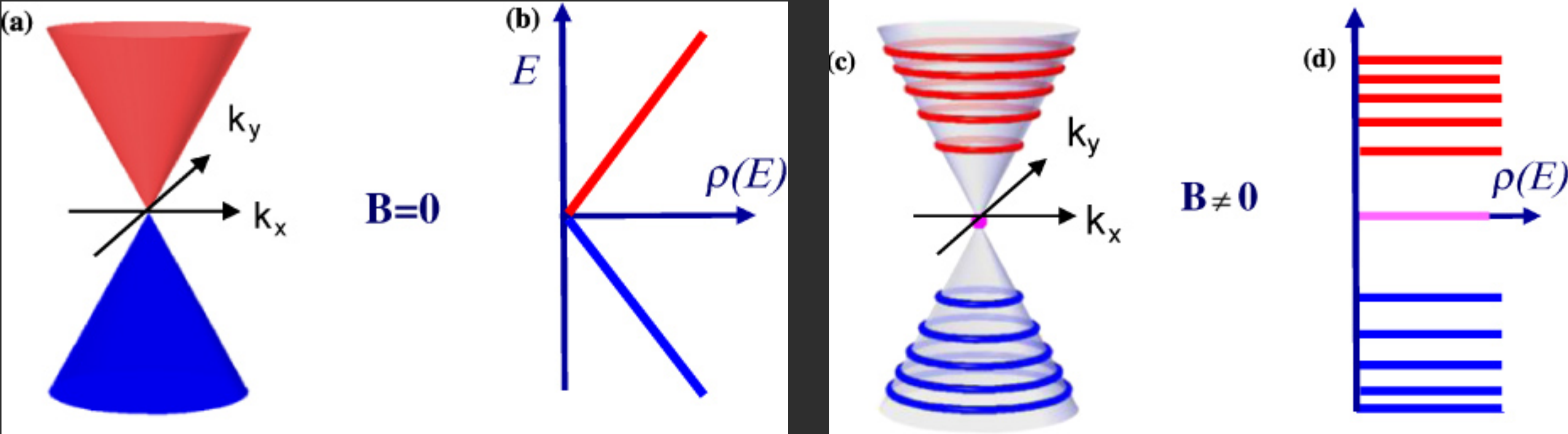
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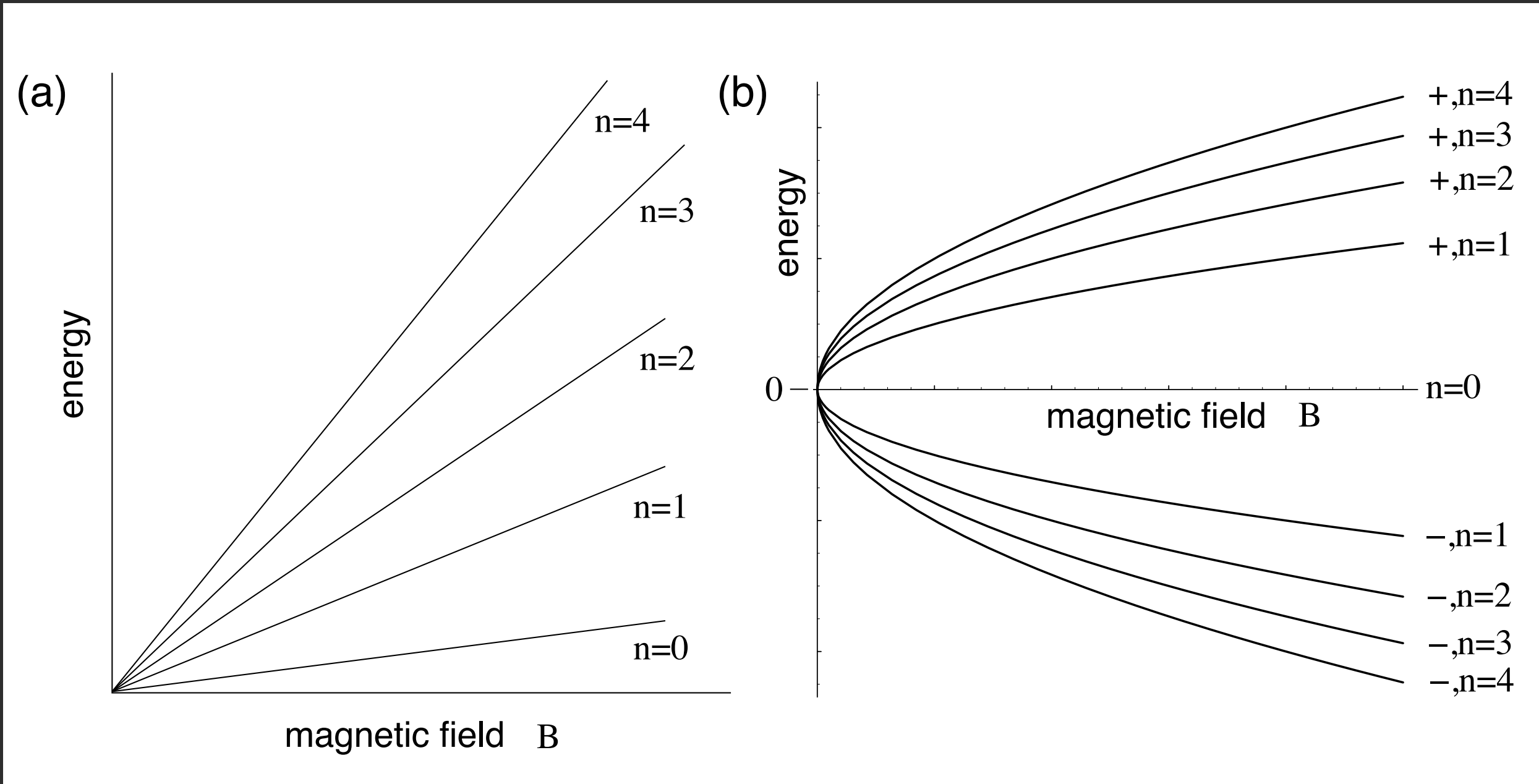
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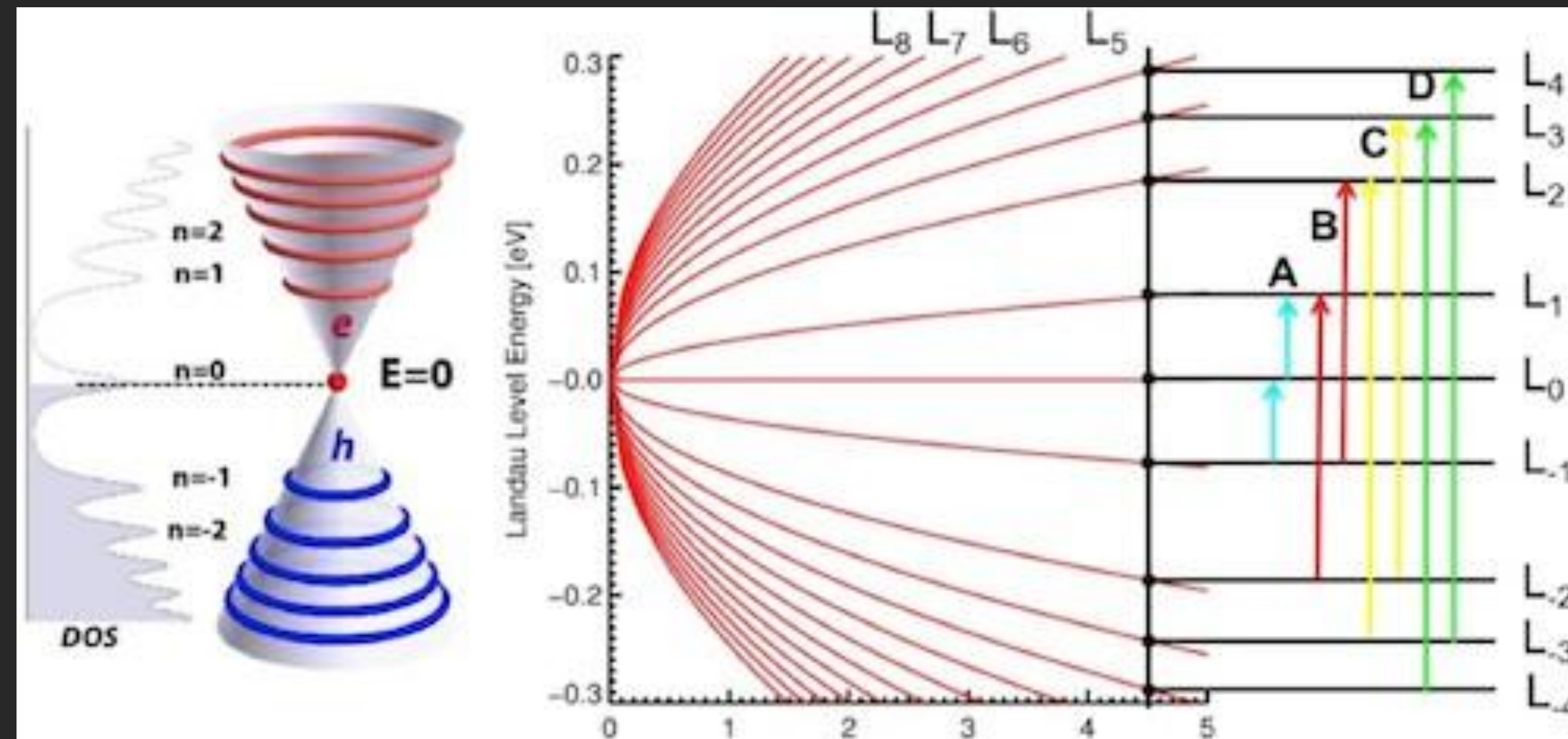
Parabolic band semiconductor

Dirac semiconductor

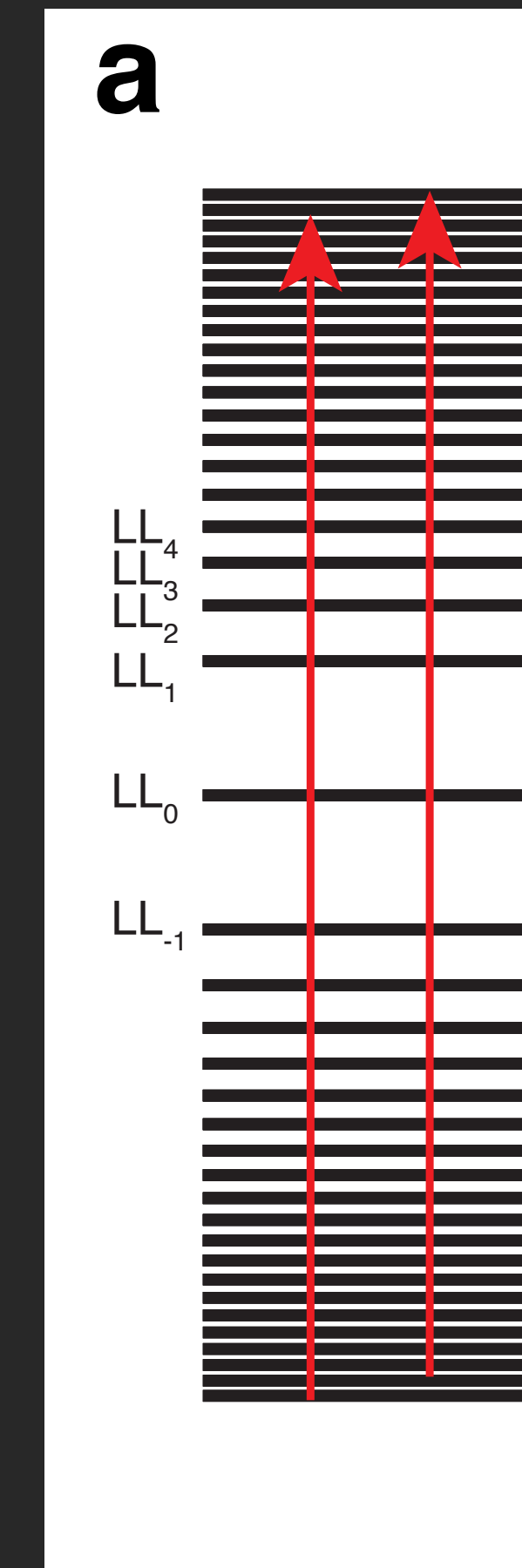
- *Normal* semiconductors:
- $\epsilon_n = \hbar\omega_c (n + 1/2) \propto B (n + 1/2)$



Part IV: In the non-equilibrium regime using light



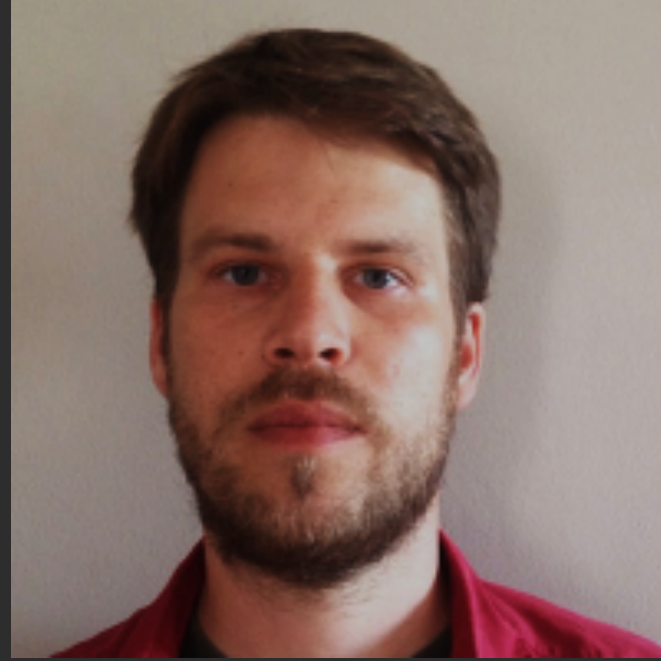
- The anharmonic energy level spacing in graphene allows unique energy transitions (within a factor of 2).
- Dipole-allowed transitions for $|n_f| - |n_i| \pm 1$.
- With graphene in the integer quantum-Hall regime; Excite holes below E_f to empty electron states above E_f ; Using selection rules: $|n_f| - |n_i| \pm 1$.



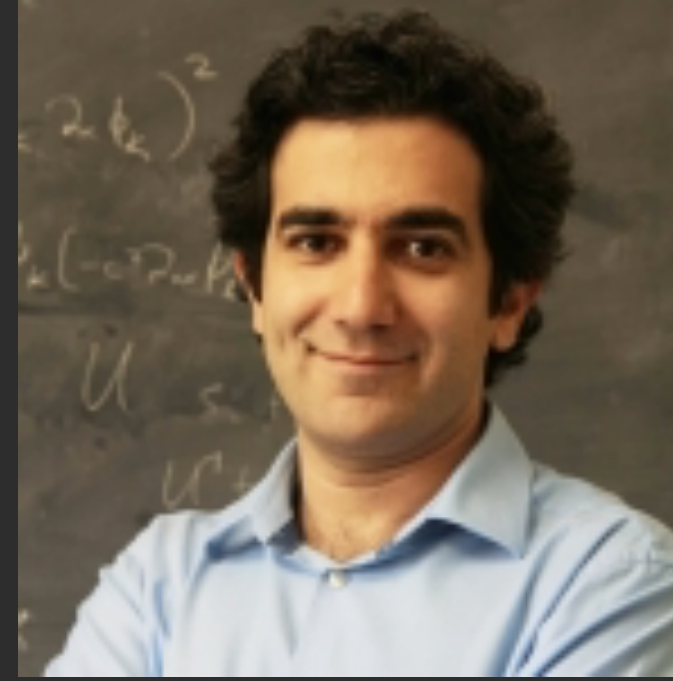
Now an experiment, and a cast of characters



Bin Cao



Tobias Grass



Mohammad Hafez

Optical and electrical measurements:

Olivier Gazzano, Tobias Huber-Loyola, Markus Müller

Theory: Michael Gullens

Joint Quantum Institute,
NIST and University of Maryland
College, MD USA

Sample Prep: 1

Jiuning Hu, Dave Newell,
National Institute of Standards and Technology
Gaithersburg, MD USA

Sample Prep: 2:

Kishan Ashokbhai Patel, Luca Anzi, Roman Sordan
L-NESS, Department of Physics,
Politecnico di Milano, Como

BN:

Kenji Watanabe, Takashi Taniguchi
National Institute for Materials Science,
Tsukuba, Japan

B. Cao, T. Grass, *et al.*, ACS Nano 2022, 16, 11, 18200–18209

The graphene sample

- **A typical structure:**

Exfoliated hBN, graphene, BN encapsulated graphene

Back-gated SiO₂/Si, with metallic back contact

Sizes range from 3 x 3 μm to 10 x 10 μm

- **Ohmic contacts:**

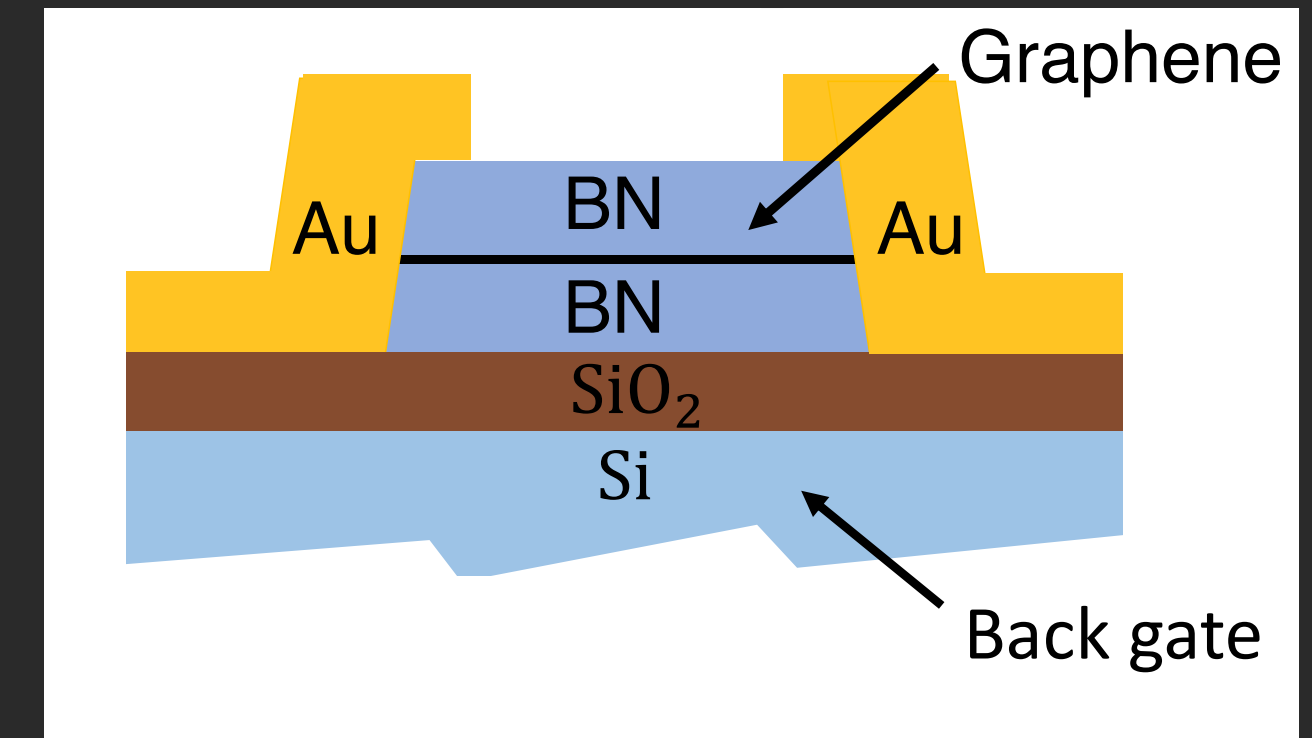
Through an e-beam deposits Al hard mask

Graphene & hBN etched selectively by O₂ and SF₆ RIE plasma

Leaves pristine graphene exposed edges

Ohmic contacts: Cr/Pd/Au (2/5/80 nm) patterned by e-beam lithography and e-beam

Two-terminal device with back gate



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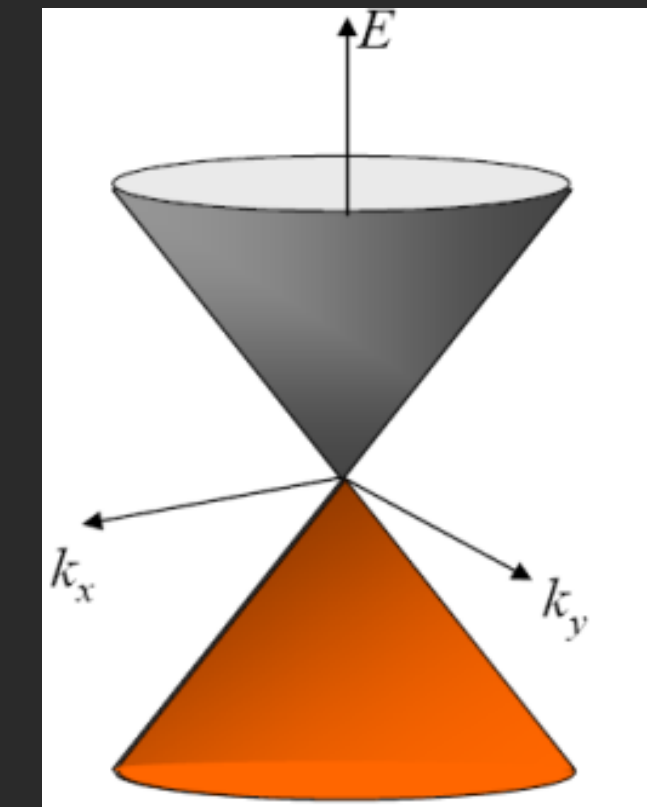
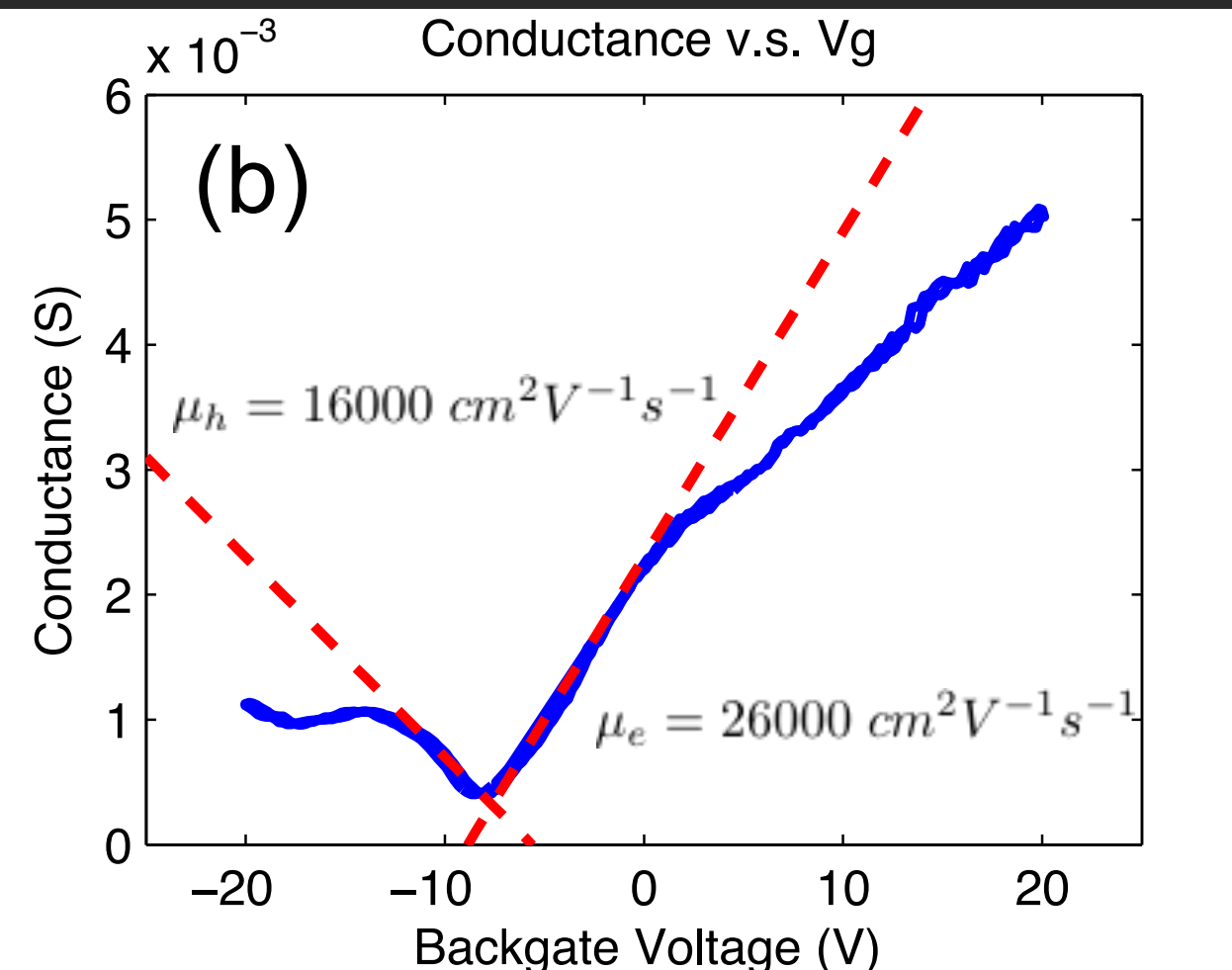
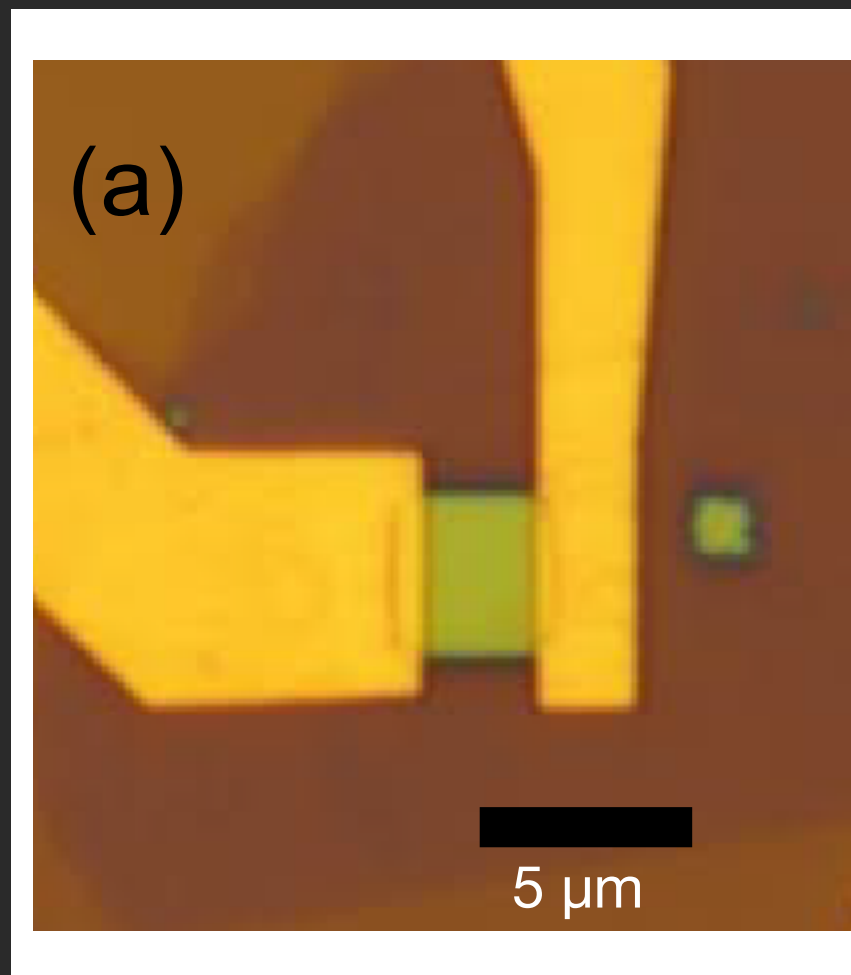
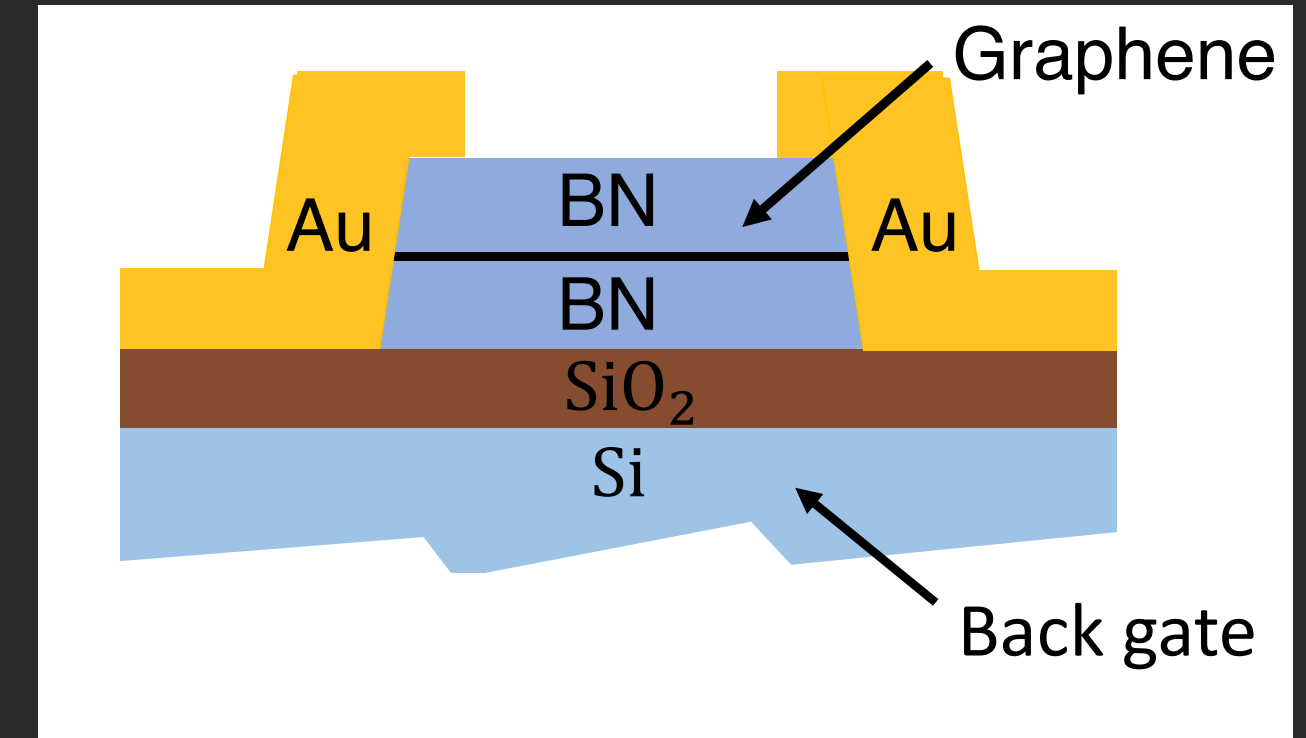
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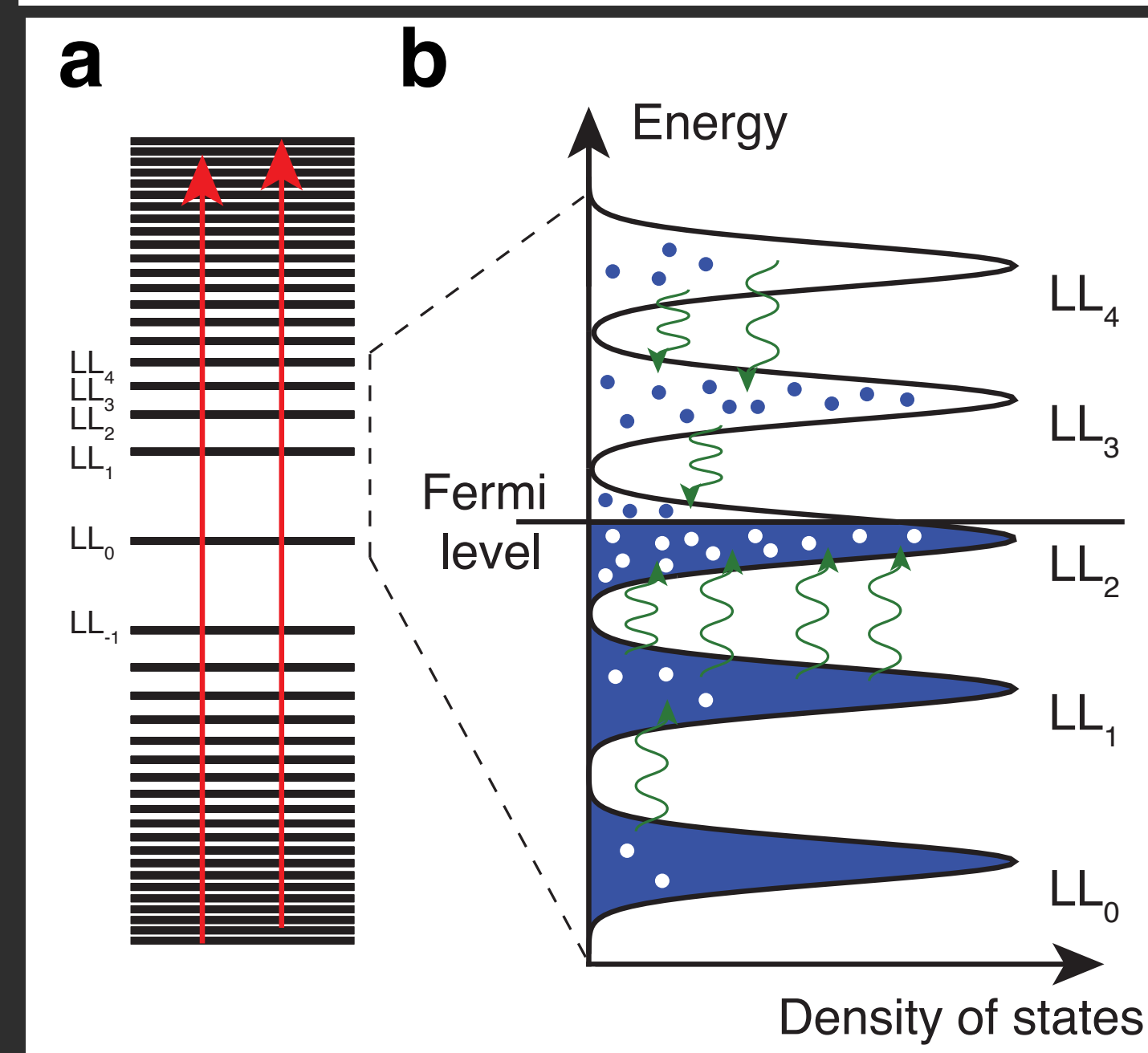
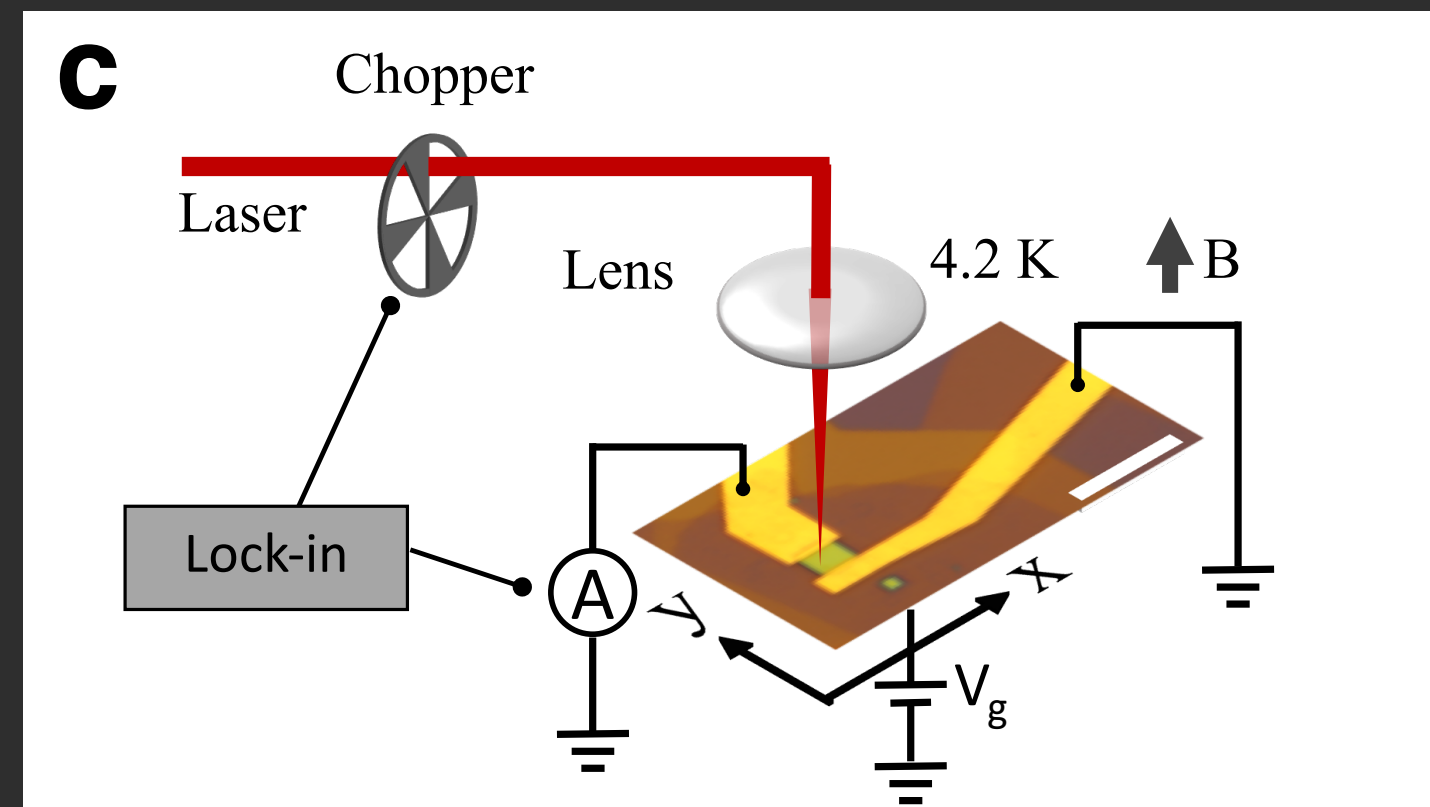


Size:
2.49 μm * 3.87 μm

$\mu_e = 26,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$
 $\mu_h = 16,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$

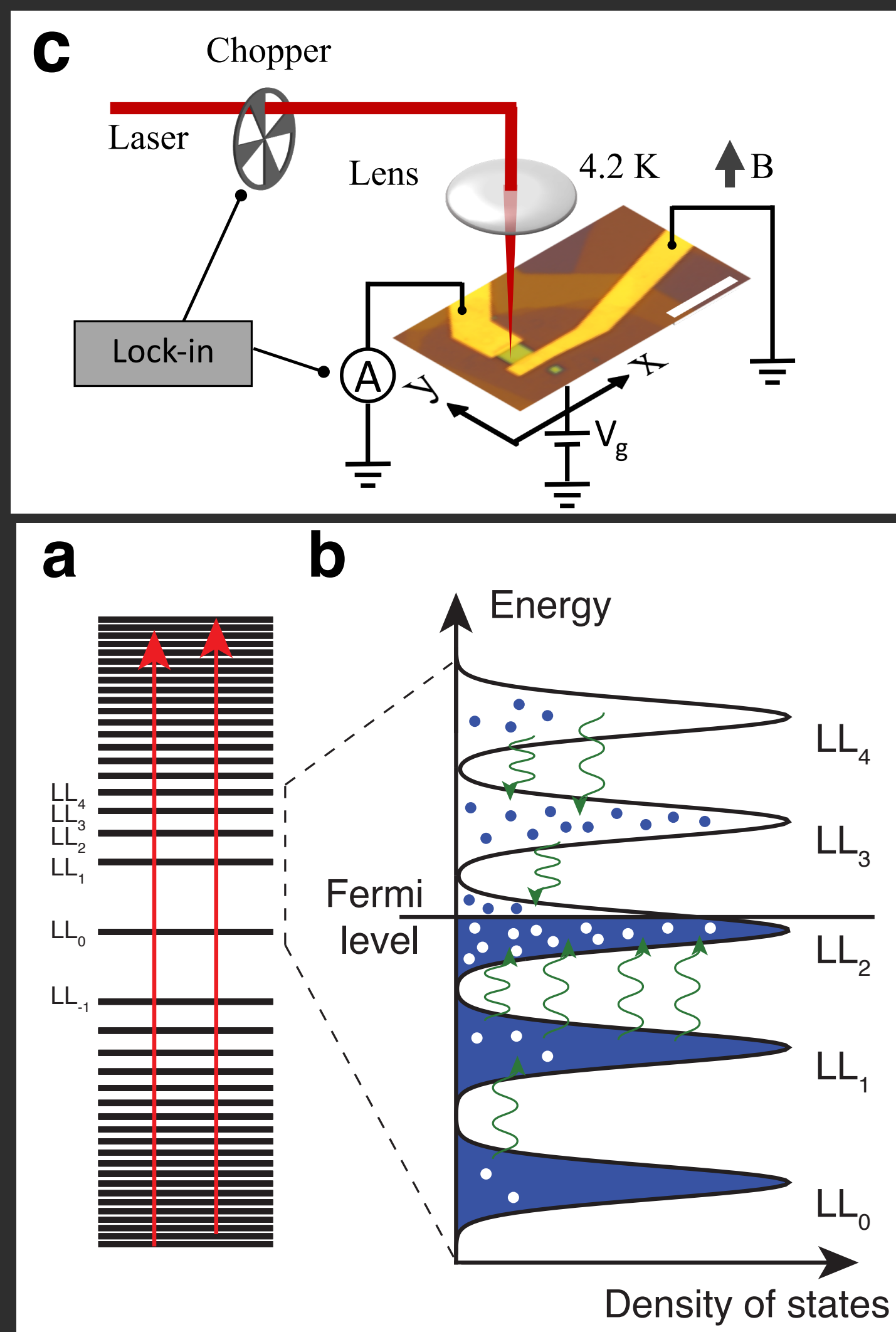
Photocurrent in the IQH - Non-equilibrium transport

- Excite with 930 nm light, create e and h populations at $|n| \approx 36$;
- Fast (sub ps) relaxation and diffusion, creating photocurrents.

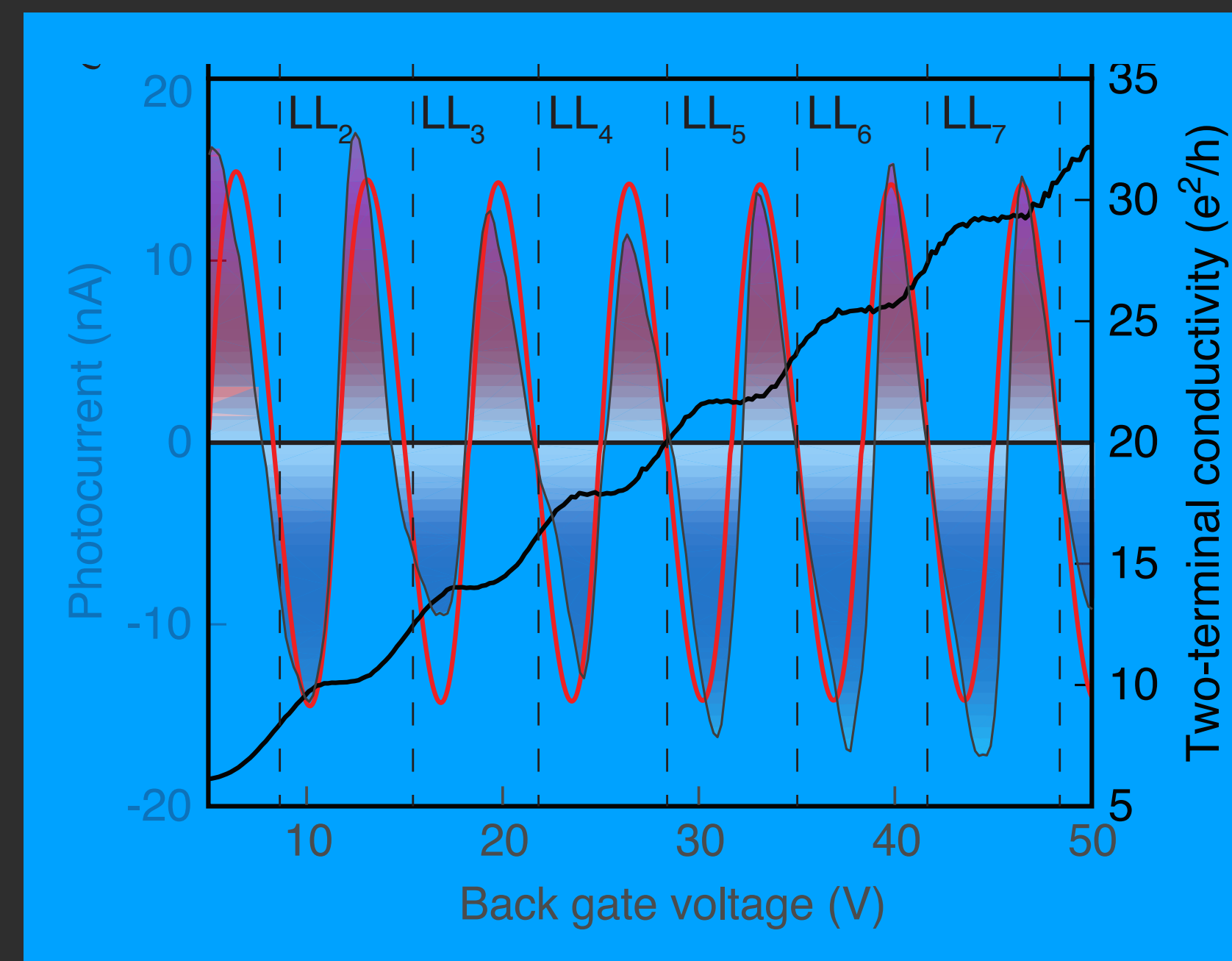


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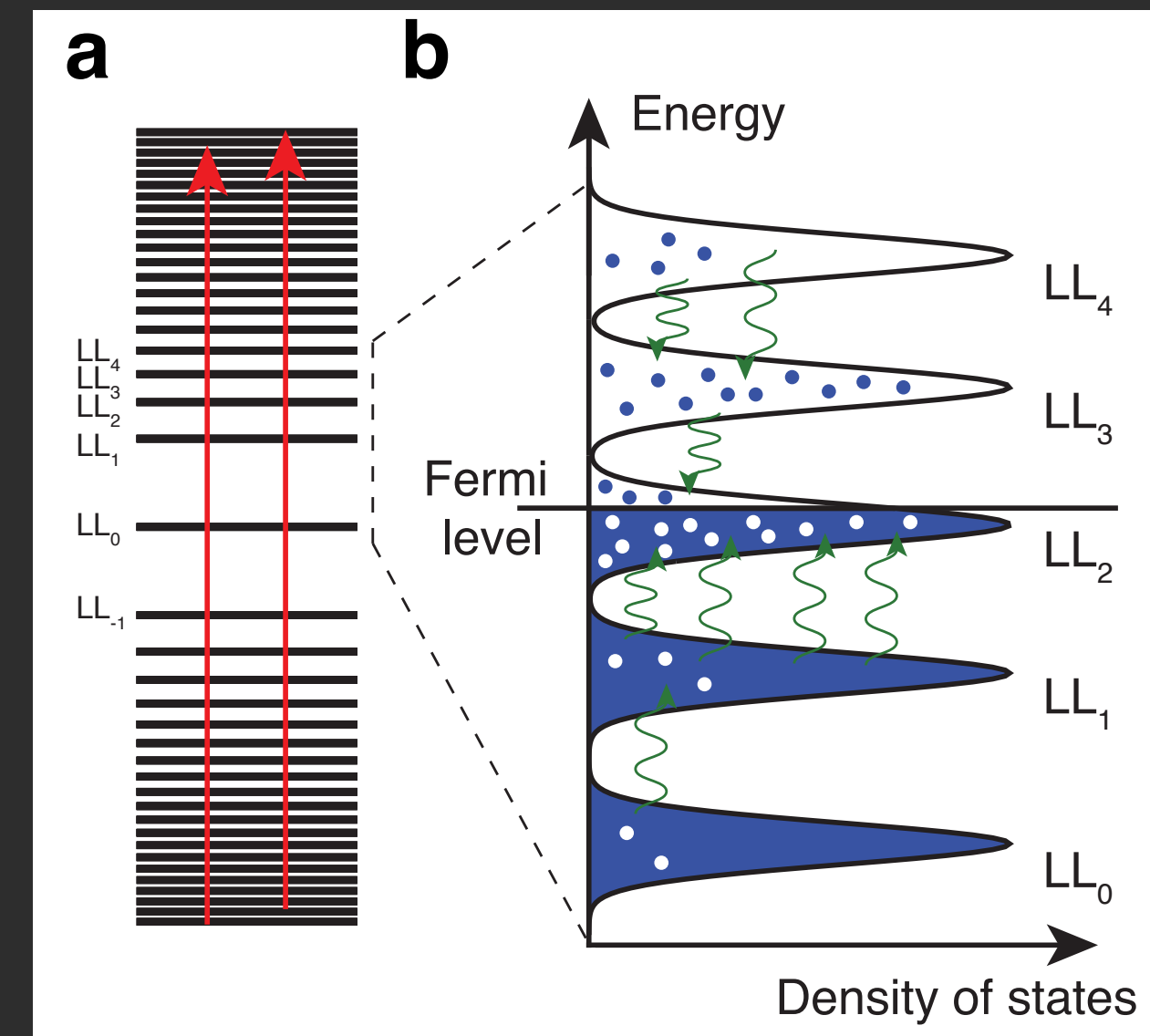
4.2K B= 4T



- Backgate voltage control E_f , no injected current.
- Photocurrent oscillations track LL's.
- Zeros in photocurrent are at half filling.

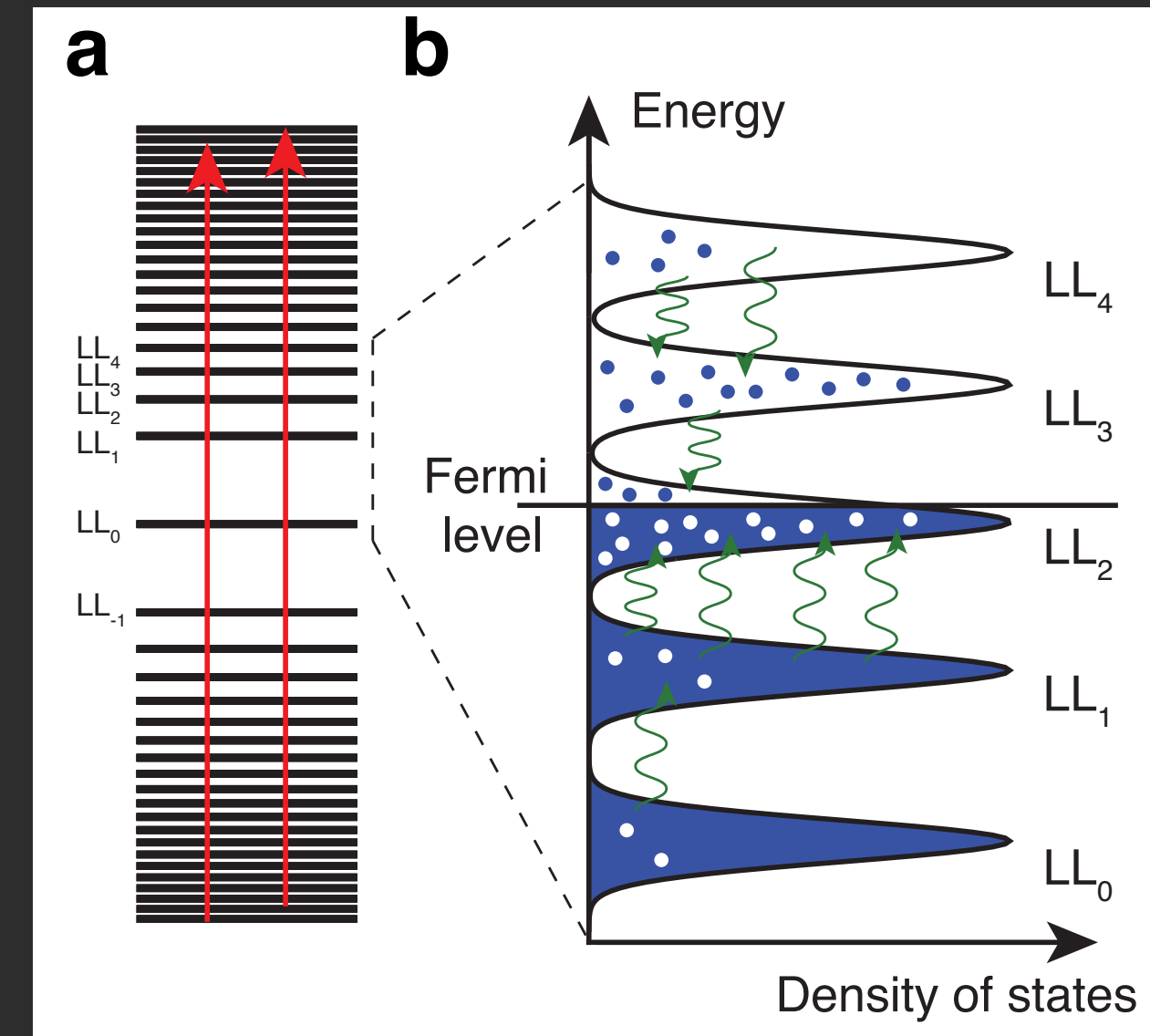
Why do we get any photocurrent?

- Why should we get any photocurrent?
- We create electrons & holes in equal amounts, they decay (~ 100 fs), and recombine.



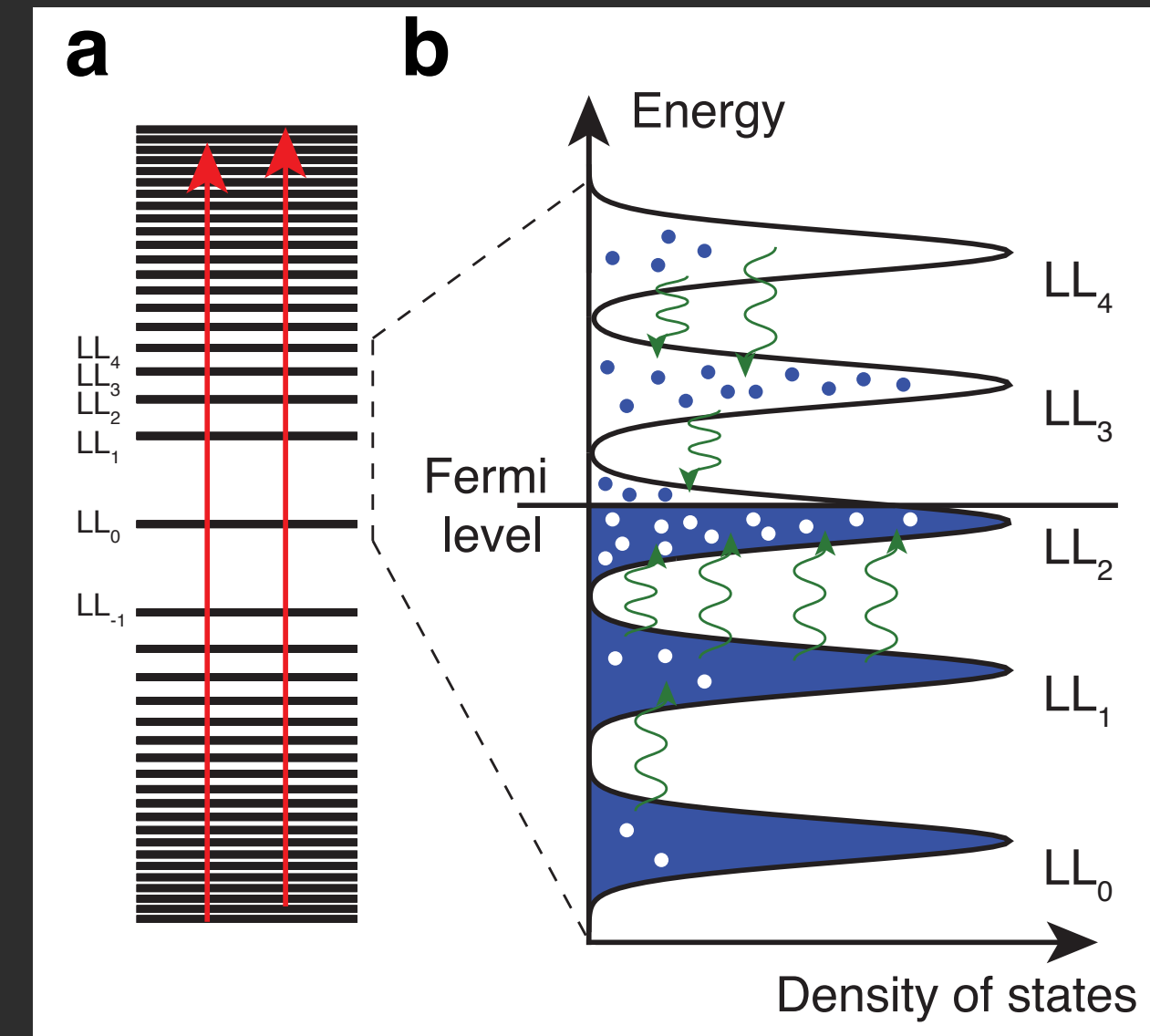
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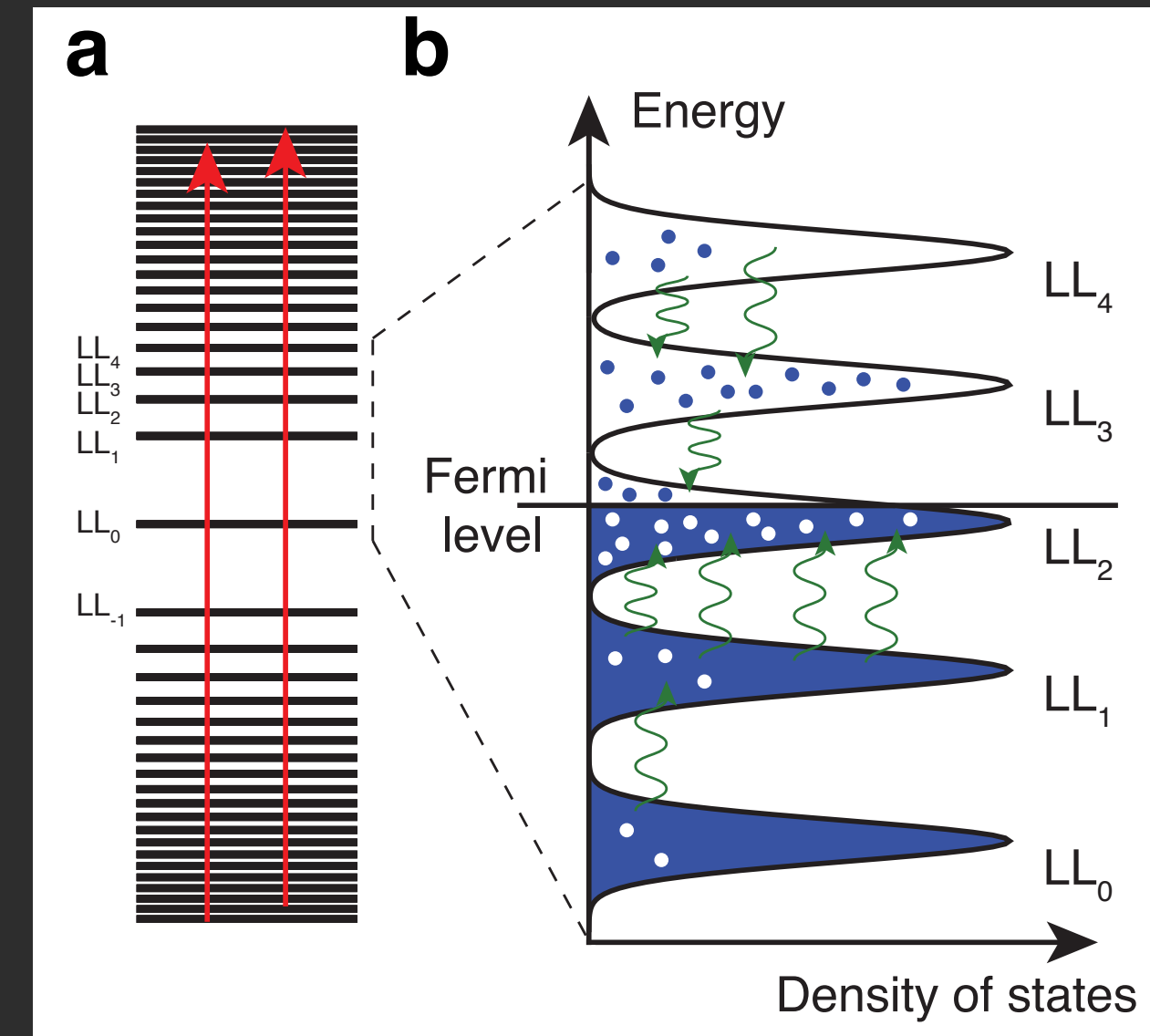
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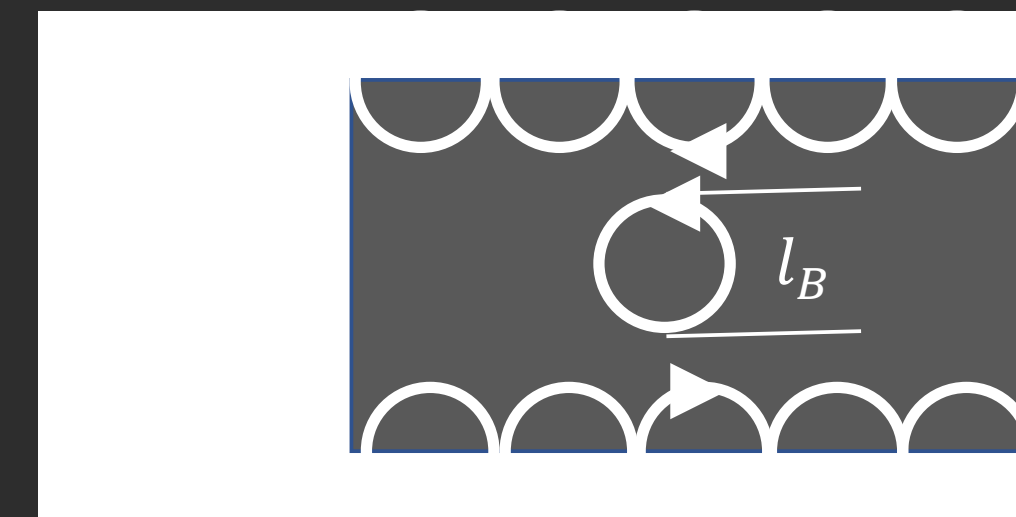
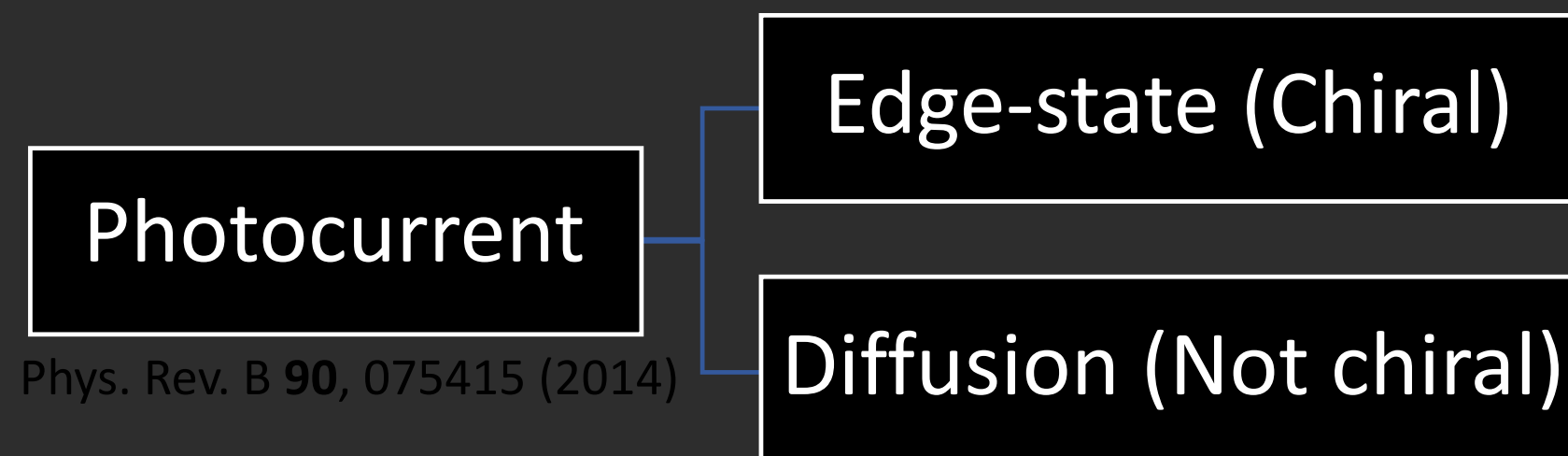
- Two processes: Relaxation of hot carriers to E_f or Diffusion of hot carriers to edges or contacts.
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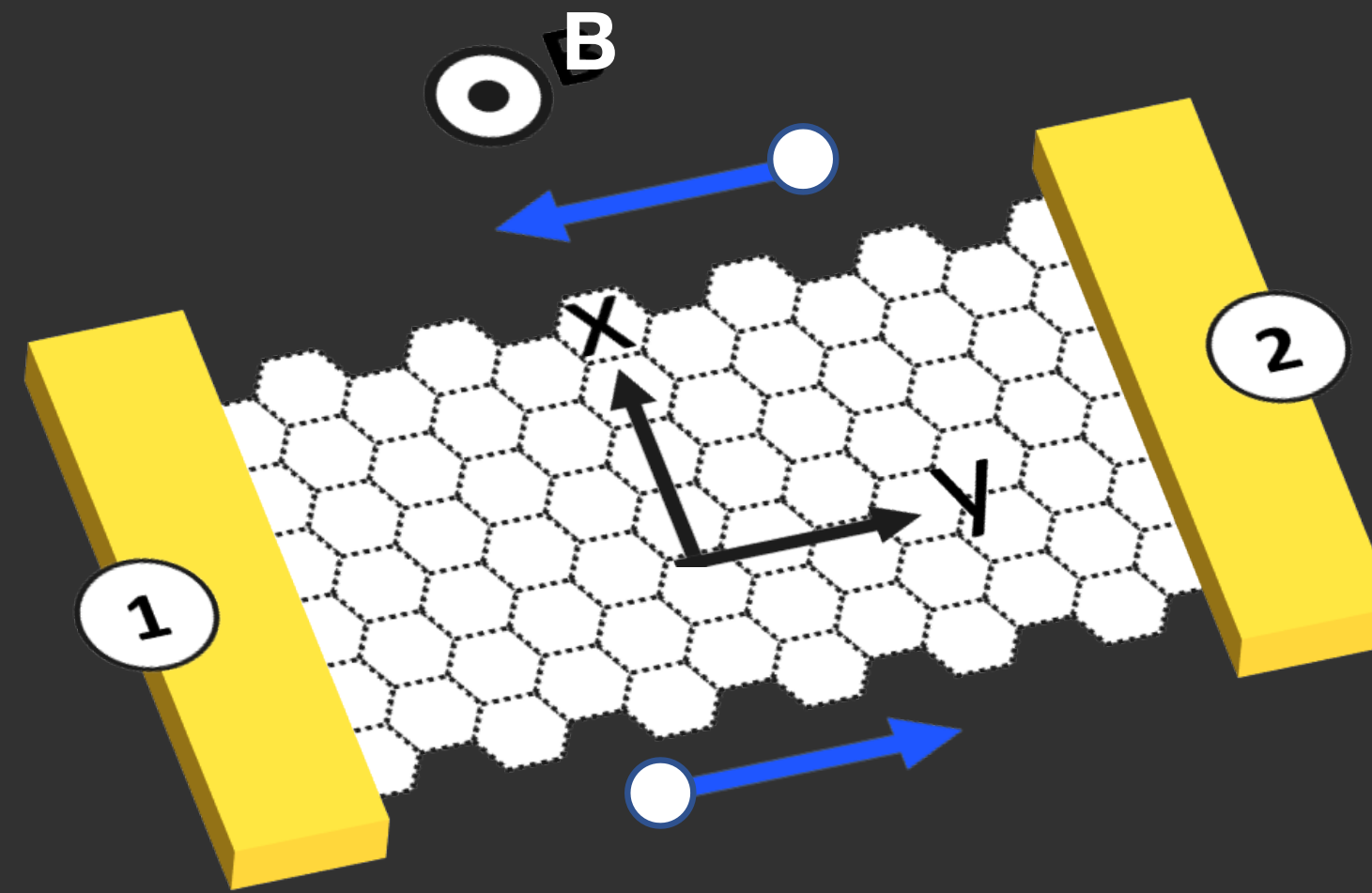
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- *Relaxation process of excess hot carriers of one type depends on unbalanced number of available states at the Fermi level.*
- Photocurrent has two components:



Separate photocurrent contributions related to edge & extended states



- When B flips direction the edge state currents flip directions. $I(B^+) - I(B^-)$ will add the edge-state currents.
- Diffusive extended-state currents (total electron and hole current) diffusing to contacts are unaffected by sign of B .

$I(B^+) - I(B^-)$ negates diffusive extended state currents, but $I(B^+) + I(B^-)$ will add the currents.

Renormalise to focus on edge states: $I(B^+) - I(B^-)$.

Chirality of edge-state photo-carriers

- Modify the energy spectrum mentioned earlier:

$$\epsilon_n = \text{sign}(n) \frac{\hbar v_F}{l_B} \sqrt{2 |n|};$$

v_F Fermi velocity,
 l_B magnetic length, $l_B = \sqrt{\frac{\hbar}{eB}}$
 n Landau level index

$$\epsilon_n = \text{sign}(n) \frac{\hbar v_F}{l_B} \sqrt{2 |n|} + \lambda \text{sgn}(q) V(x)$$

$\lambda = +1$ for conduction band •

$\lambda = -1$ for valence band

q the carrier charge

$V(x)$ the edge confining potential.

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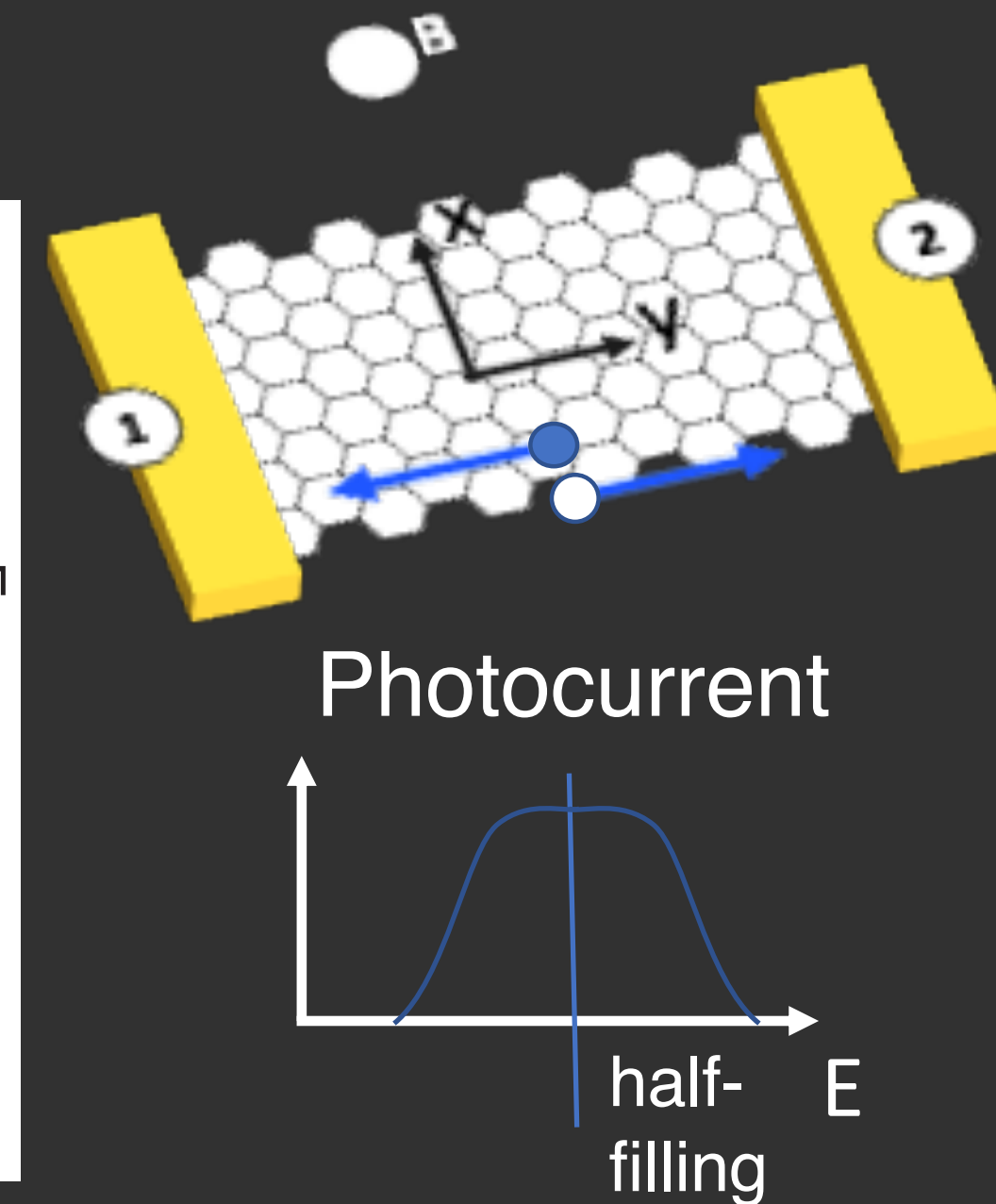
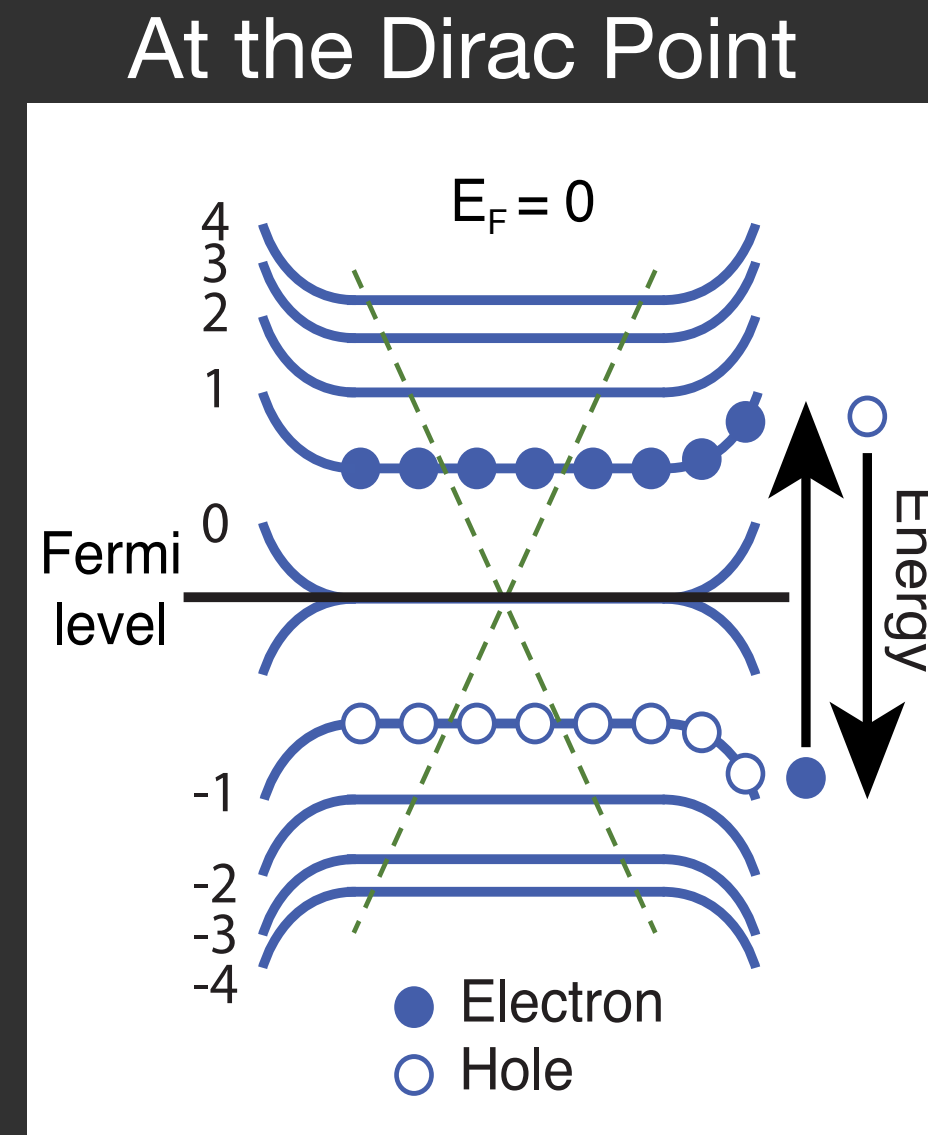
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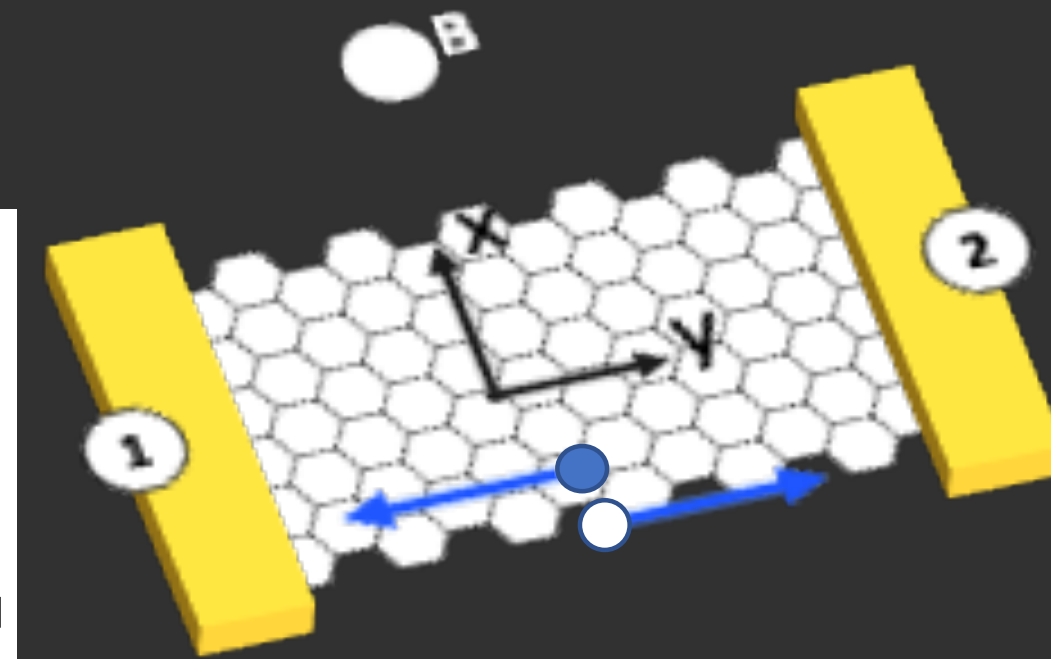
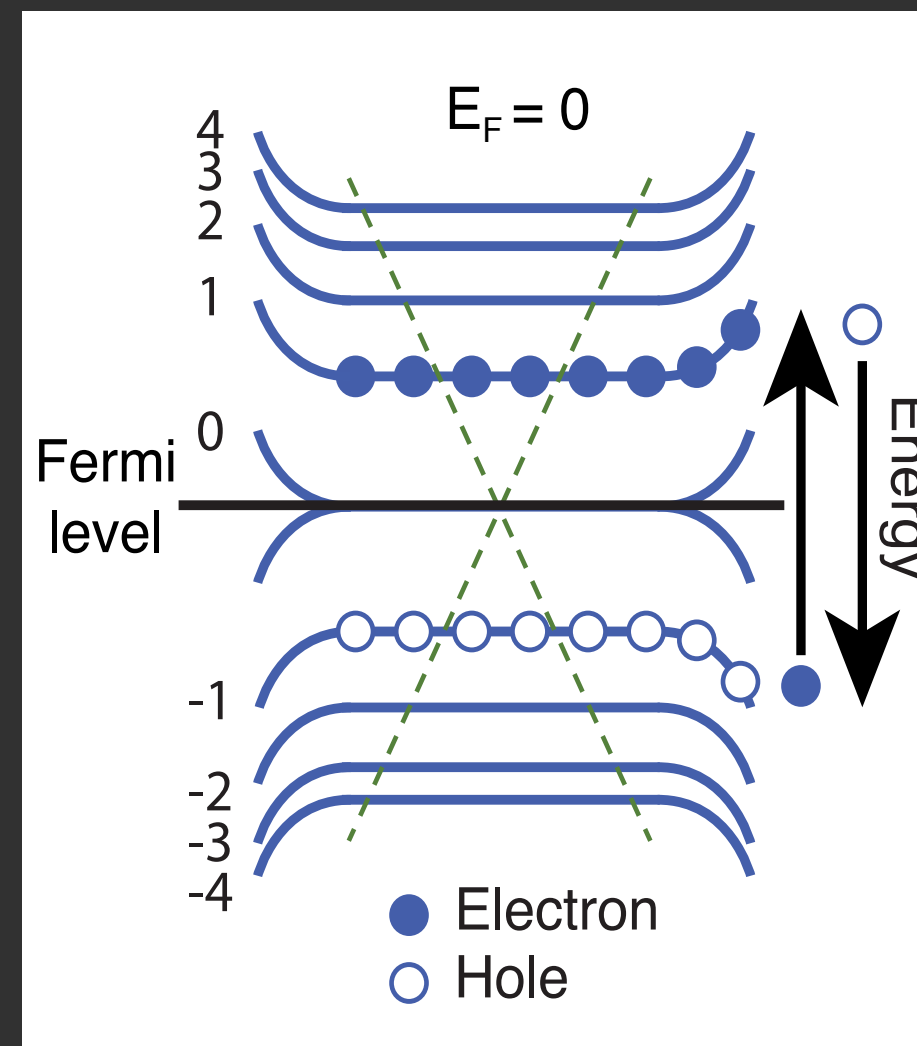
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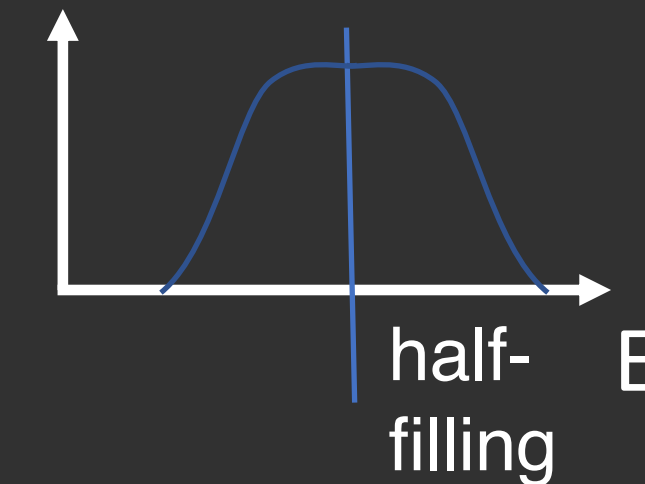
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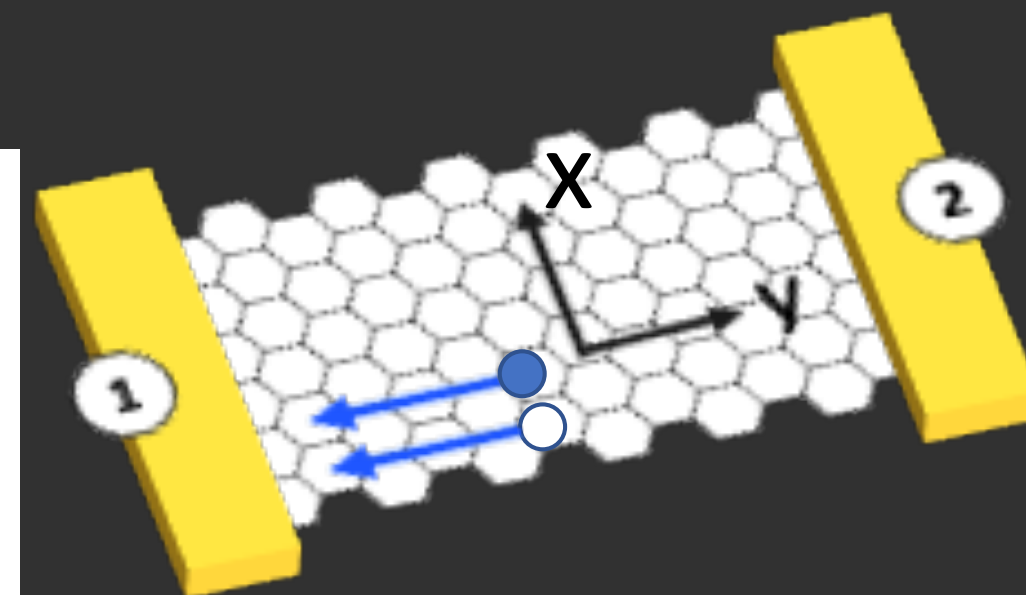
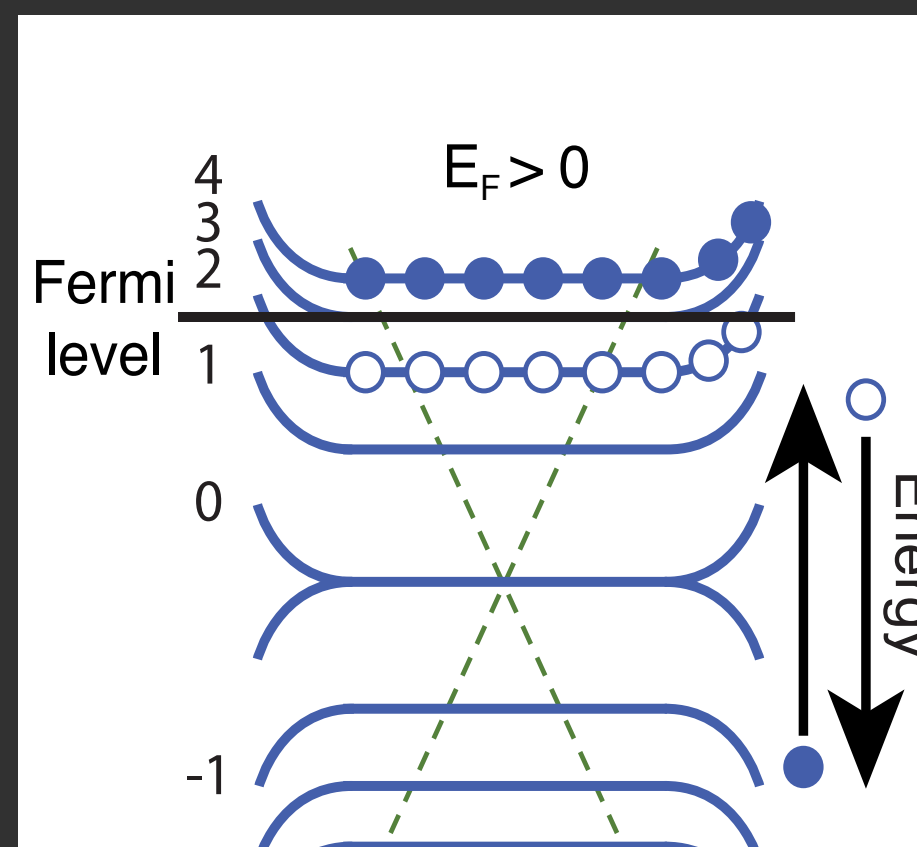
At the Dirac Point



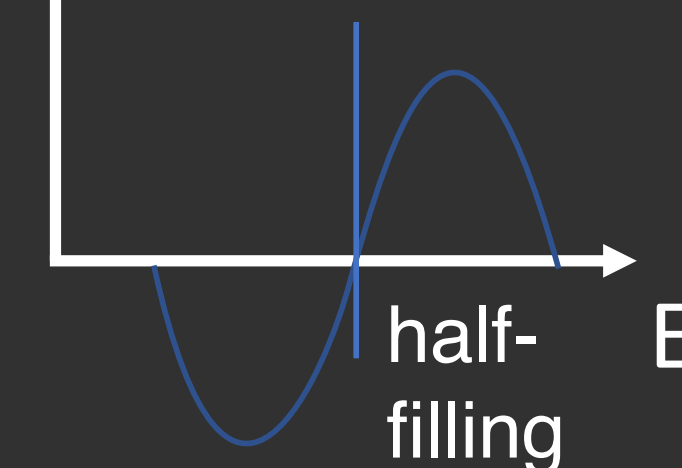
Photocurrent



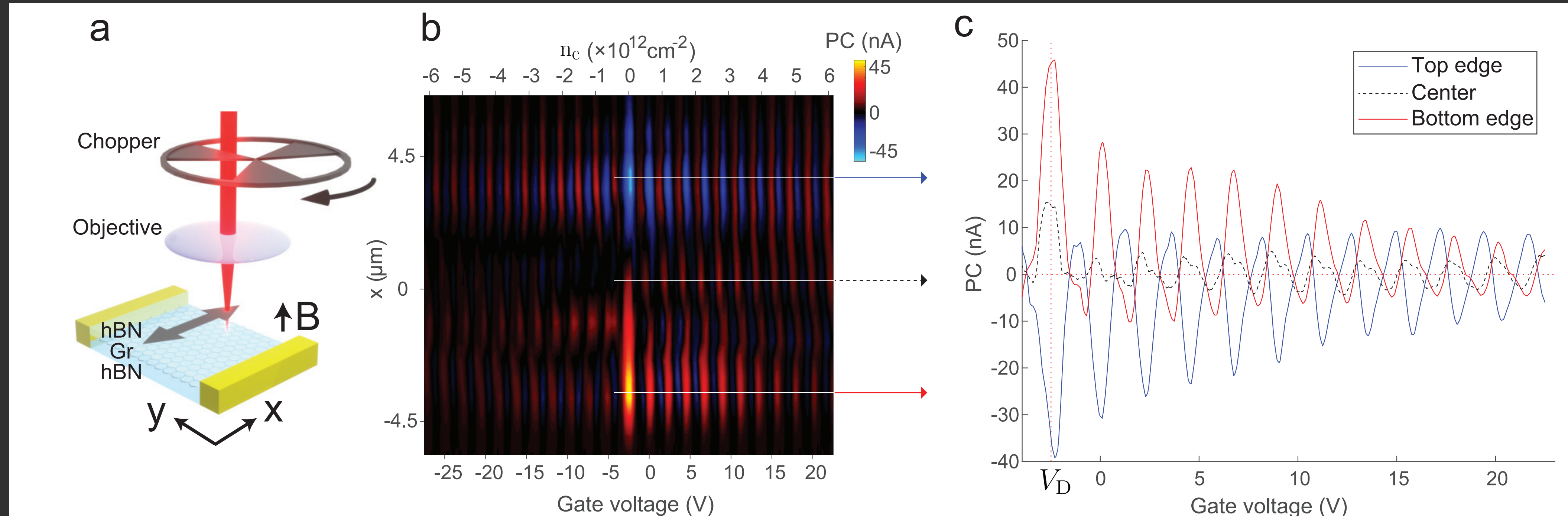
Away from Dirac Point



Photocurrent



Edge-state currents from photoexcitation



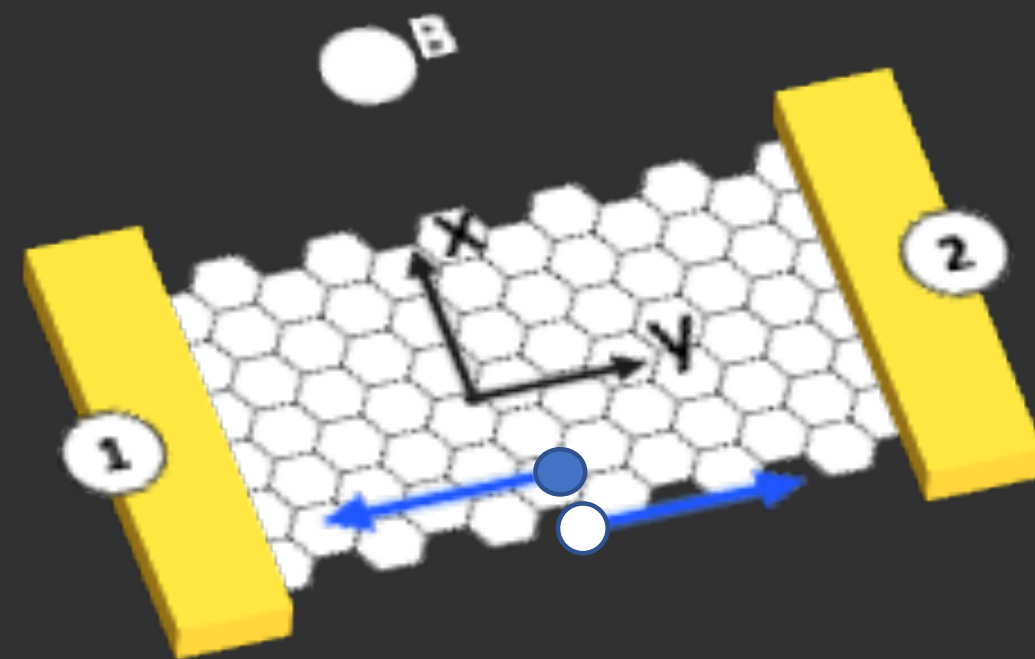
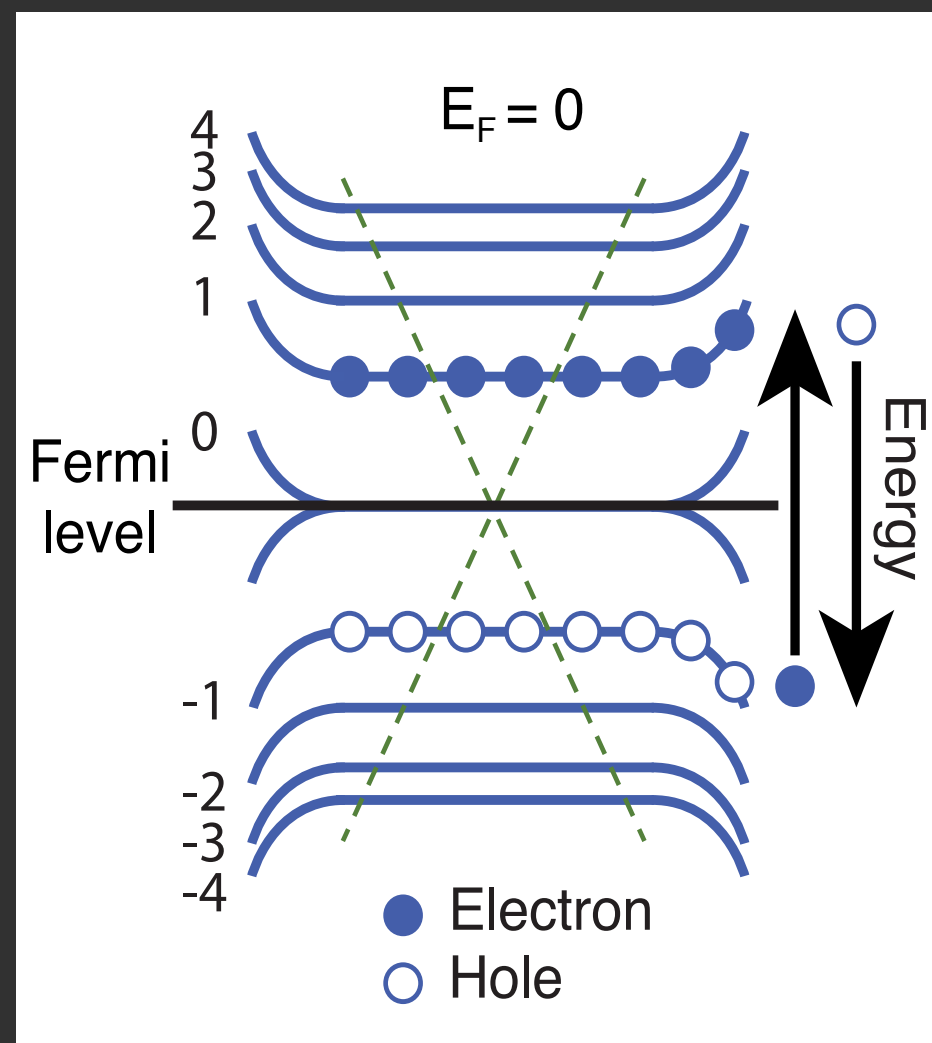
$$I(B^+ - B^-)$$

Edge-state current contributions

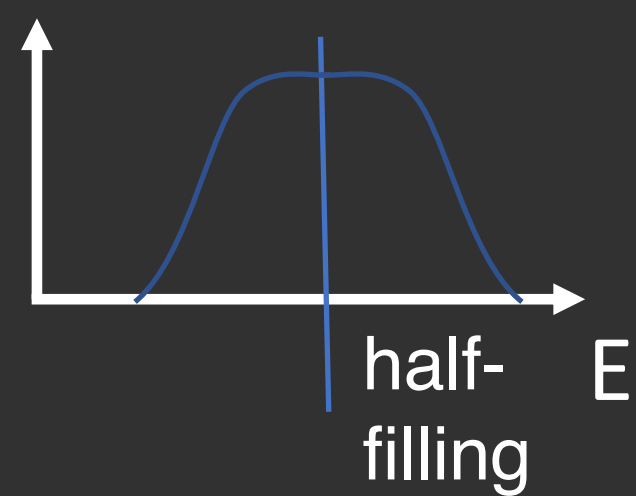
- Little B-field dependent transport in the sample center.
- Edge states currents are more concentrated at edges.

Edge-state photocurrent half-filling with LL

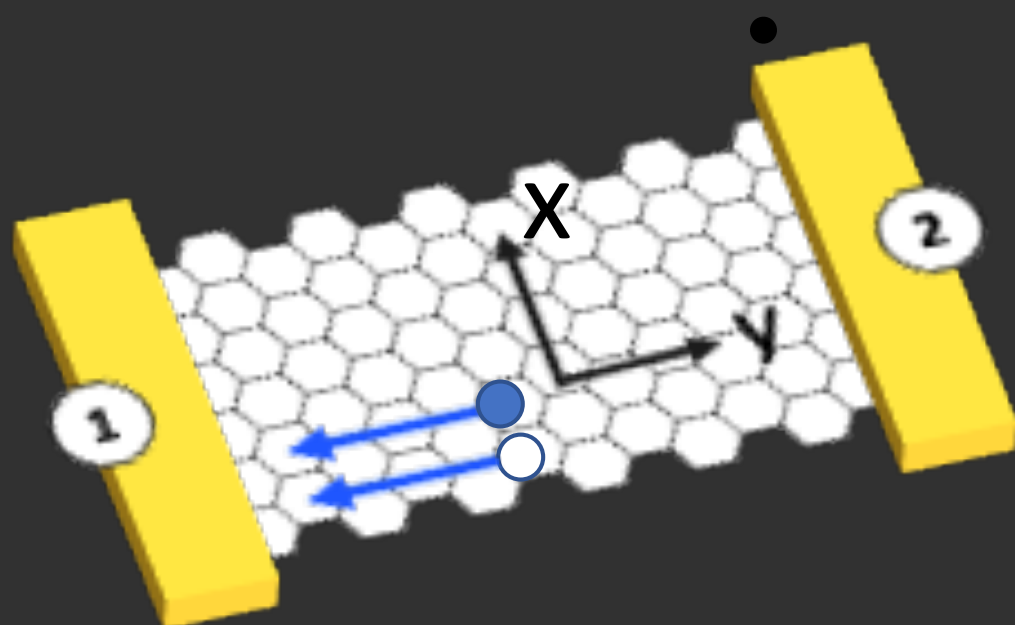
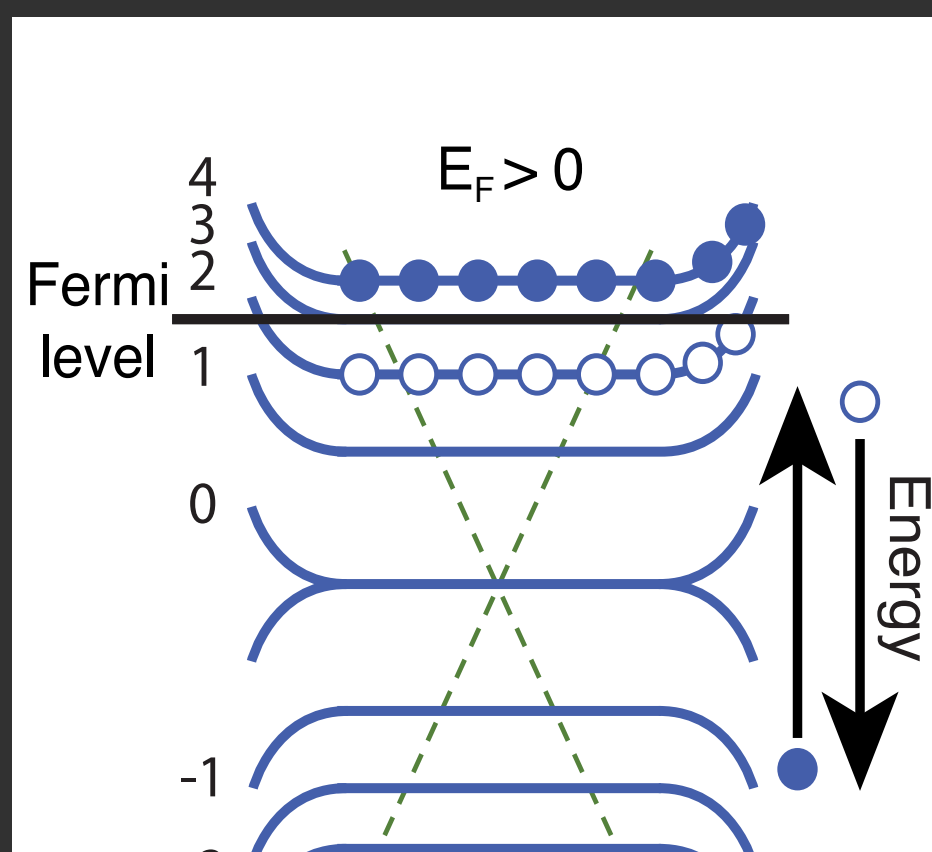
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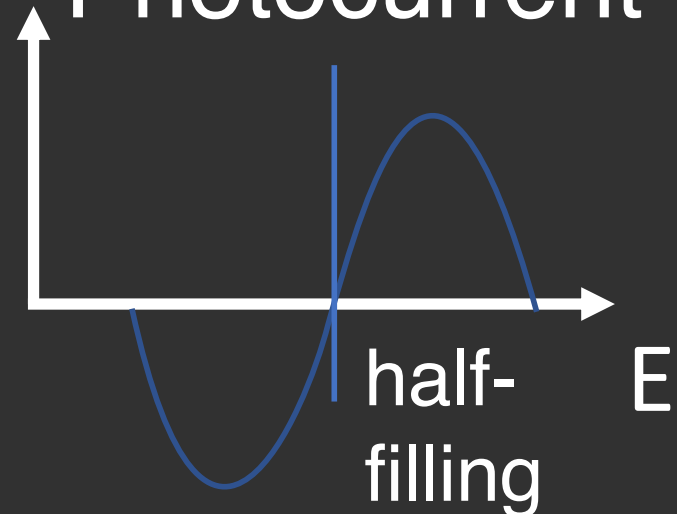
Photocurrent



Away from Dirac Point

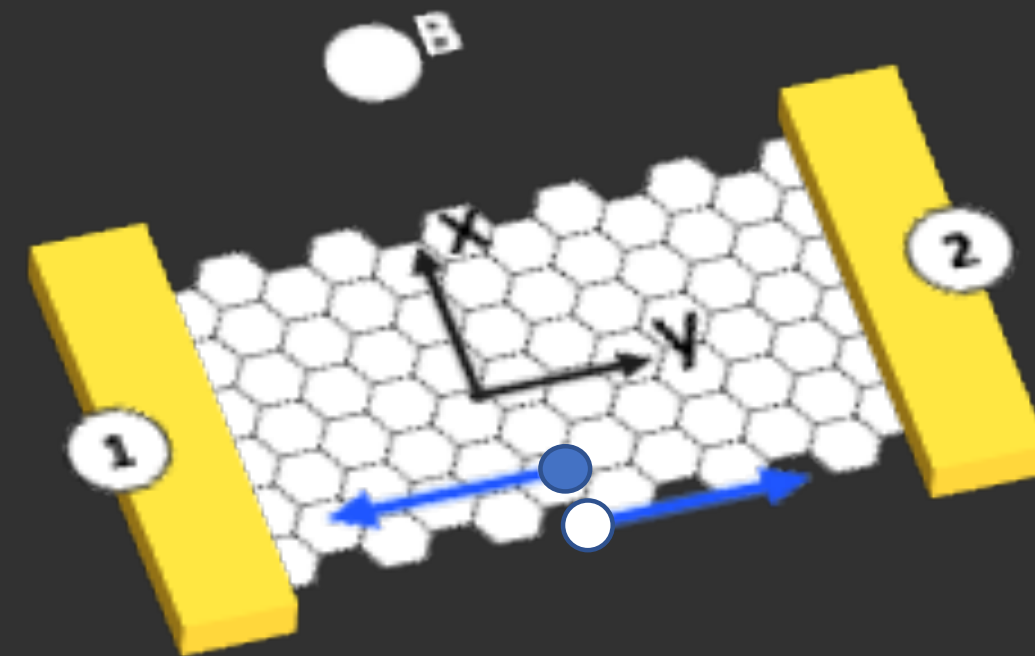
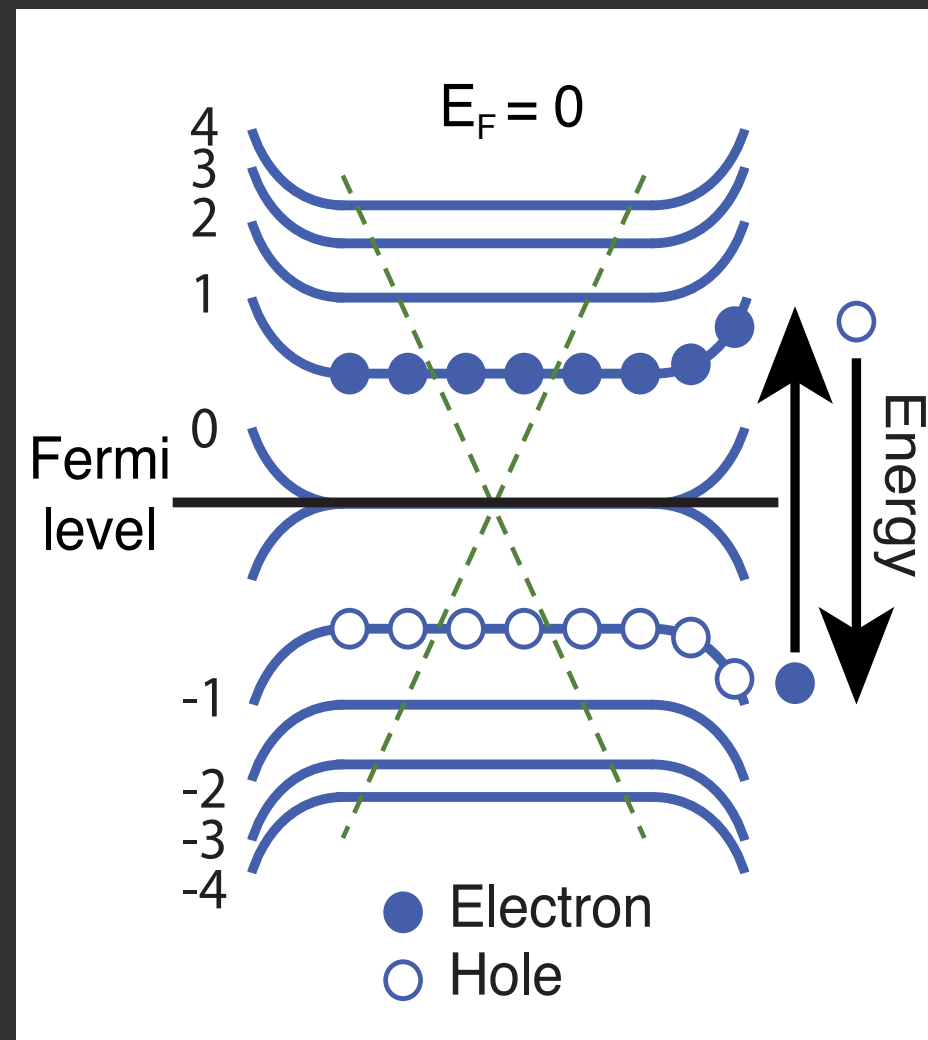


Photocurrent

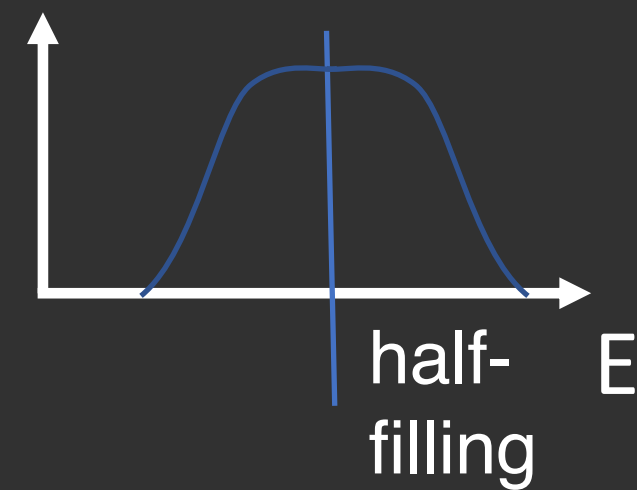


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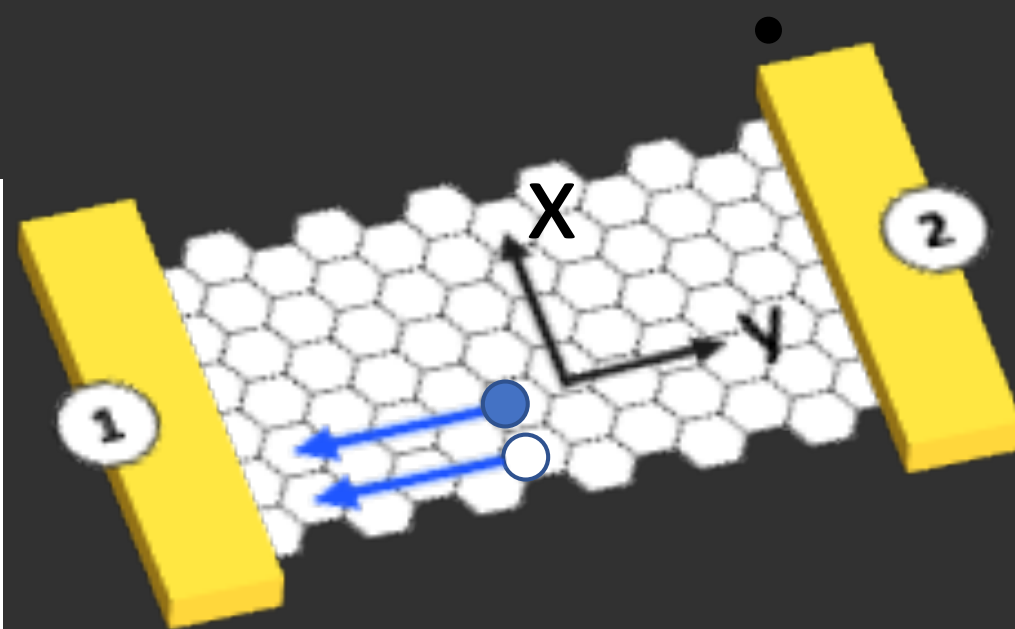
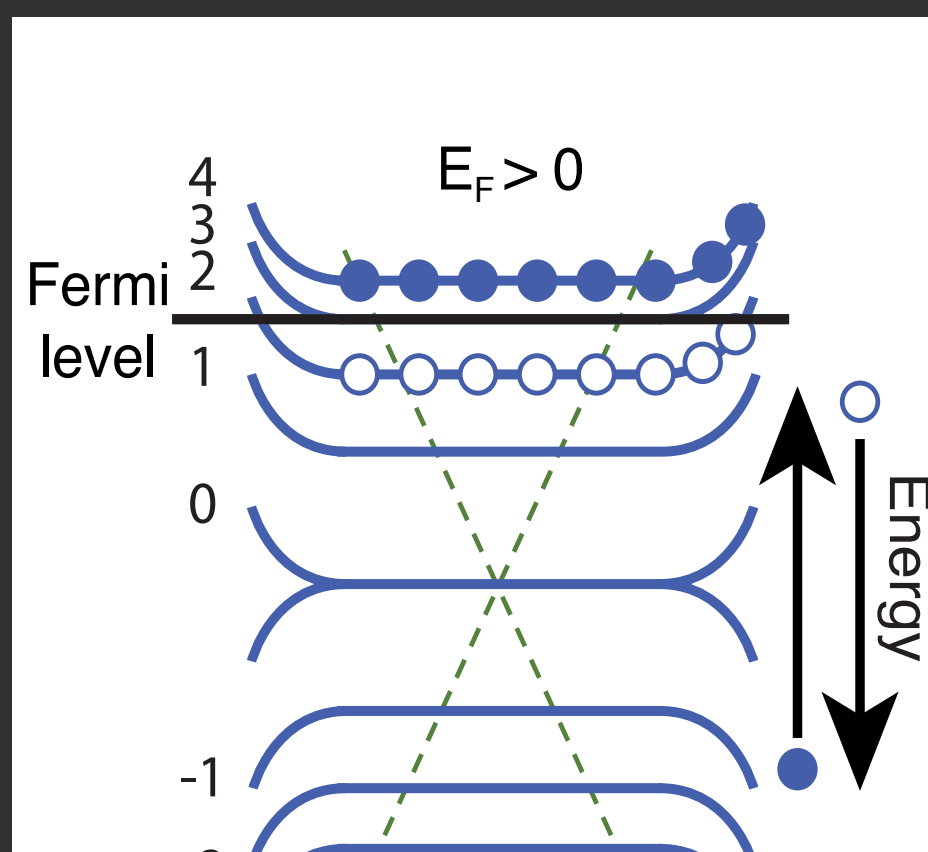


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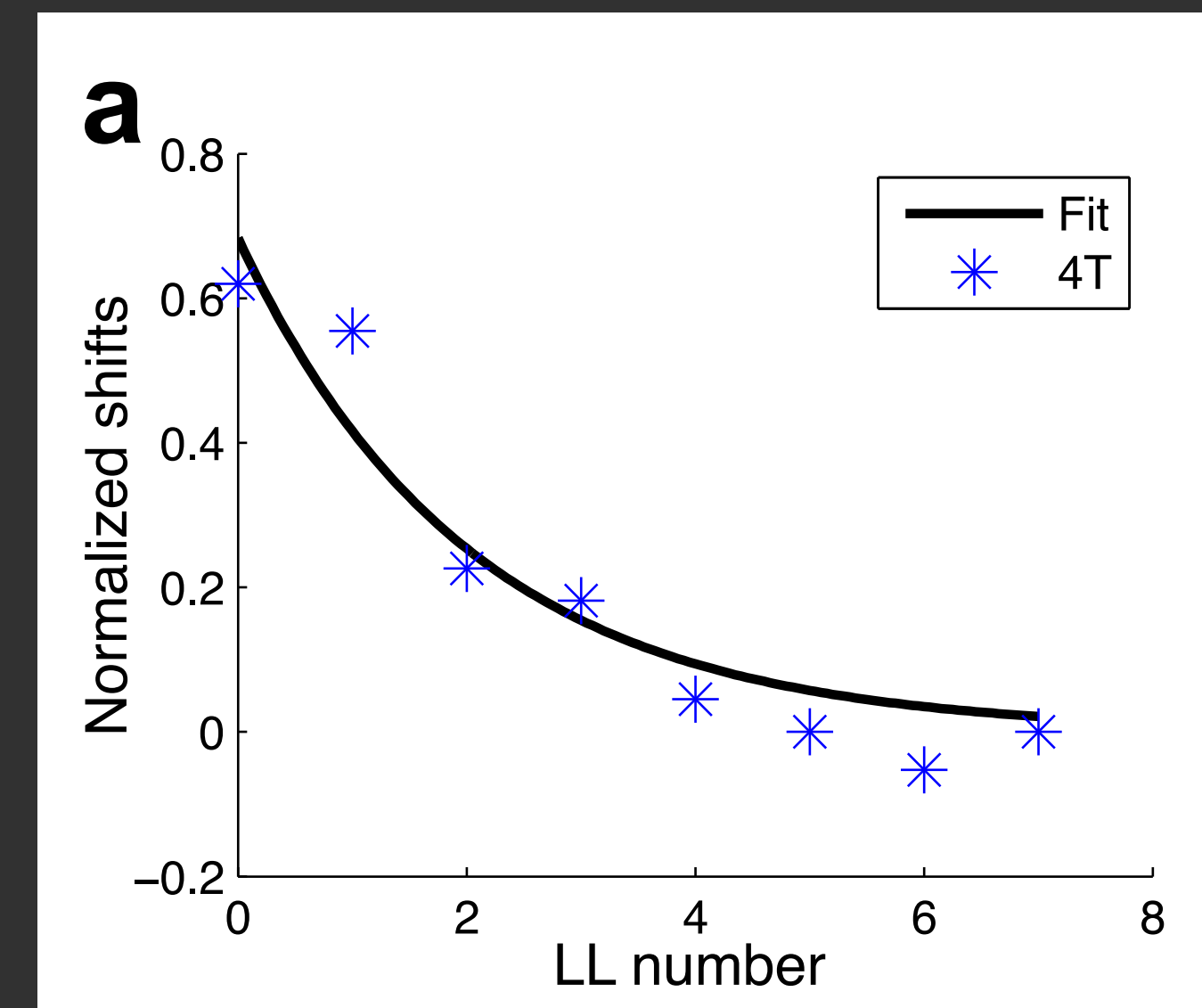
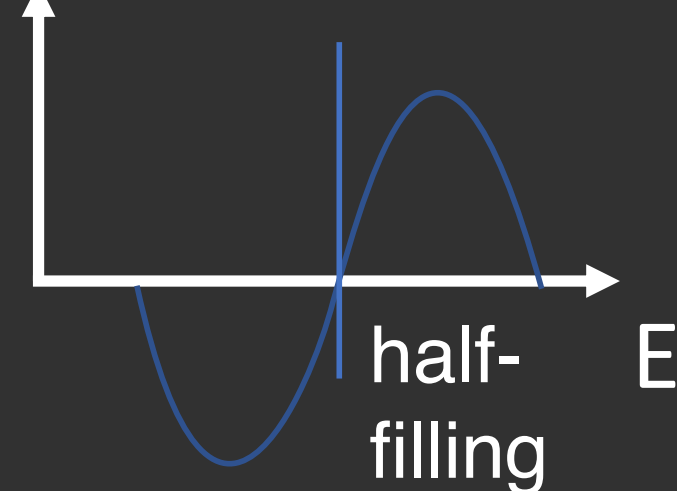


- At LL_0 half filling of LL is a PC peak
- While away from LL_0 PC is 0.
- Because of edge states have multi-LL components the PC moves from a peak value to 0 over several LL 's.

Away from Dirac Point

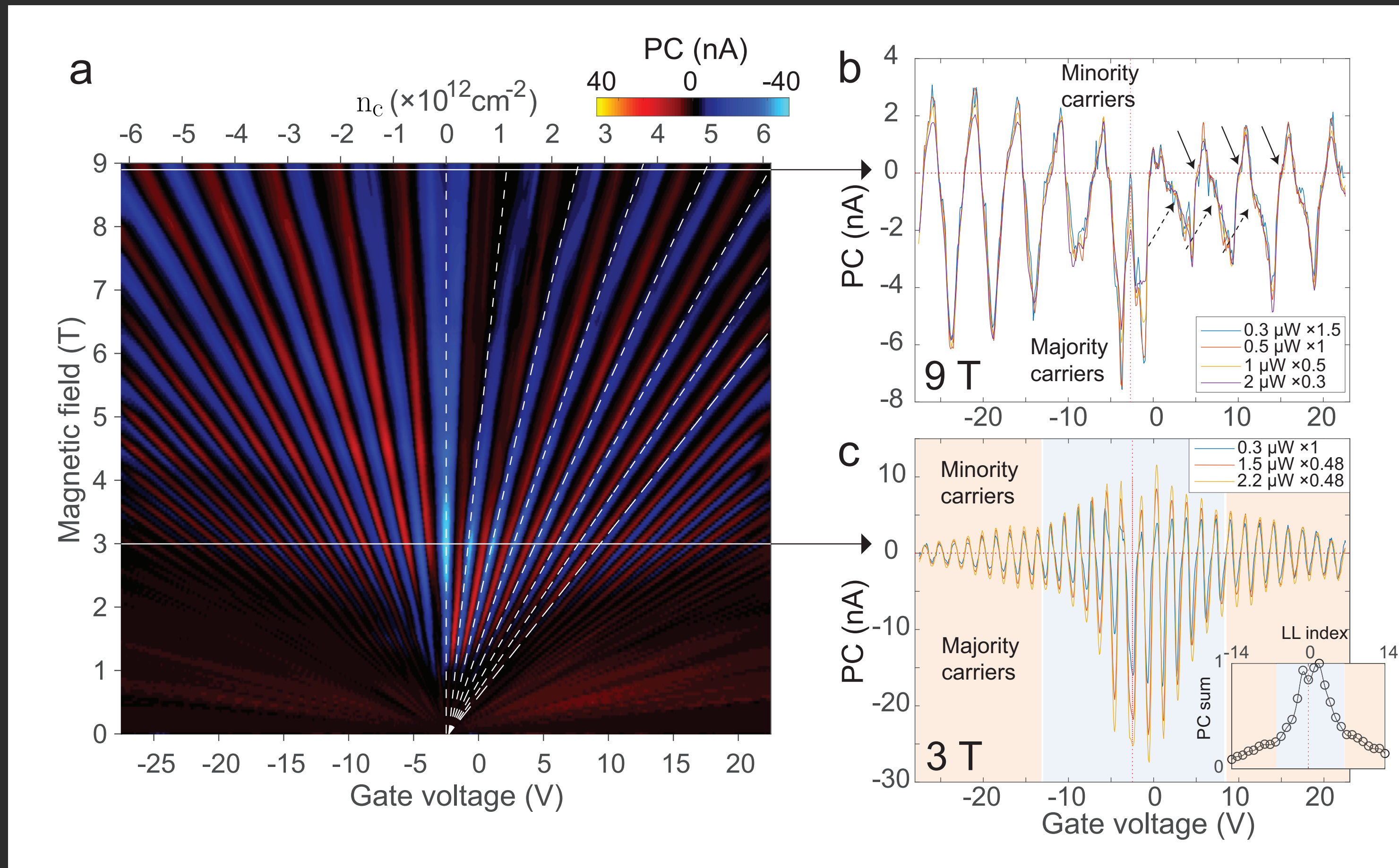


Photocurrent



Shape of the photocurrent envelop as a function of B -field

The case for carrier multiplication



9T region: The photocurrent is flat

Lower B -fields, 3T:

Large photocurrent majority (e^-) carrier envelope

But also a smaller PC minority carrier envelope.

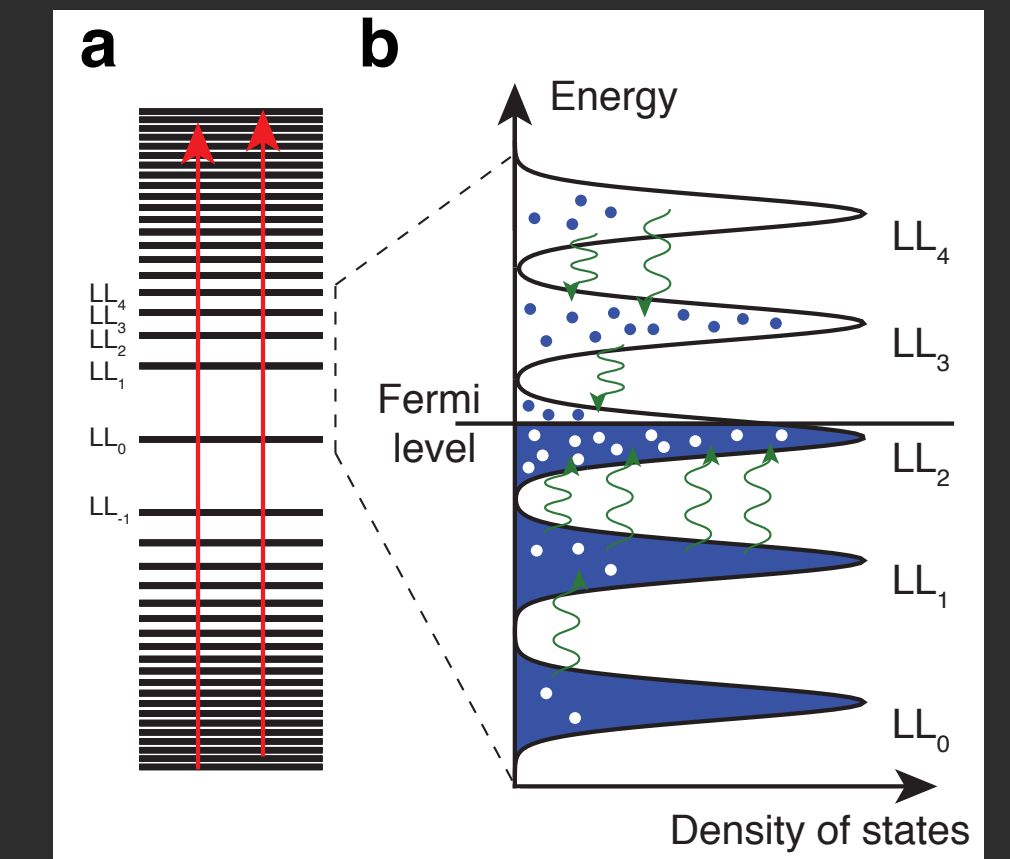
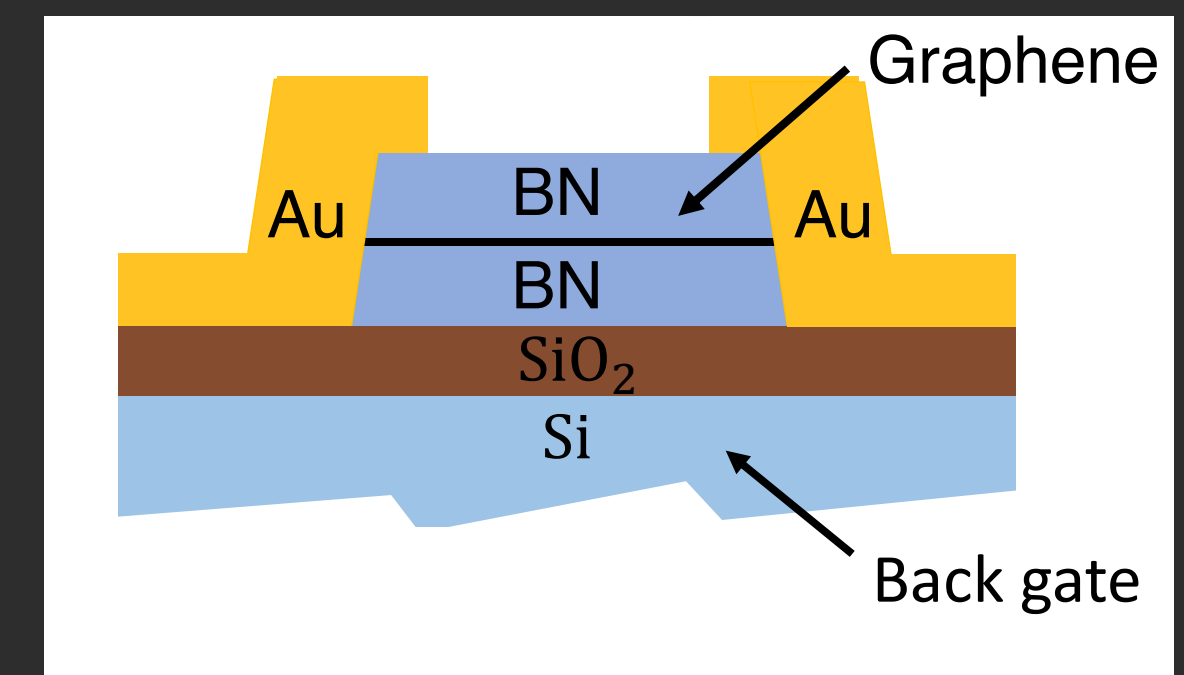
Carrier multiplication:

The majority carrier envelop could be due to e^- and h^+ adding around LL_0 .

But the **minority carrier envelope can only be explained by carrier multiplication.**

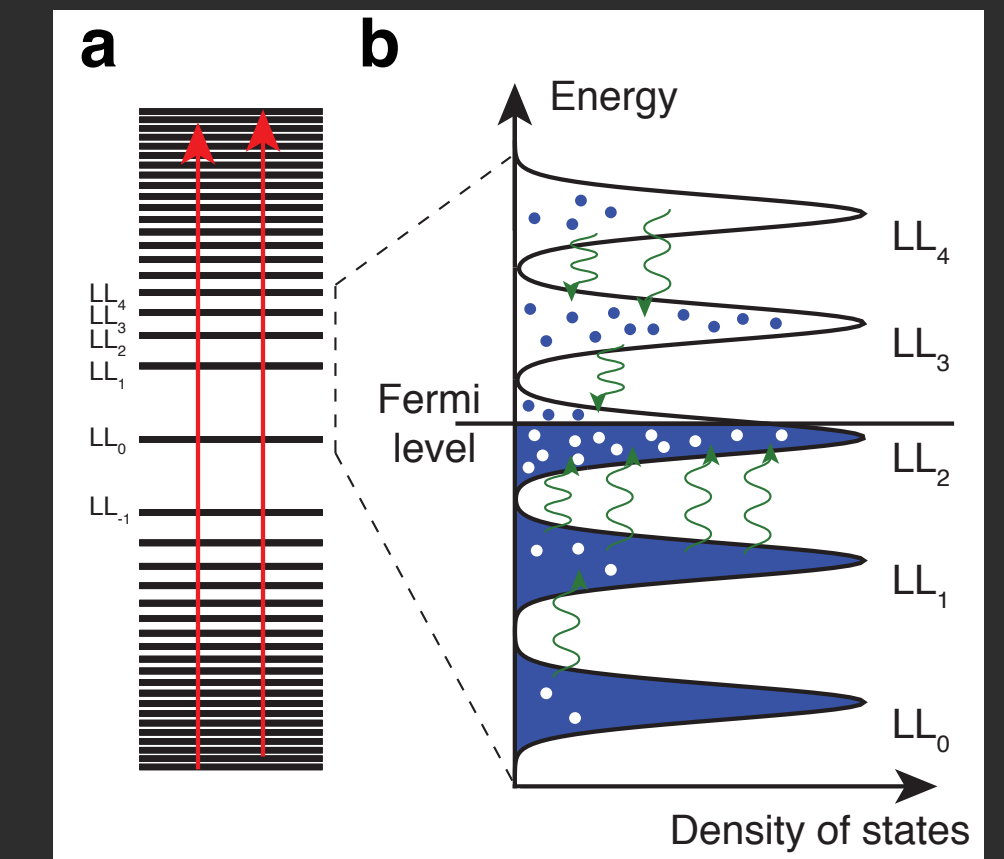
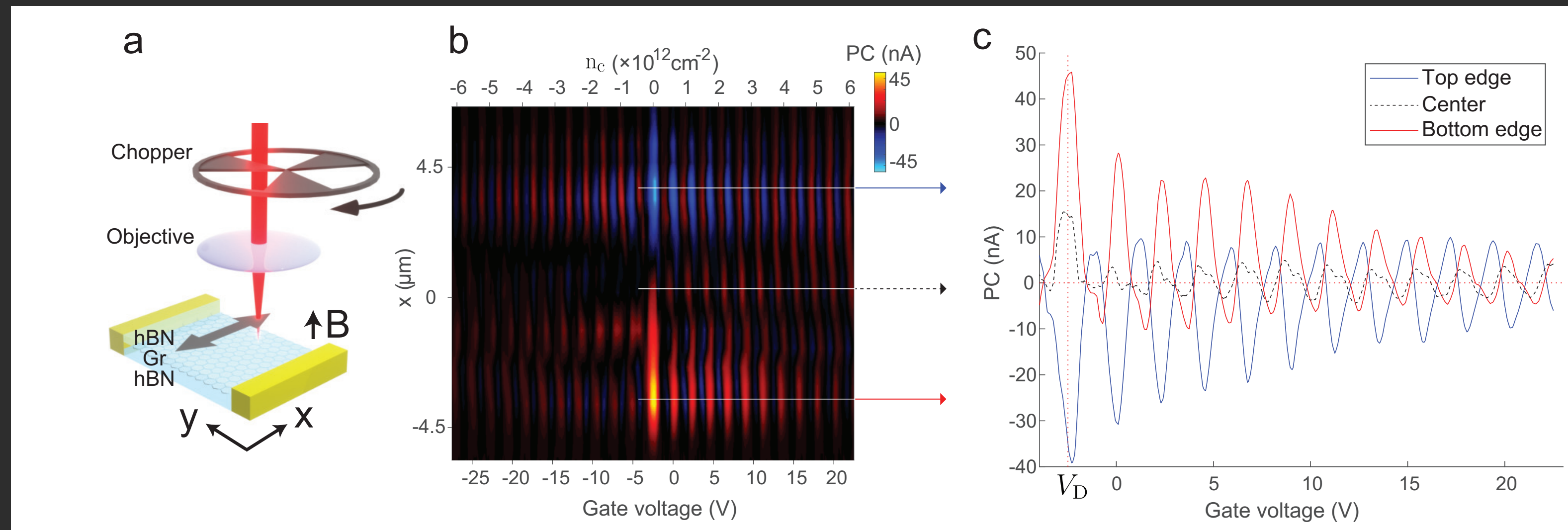
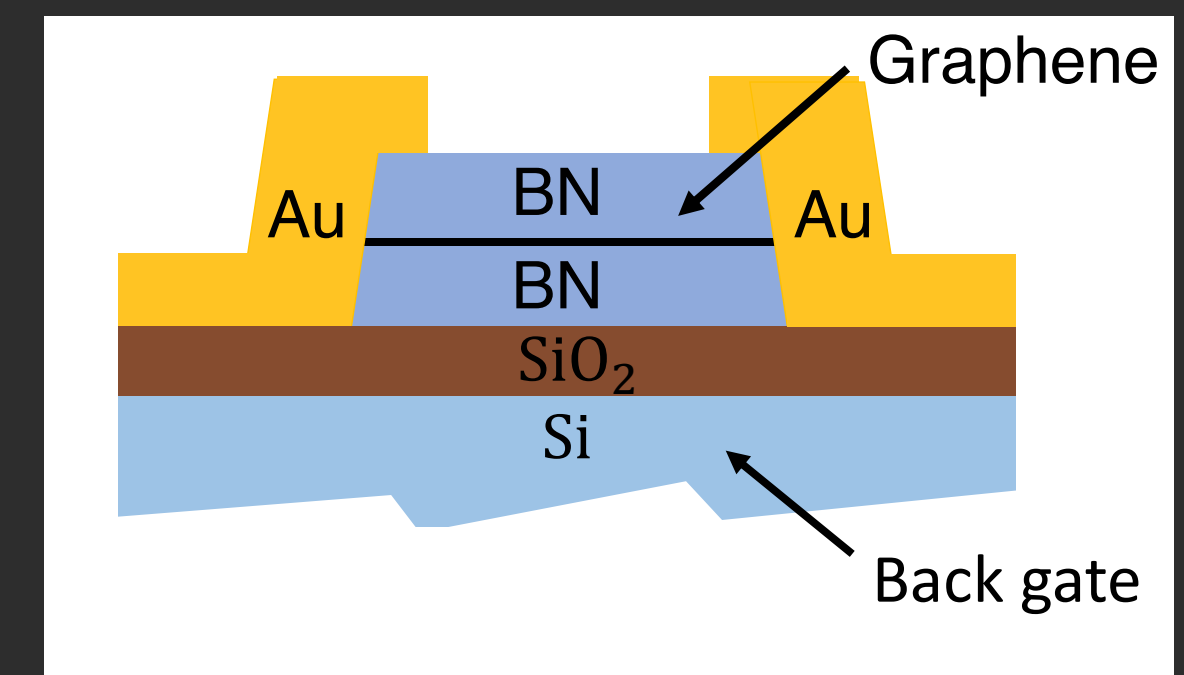
Summary and thoughts

- Using a graphene back-gated device structure at 4K
- Investigate integer quantum Hall physics with a non-equilibrium carrier distribution created by a laser



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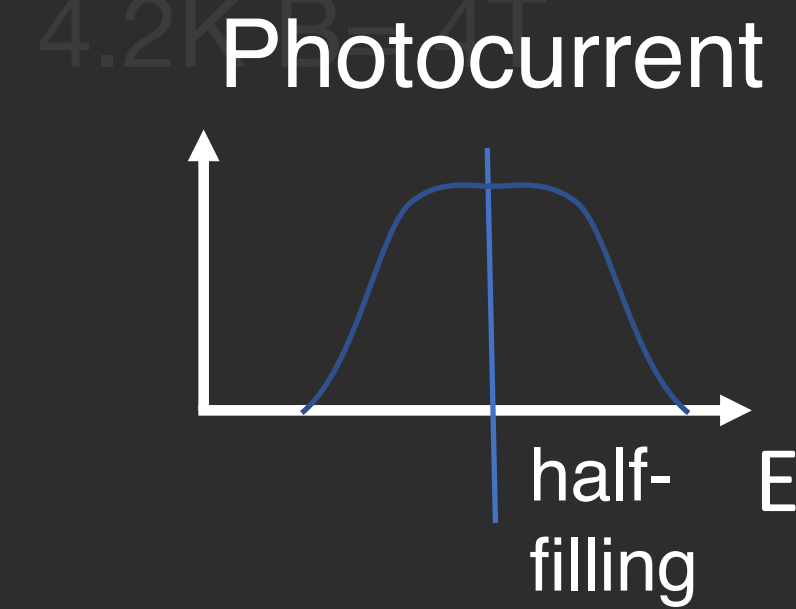
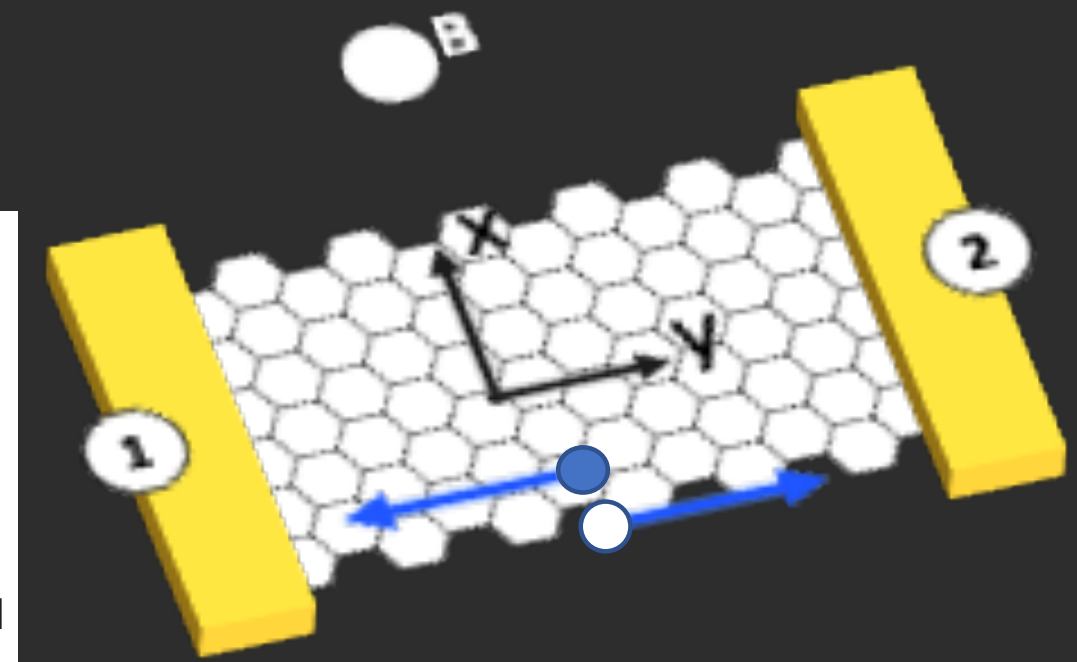
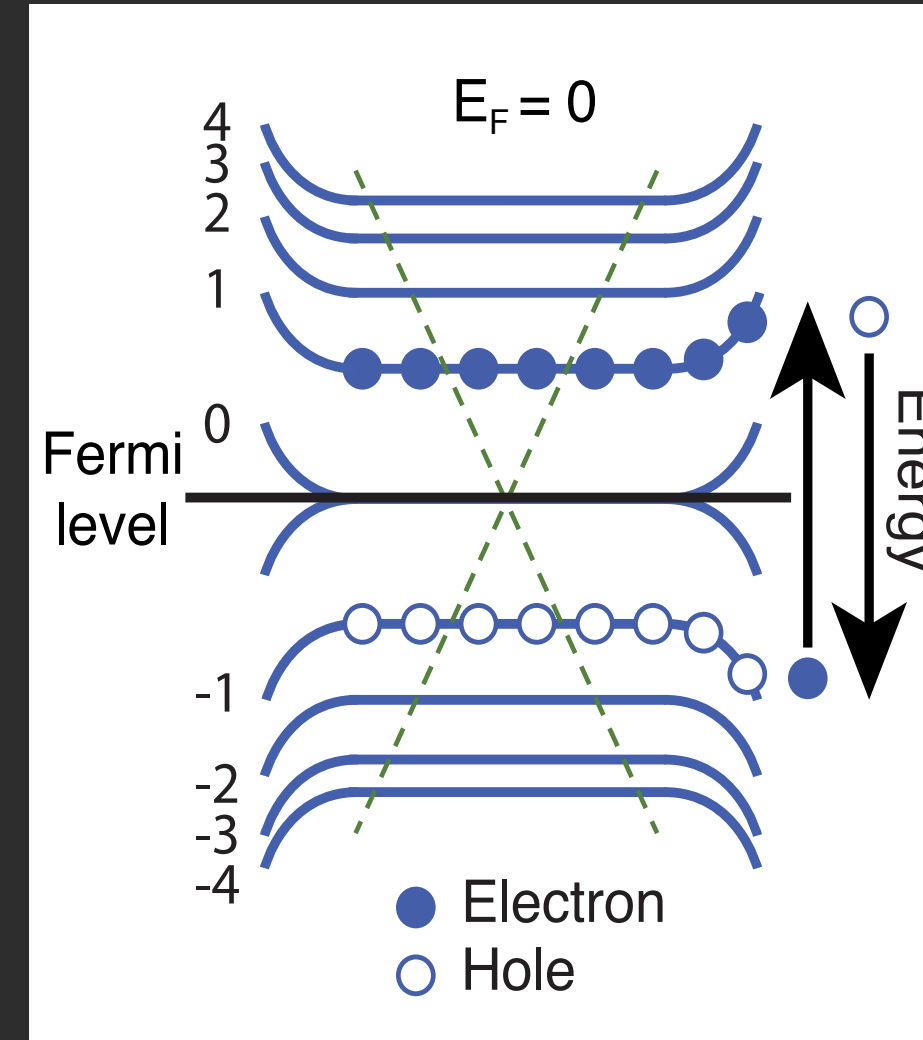
- Current occurs as carriers diffuse to edge-states before recombination
- Edge-state currents are linked to the changing availability of states for each carrier type with Fermi-level position in an LL.

- Both electron and hole currents are present in edge states.
- At the Dirac point their currents add as they come from different bands and both are majority carriers.
- Away from the Dirac point their currents subtract as the carriers are now in the same band.
- Strong evidence of carrier multiplication.

B. Cao, T. Grass, *et al.*, *ACS Nano* 2022, 16, 11, 18200–18209

glenn.solomon@adelaide.edu.au

At the Dirac Point



Away from Dirac Point

