

# Explore two-dimensional world using photoemission technique

Choongyu Hwang

*Department of Physics  
Pusan National University*

# Outline

## **1. What are the grand challenges**

## **2. Angle-resolved photoemission spectroscopy (ARPES)**

- From Einstein's photoemission to Present
- Conventional ARPES

## **3. Future directions of ARPES**

- Time resolution
- Spin and time resolution

# Grand challenges

- **Energy problem**

Solar energy, hydrogen fuel, nuclear energy ??

- **Understanding Emergent Phenomena**

Phenomena which are not the properties of the individual elementary components BUT of the assembly of such components:

Strongly correlated electron systems : high  $T_C$  superconductors

Magnetism made of non-magnetic elements

- **The ultra-fast**

Sciences in the picosecond, femtosecond, attosecond time scale

# Why do we need to study electrons?

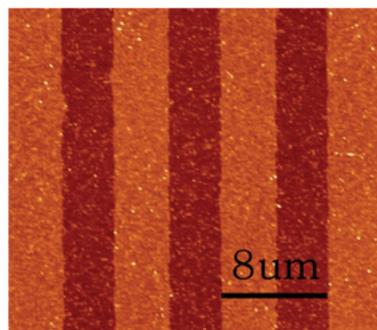
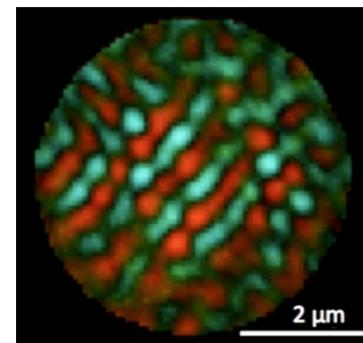
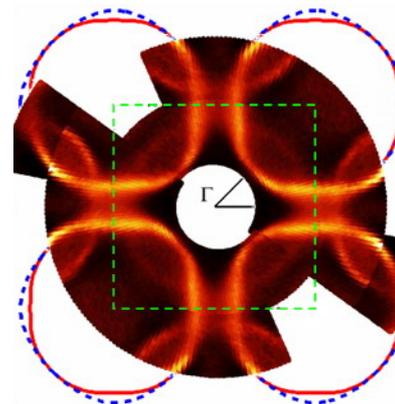
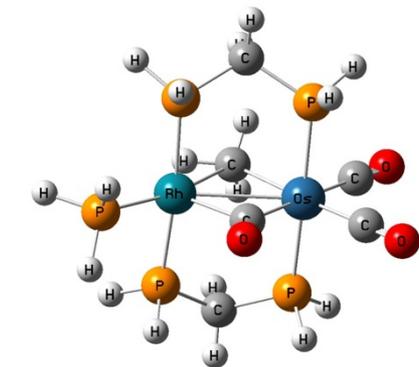
Ultimately, the

electric,

magnetic,

chemical,

mechanical,



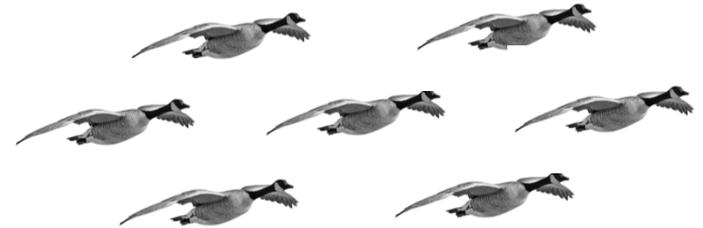
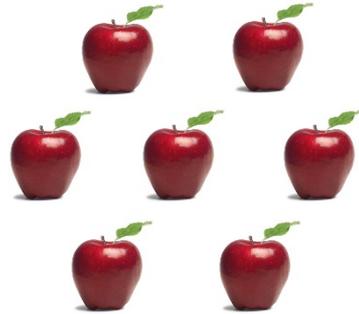
optical, thermal, and structural properties of matter depend on the behavior of electrons and location of atoms.

# Highly correlated systems

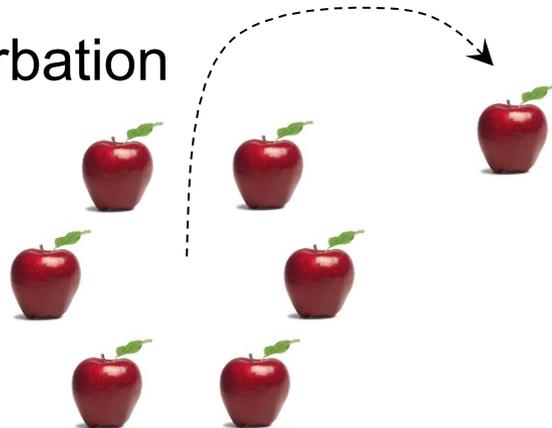
Non-correlated system

Correlated system

Ground state



With external perturbation



The responses are different due to **correlation effects!**

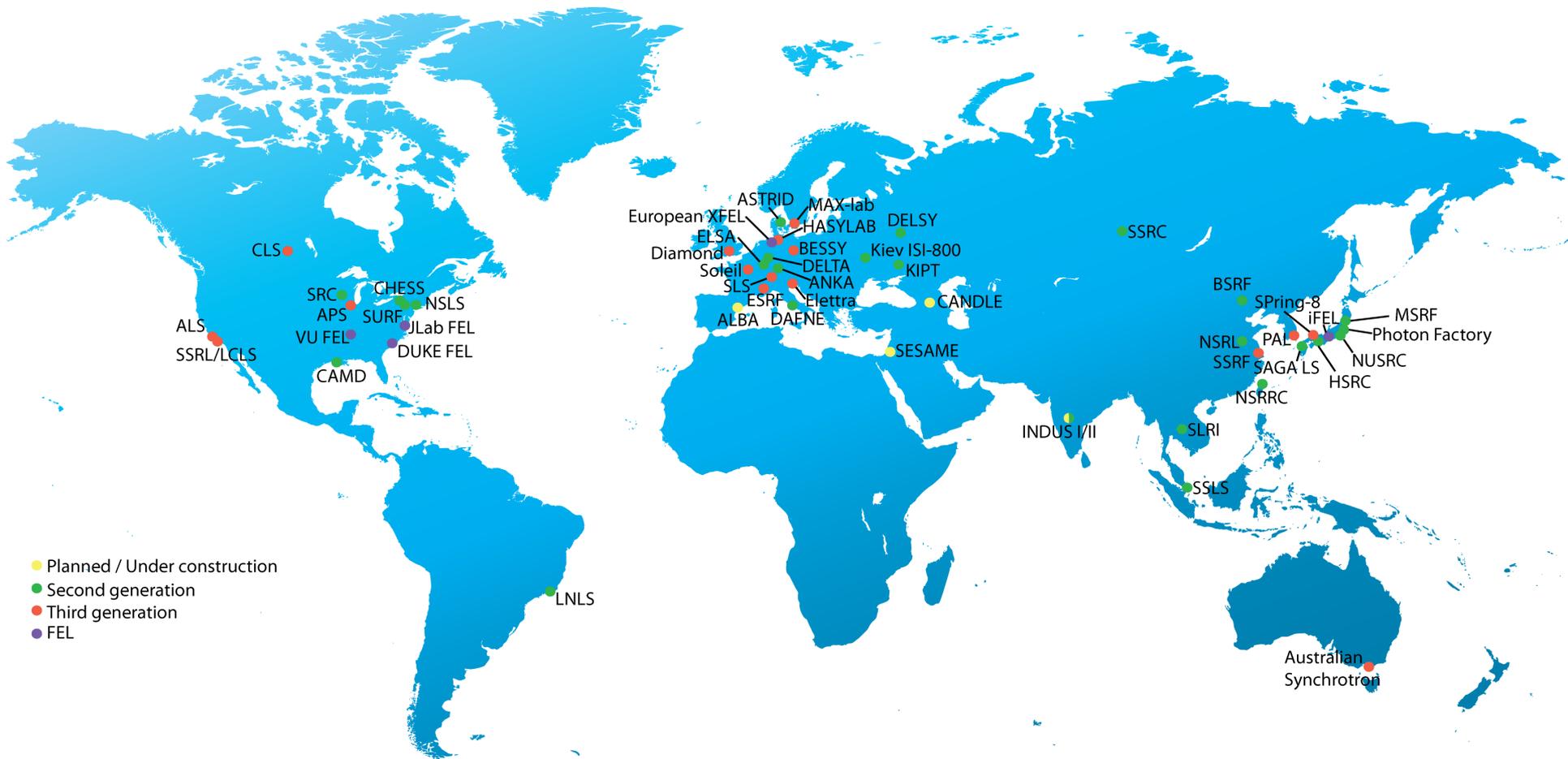


# Energy scale of importance

- **Superconducting gap** ~ 1 – 100 meV
- **Optical phonons** ~ 40 – 200 meV
- **Magnons** ~ 10 – 40 meV
- **Pseudogap** ~ 30 – 300 meV
- **Multiphonons and multimagnons** ~ 50 – 500 meV
- **Orbital fluctuations** ~ 100 meV – 1.5 eV
- **Mott gap** ~ 1 eV – 3 eV

**Requirement:** High **energy** and **momentum** resolution  
with **tunable** photons with high **flux**  
at very low **temperature**

# World map of synchrotron



# Dimensionality: 3-dimension



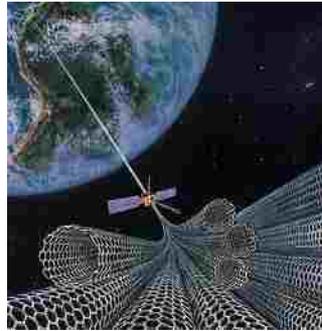
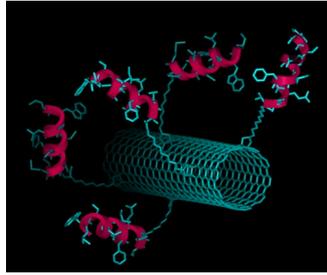
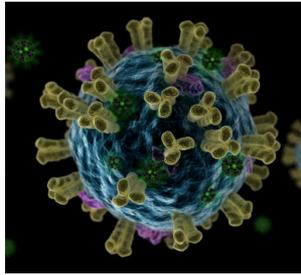
# Dimensionality: 2-dimension



# Dimensionality: 1-dimension



# Carbon Allotropes



Soft, layered substance  
Pencil, lubricant, electrode

Hardest substance  
Jewellery

Potential medical use  
- Drug delivery  
- Target cancer cells

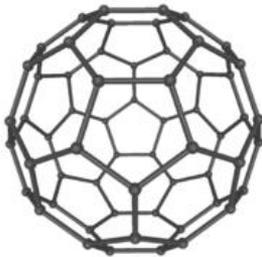
Space elevator

0d

1d

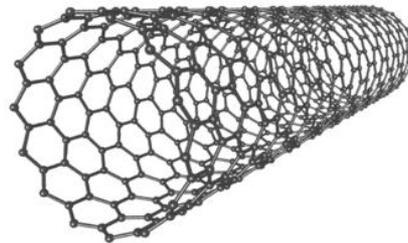
2d

3d

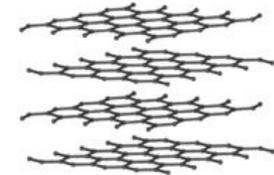


**Bulkyball  
Fullerene**

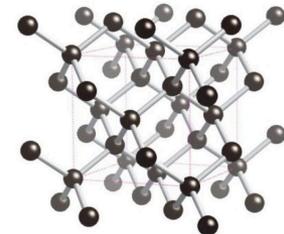
**Nobel prize in chemistry  
(1996)**



**Carbon Nanotube  
Benjamin Franklin Medal  
(2002)**



**Graphite**

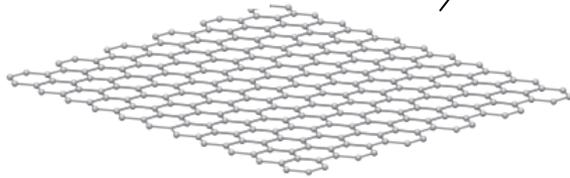


**Dimaond**

# Carbon Allotropes

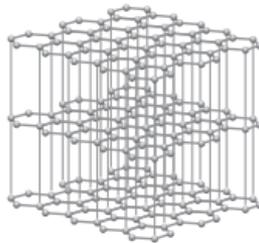
1947

Theory of Graphene  
P. R. Wallace



Massless Dirac equation of Graphene  
G. W. Semenoff

1984

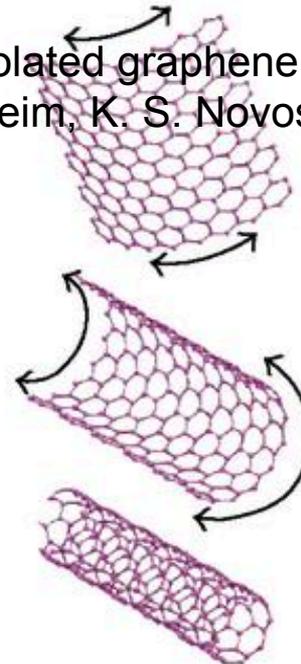


Considered only as a toy model

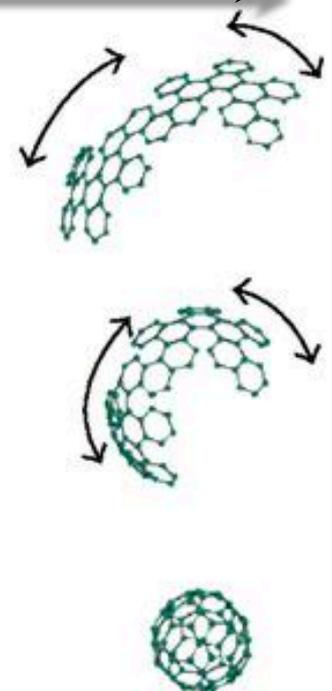
Massless Dirac Fermion proved  
K. Geim  
P. Kim

2004

Isolated graphene  
A. K. Geim, K. S. Novoselov



2005



Abnormal quantum  
Hall effect

One atom thick  
honeycomb lattice

Half metallicity

Linear  
energy spectrum

Chirality

Flexible  
display

Spin valve

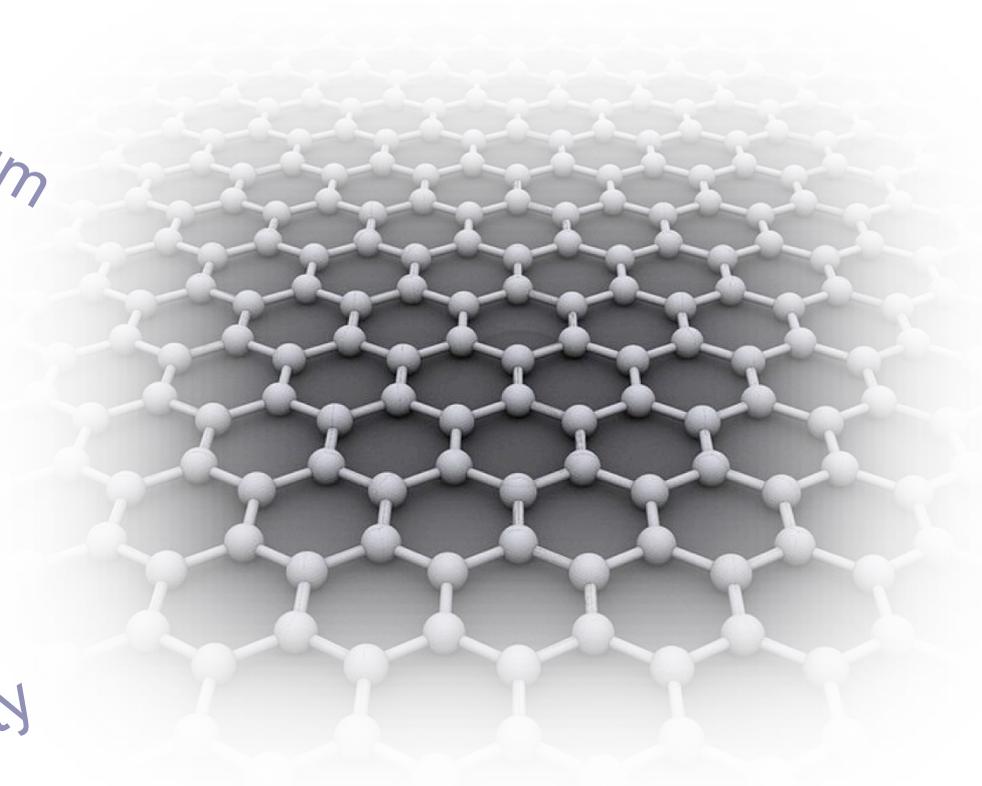
High  
Conductivity

Magnetism

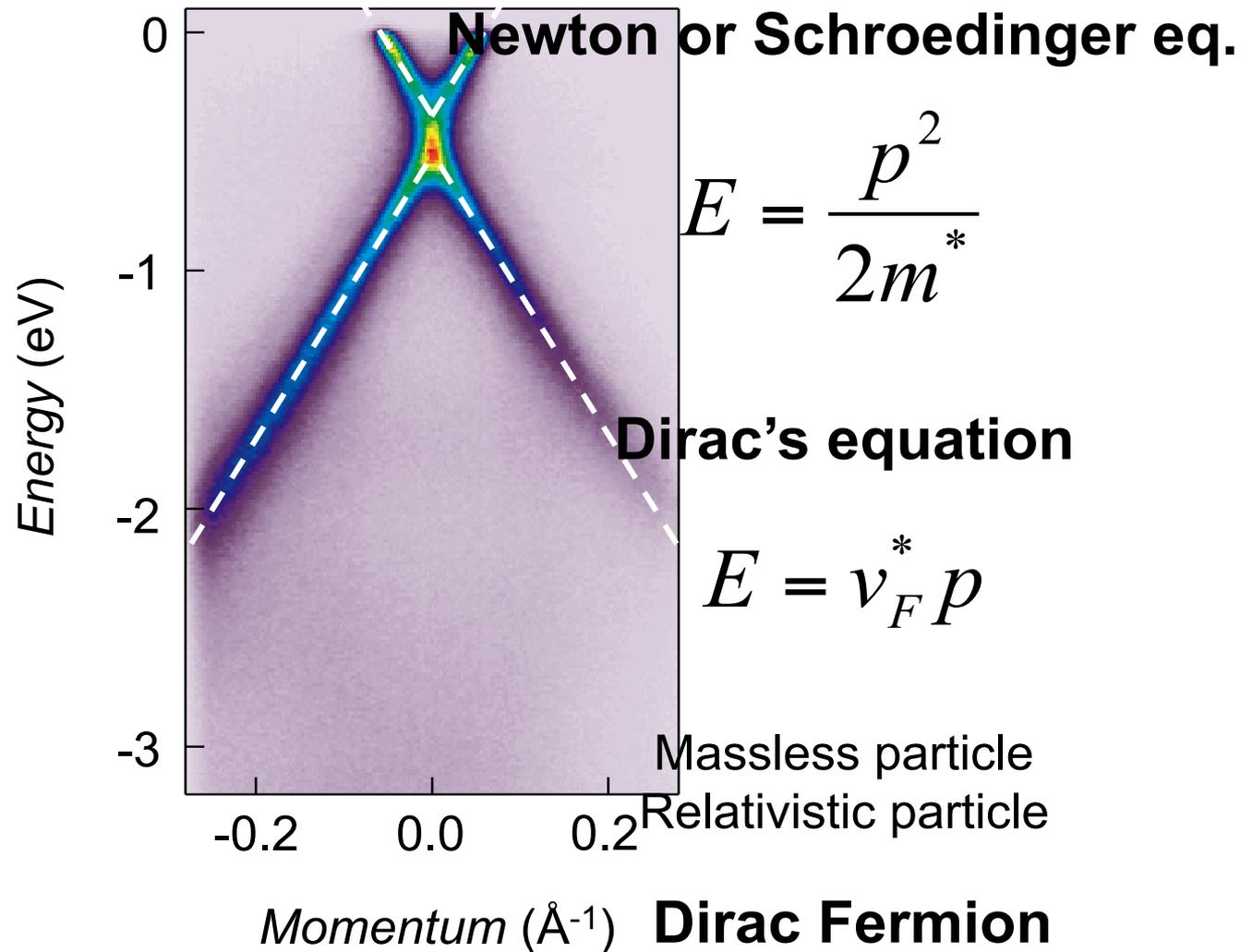
1 THz transistors

Strength and plasticity

Quantum spin  
Hall effect

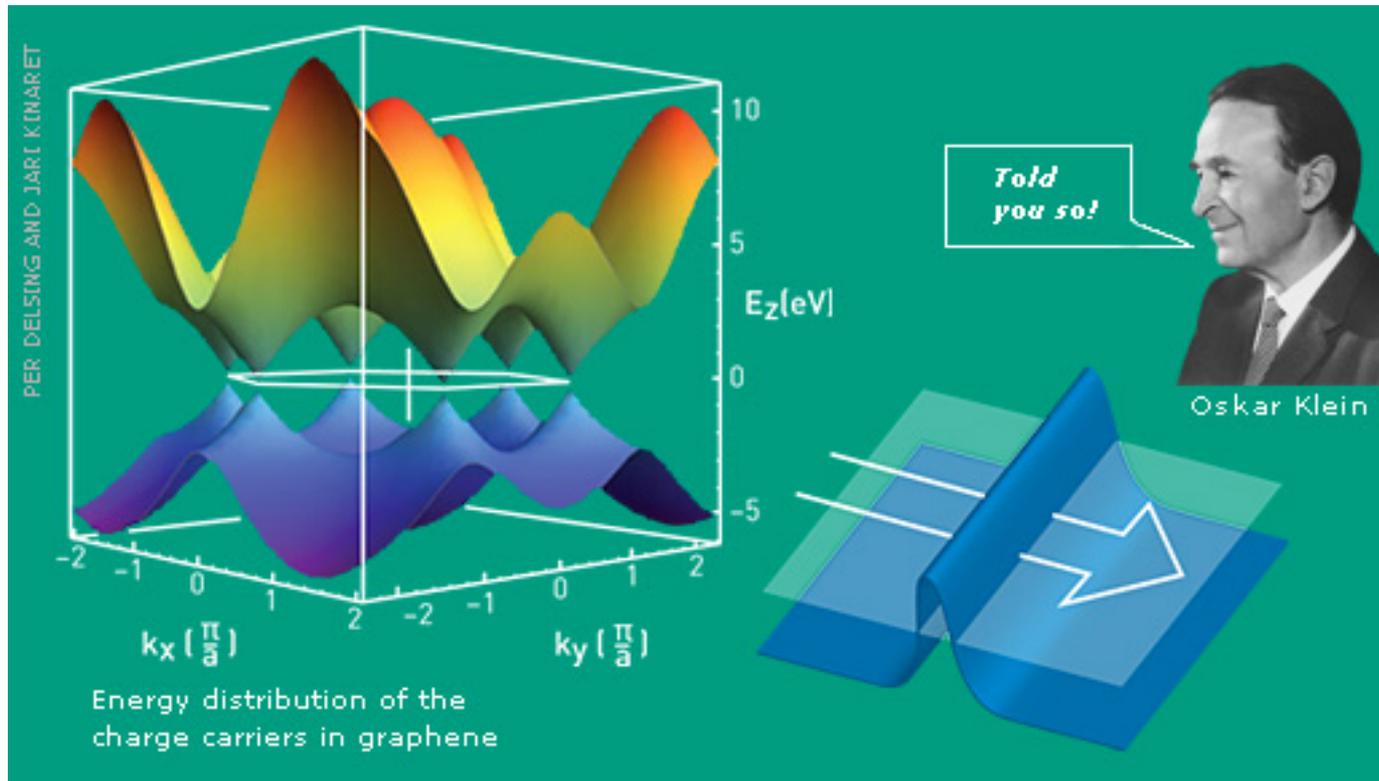


# Dirac Fermions realized in the Lab



# High energy physics in the space realized in a Lab

## Klein tunneling



- O. Klein, Zeitschrift für Physik 53, 157 (1929)
- M. I. Katsnelson et al., Nat. Phys. 2, 620 (2006)
- A. F. Young et al., Nat. Phys. 5, 222 (2009)

# What has graphene done for us so far?

## Companies

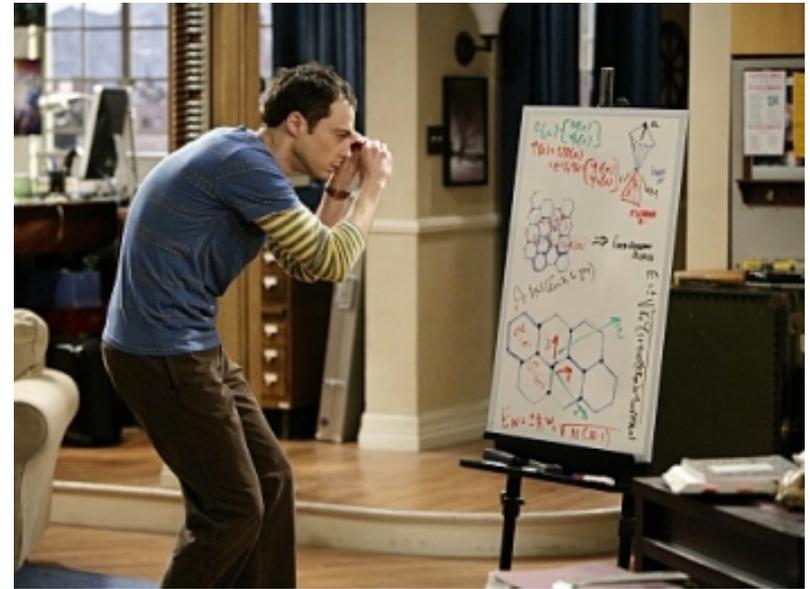


# What has graphene done for us so far?

## Companies



## Entertainment



**BANG**  
the **BIG THEORY WIKI**

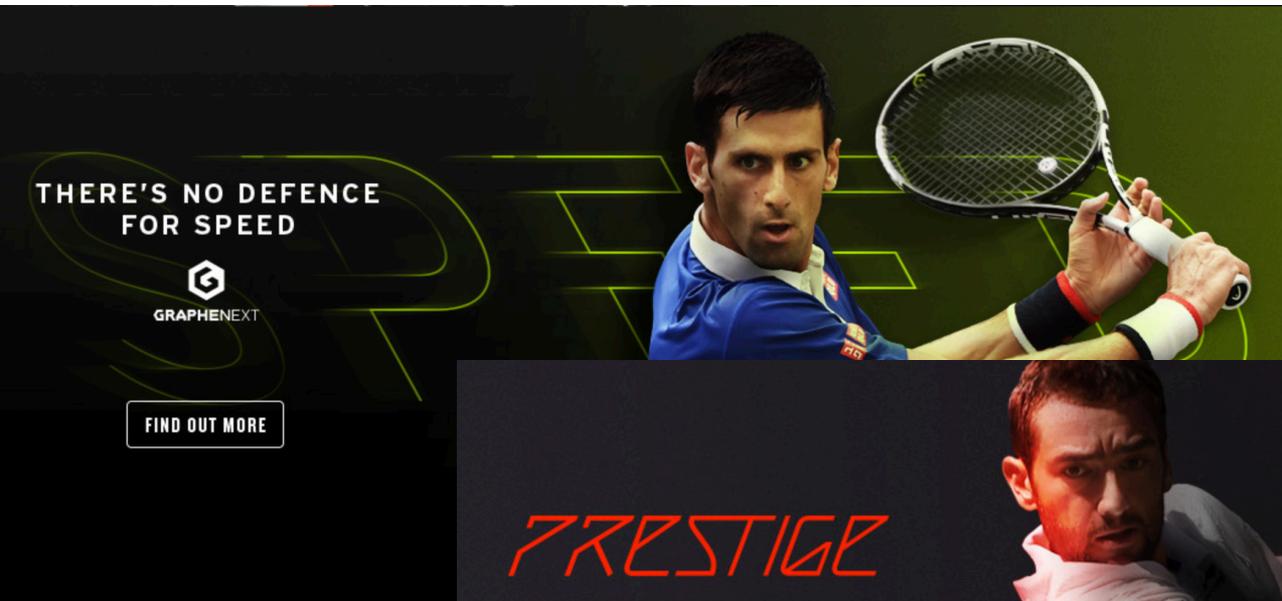
Season 3, Episode 14

# What has graphene done for us so far?

Companies

Entertainment

Sports



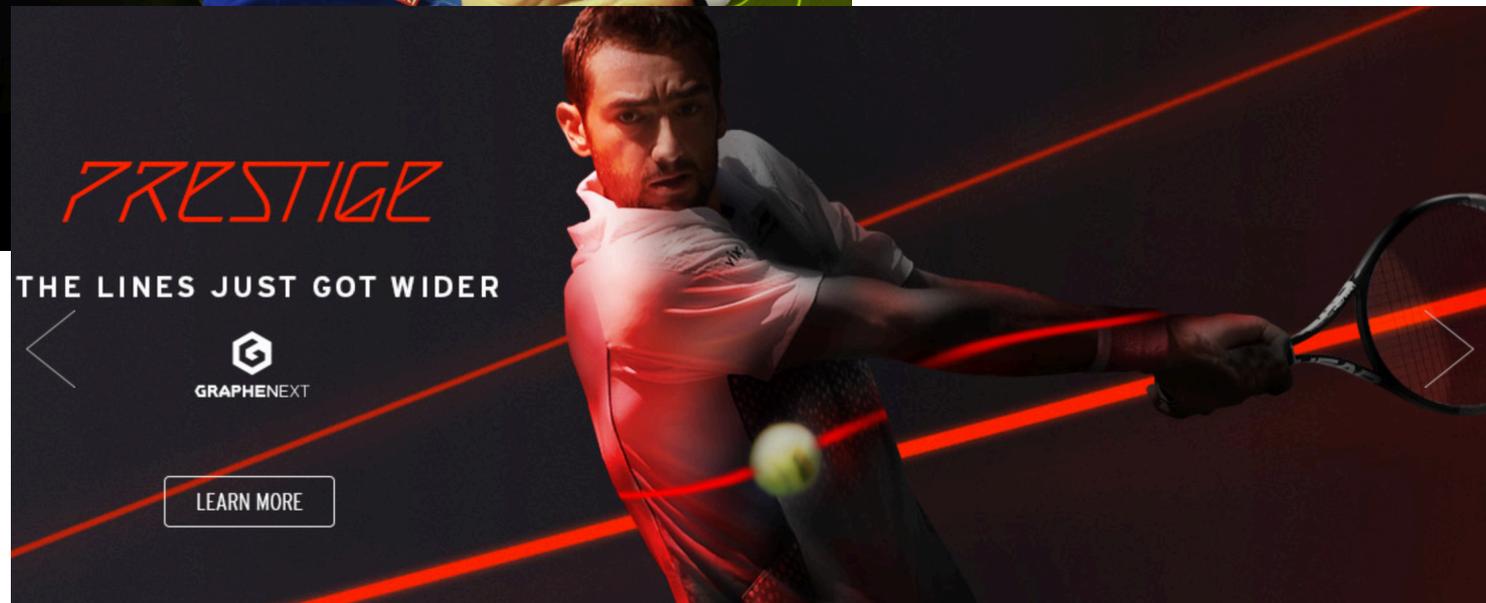
THERE'S NO DEFENCE FOR SPEED

GRAPHENEXT

FIND OUT MORE

This advertisement features a photograph of tennis player Novak Djokovic in a blue shirt, captured in a backhand swing. The background is dark with green neon-style lines forming a grid pattern. The text 'THERE'S NO DEFENCE FOR SPEED' is in white, with 'GRAPHENEXT' below it and a 'FIND OUT MORE' button at the bottom left.

WTA Rank 1  
Novak Djokovic



PRESTIGE

THE LINES JUST GOT WIDER

GRAPHENEXT

LEARN MORE

This advertisement features a photograph of tennis player Andy Murray in a white shirt, captured in a forehand swing. The background is dark with red neon-style lines forming a grid pattern. The text 'PRESTIGE' is in red, 'THE LINES JUST GOT WIDER' is in white, 'GRAPHENEXT' is below it, and a 'LEARN MORE' button is at the bottom left.

WTA Rank 2  
Andy Murray

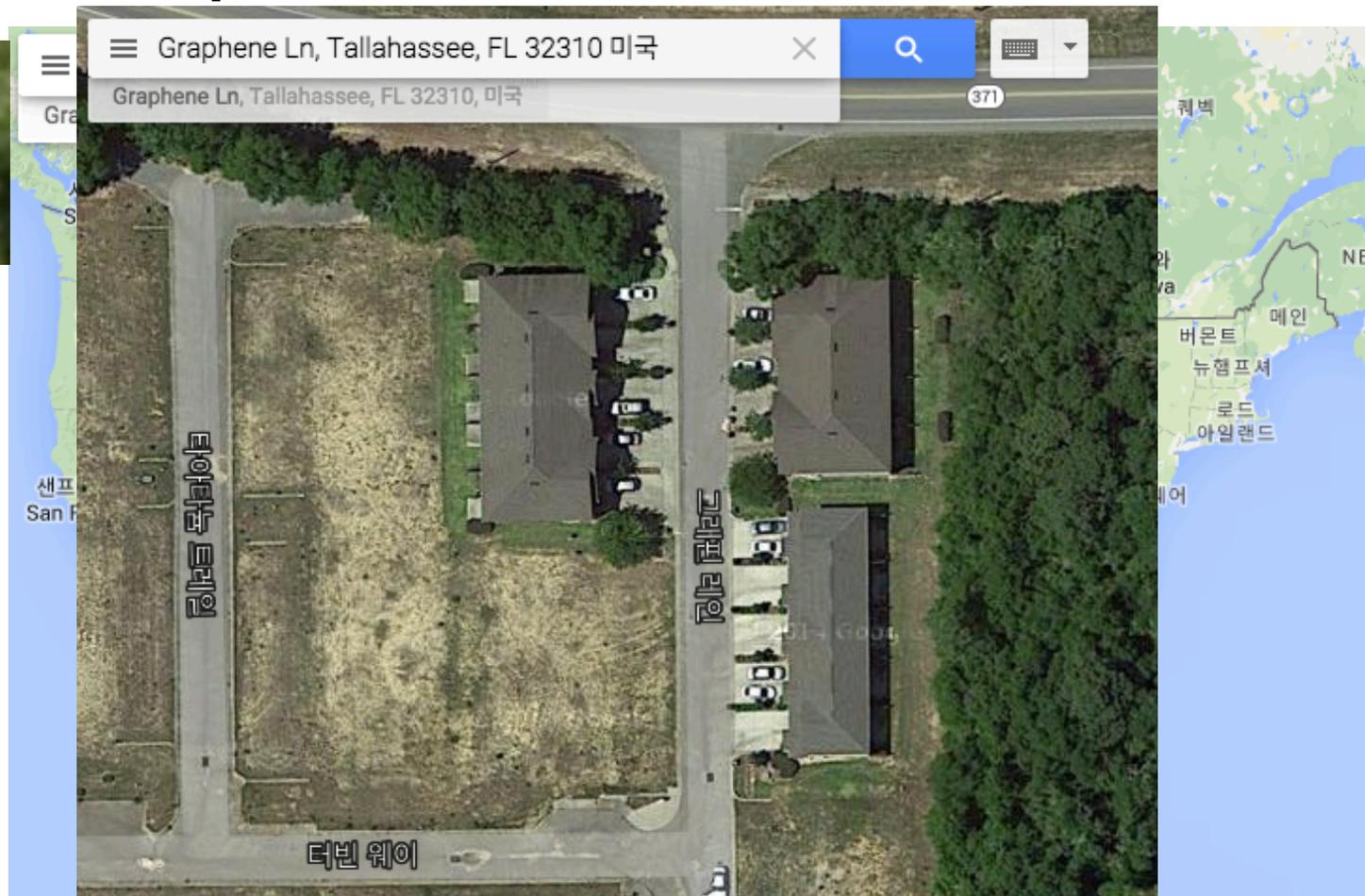
# What has graphene done for us so far?

Companies

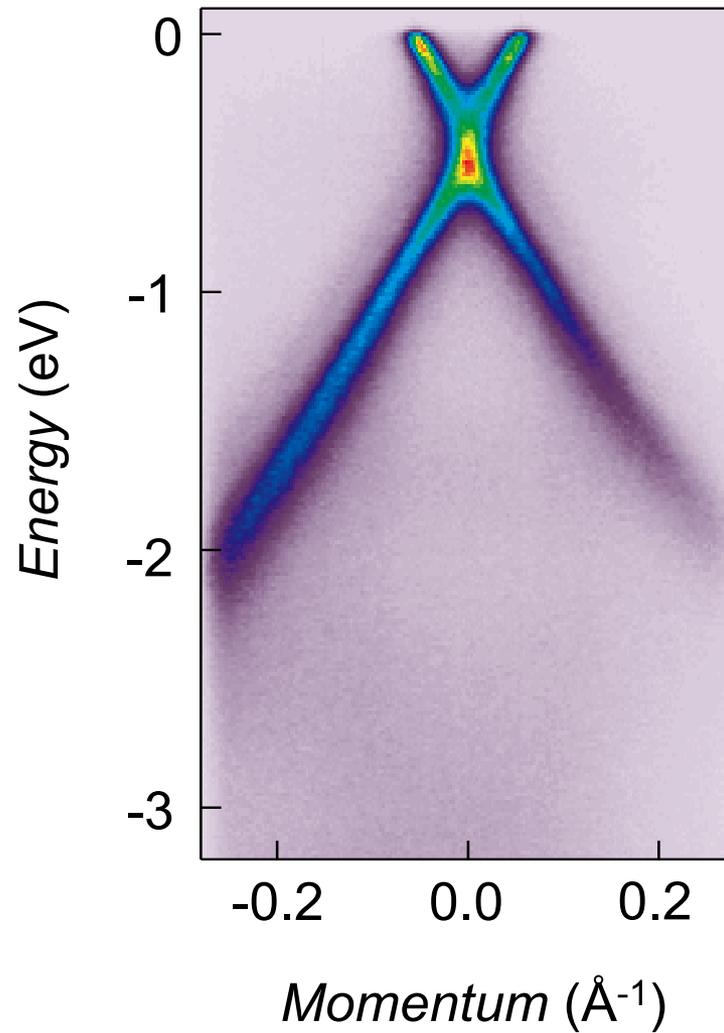
Entertainment

Sports

Graphene Ln, Tallahassee, FL, USA



# Dirac Fermions realized in the Lab



# Outline

## 1. What are the grand challenges

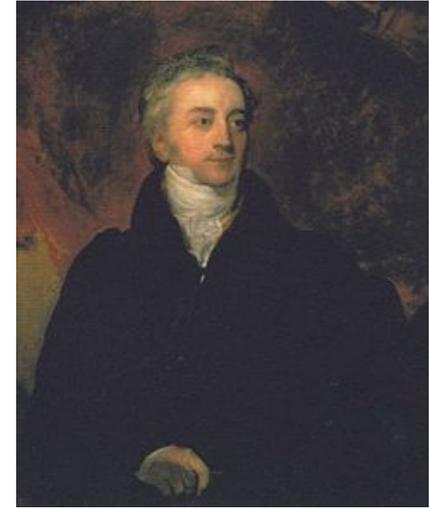
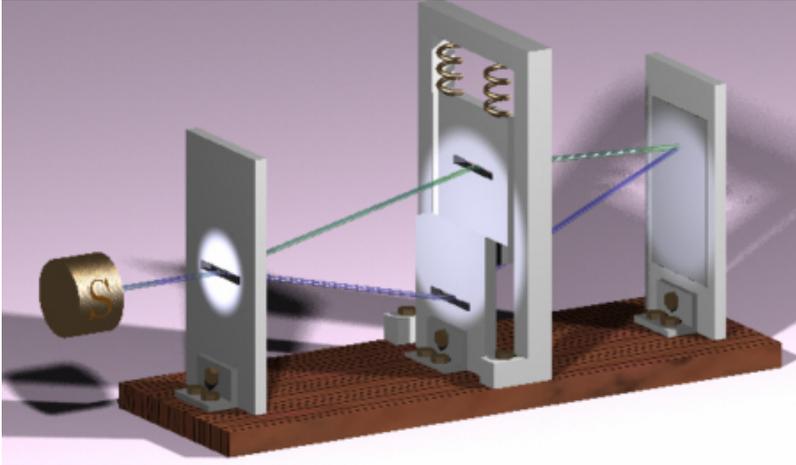
## 2. Angle-resolved photoemission spectroscopy (ARPES)

- From Einstein's photoemission to Present
- Conventional ARPES

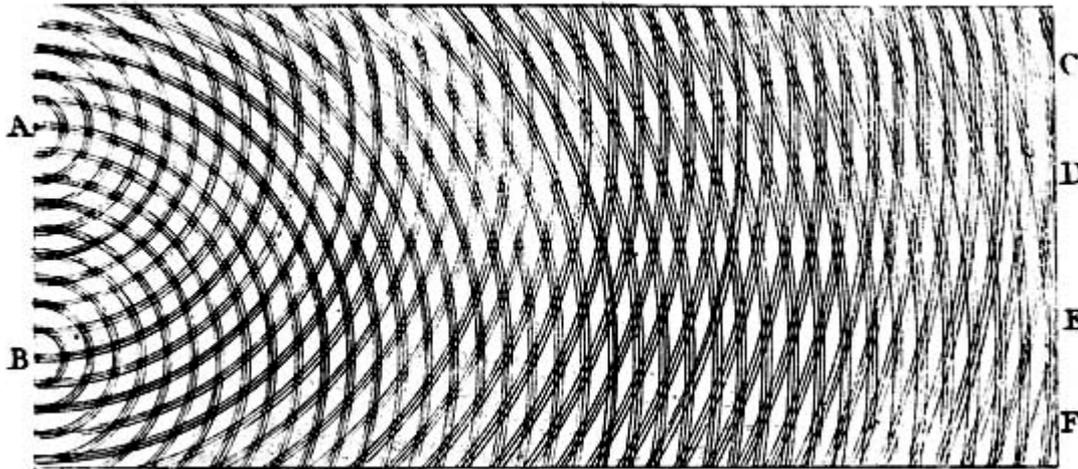
## 3. Future directions of ARPES

- Time resolution
- Spin and time resolution

# Light as a wave: 1803



Thomas Young  
Light as a wave (1803)



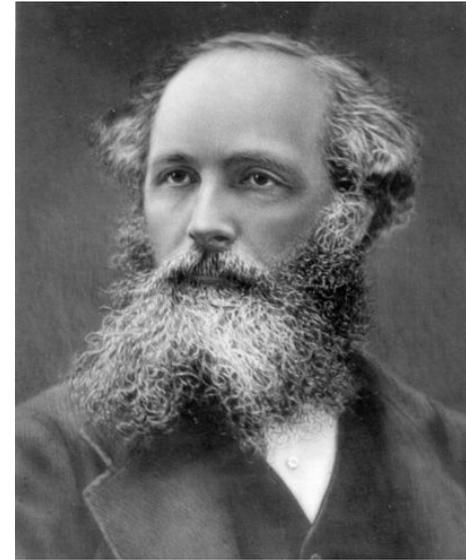
Young's sketch of the light waves emerging from two narrow slits  
*A course of lectures on natural philosophy and the mechanical arts, Vol. 1 (1807)*

# Maxwell

Describe the propagation of Electromagnetic radiation

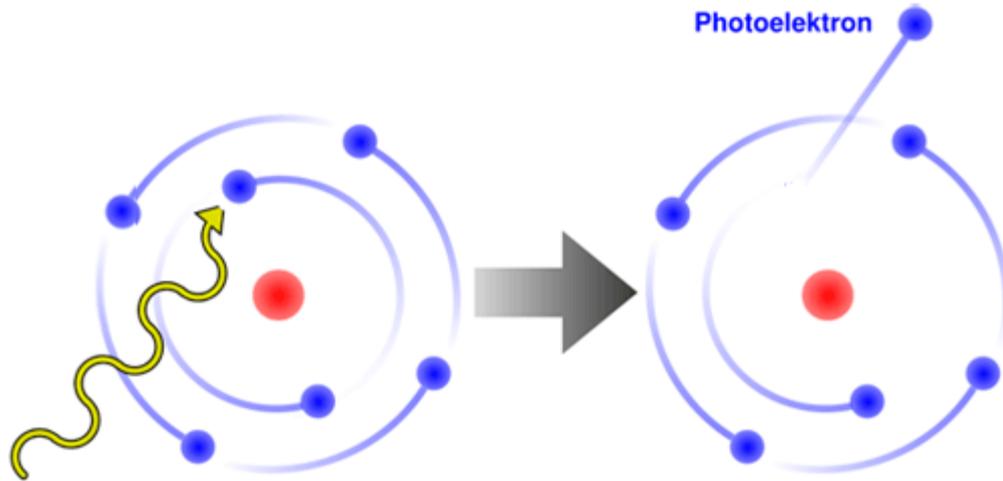
$$E_z = E_{z0} \cos\left[w\left(t - \frac{z}{c}\right) + \theta\right]$$

$$c = \frac{1}{(\epsilon_0 \mu_0)^{1/2}}$$



James C Maxwell  
Electromagnetism (1865)

# Photoelectric effect



Heinrich Hertz  
Photoelectric effect (1887)

# Einstein's equation

## Einstein's Photoelectric Equation

The electron leaves  
the body with energy

$$\frac{1}{2}mv^2 = h\nu - P$$

where  $h$  is planck's constant,  $\nu$  is  
the light frequency, and  $P$  is the  
work that electron has to do in  
leaving the body.



Albert Einstein  
Photoelectric equation (1905)

# Einstein's 1905 – age 26

**“ A storm broke loose in my mind”**

**March**

**Quantum nature of light and photoelectric effect**

**1921 Nobel prize**

**May**

**On “Brownian Motion” or random bouncing of small particles in a surrounding fluid**

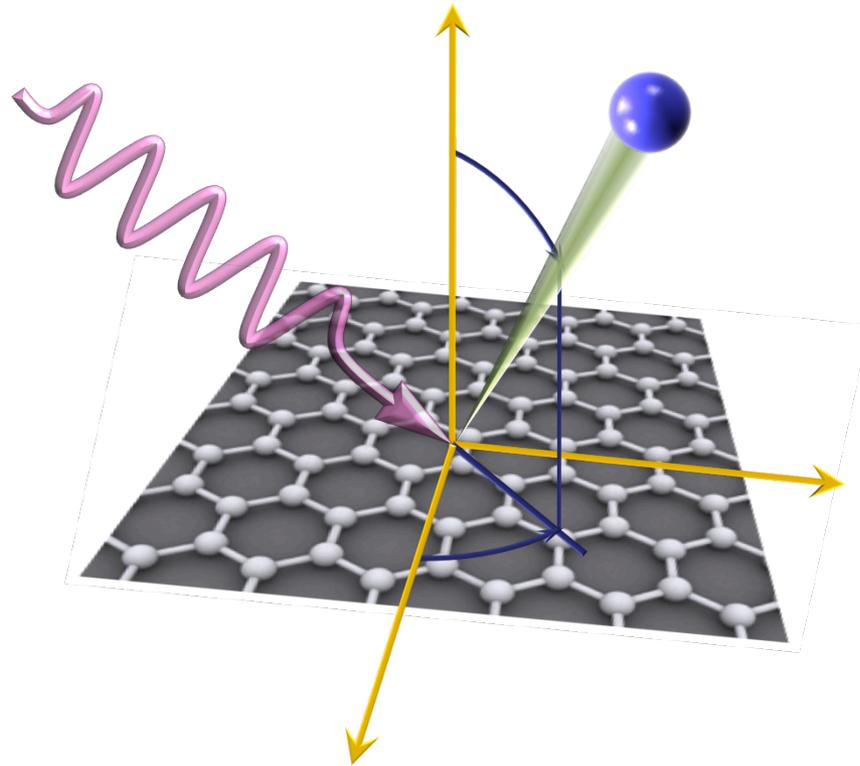
**June**

**Special theory of relativity  $c$ =speed of light  
 $c$  same in all reference frames and is max speed**

**September**

**Consequence of Special Relativity  $E = mc^2$**

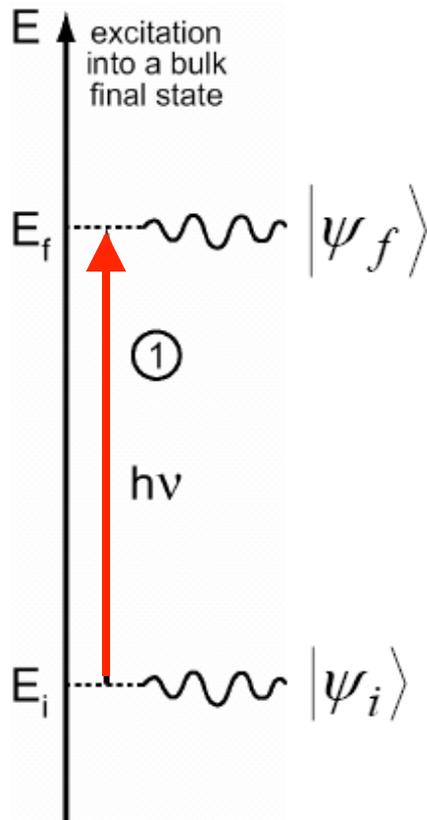
# How to measure electrons?



# Three step model

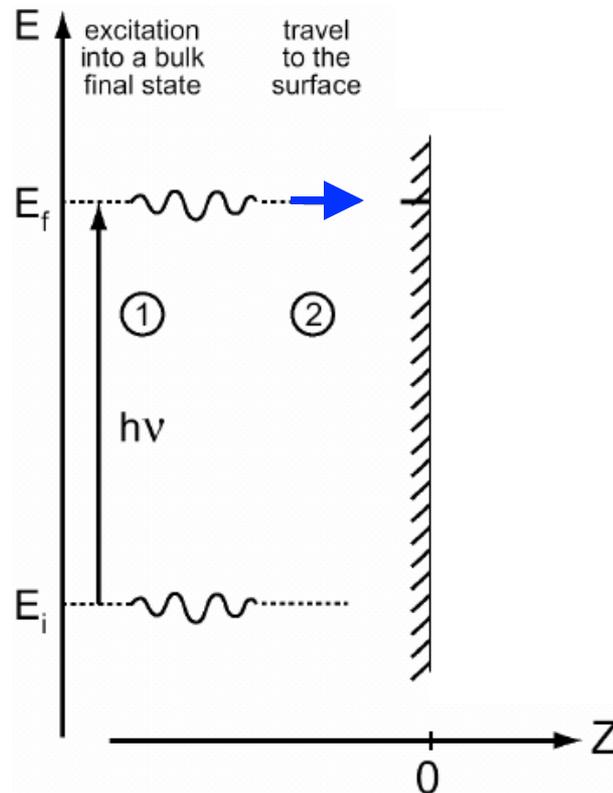
## Step I

### Transition in the solid



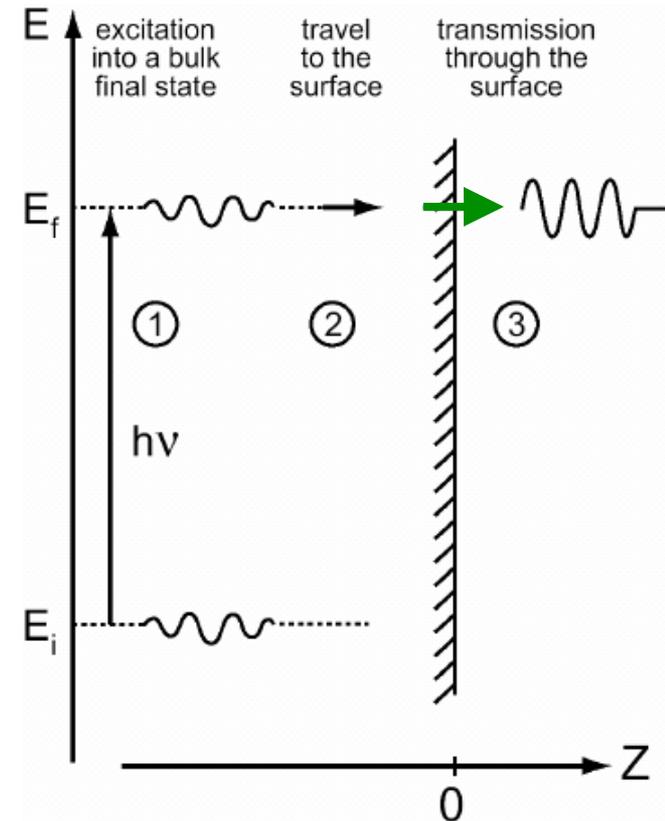
## Step II

### Transmission to surface



## Step III

### Transmission into vacuum

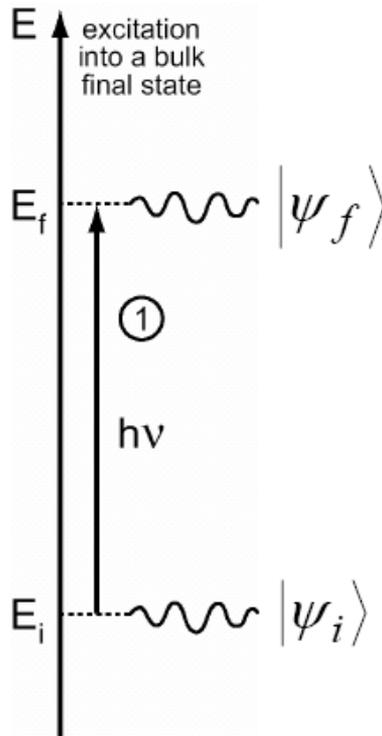


# Step I: photoexcitation process

$$w_{i \rightarrow f} \propto \left| \langle \psi_f | \vec{A} \cdot \vec{p} | \psi_i \rangle \right|^2 \delta(E_f - E_i - h\nu)$$

Typical photon wavenumber

$$\kappa = 2\pi \frac{E}{hc} = 2\pi \frac{E [\text{eV}]}{12400 [\text{eV} \cdot \text{\AA}]}$$
$$= .01 \text{ to } .05 \text{ \AA}^{-1} \quad (\text{for } E = 20 \text{ to } 100 \text{ eV})$$



- Photon impact very little momentum in PES process
- Photon-stimulated transition not allowed

Energy conservation:

$$E_f = E_i + h\nu$$

Momentum conservation:

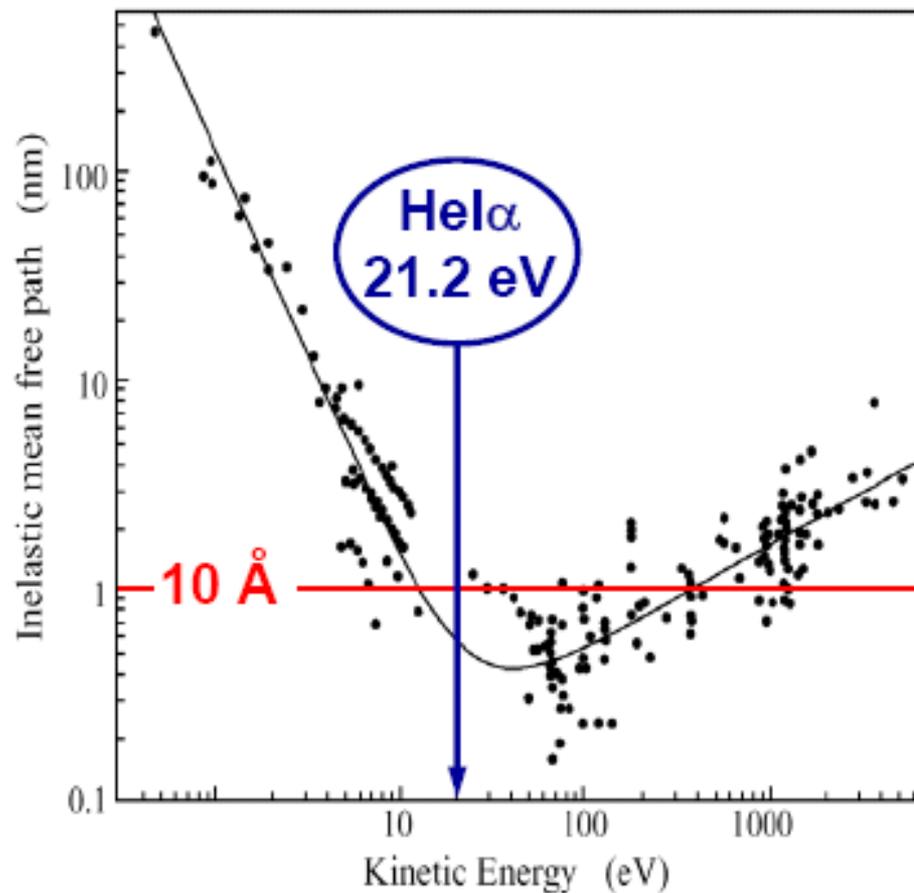
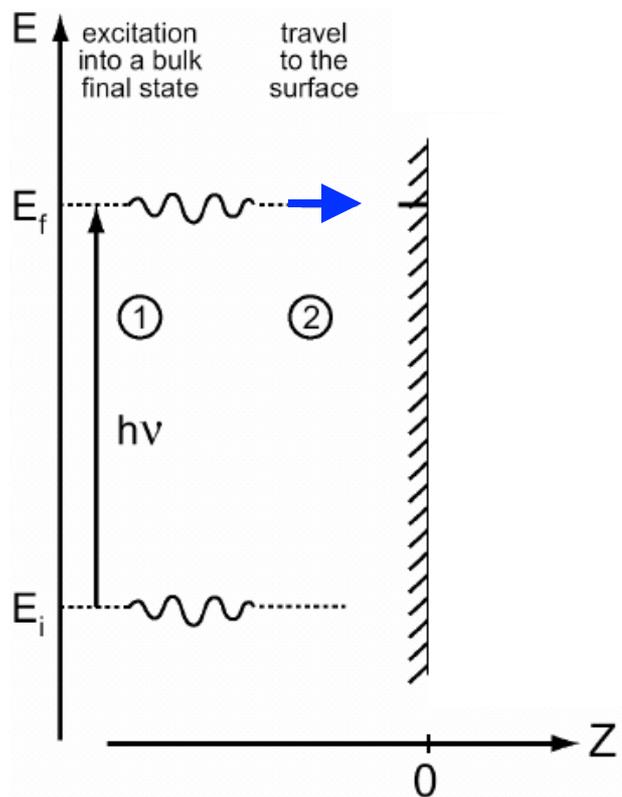
$$\underline{k}_f = k_i + \mathbf{G} + \cancel{k_{\text{photon}}}$$

only "vertical" transitions

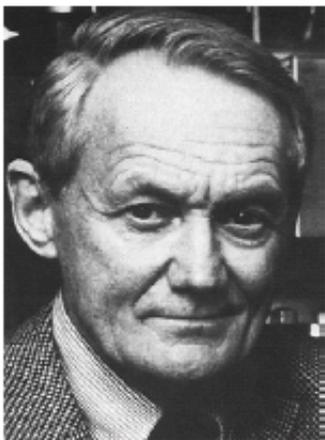
# Step II: transport to surface

## Step II

### Transmission to surface



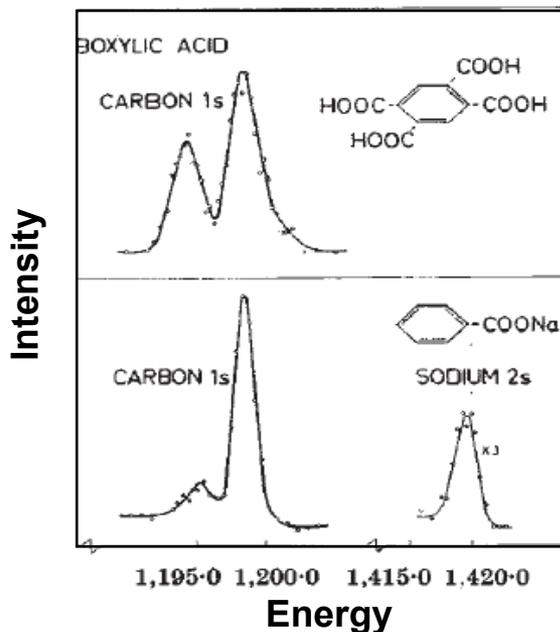
# Photoemission core-level spectroscopy



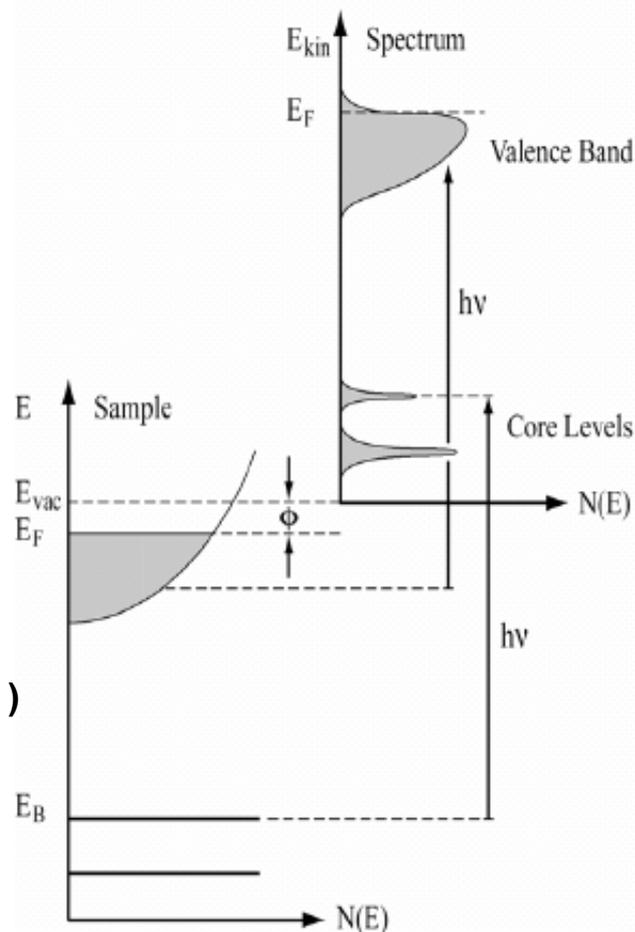
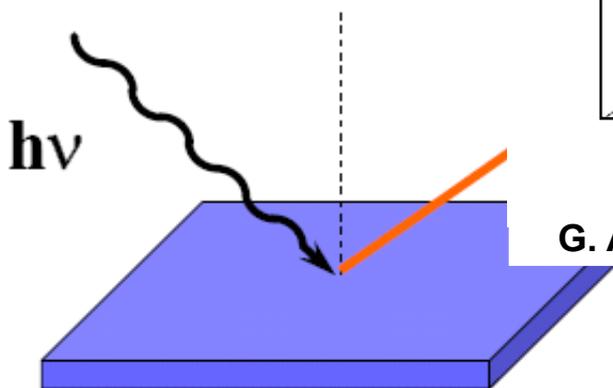
K.M. Siegbahn

energy conservation:

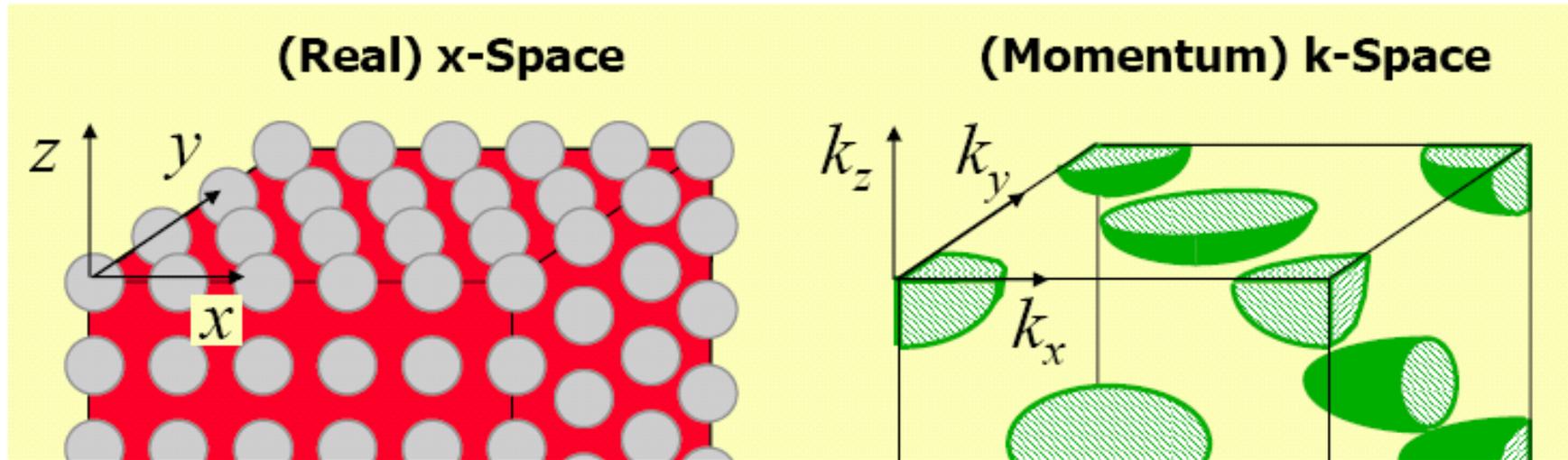
$$E_{kin} = h\nu - E_B - \Phi$$



G. Axelson *et al.*, Nature (1967)



# valence electron & momentum resolution?



Many properties of solids are determined by  
electrons near  $E_F$

(conductivity, magnetoresistance, superconductivity,  
magnetism)



Only a narrow energy slice around  $E_F$   
is relevant for these properties  
( $kT = 25$  meV at room temperature)

# Outline

## 1. What are the grand challenges

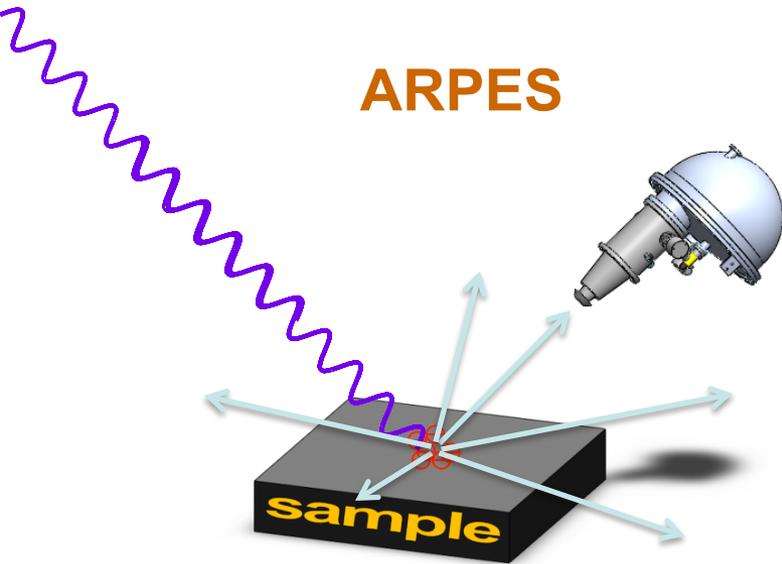
## 2. Angle-resolved photoemission spectroscopy (ARPES)

- From Einstein's photoemission to Present
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## 3. Future directions of ARPES

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- Spin and time resolution

# Providing momentum resolution



ARPES

Successful with QMs:

HTC cuprates

CMR manganites

Graphene

Topological Insulators

⋮

**Energy conservation**

$$E_B = \hbar\omega - E_K - \Phi_A$$

**Angle / momentum conservation**

$$k_{\parallel} \propto \sqrt{E_K} \sin \theta$$

**Photoelectron intensity**

$$I(k, \omega) \propto A(k, \omega) |M(k, \omega)|^2 f(\omega)$$

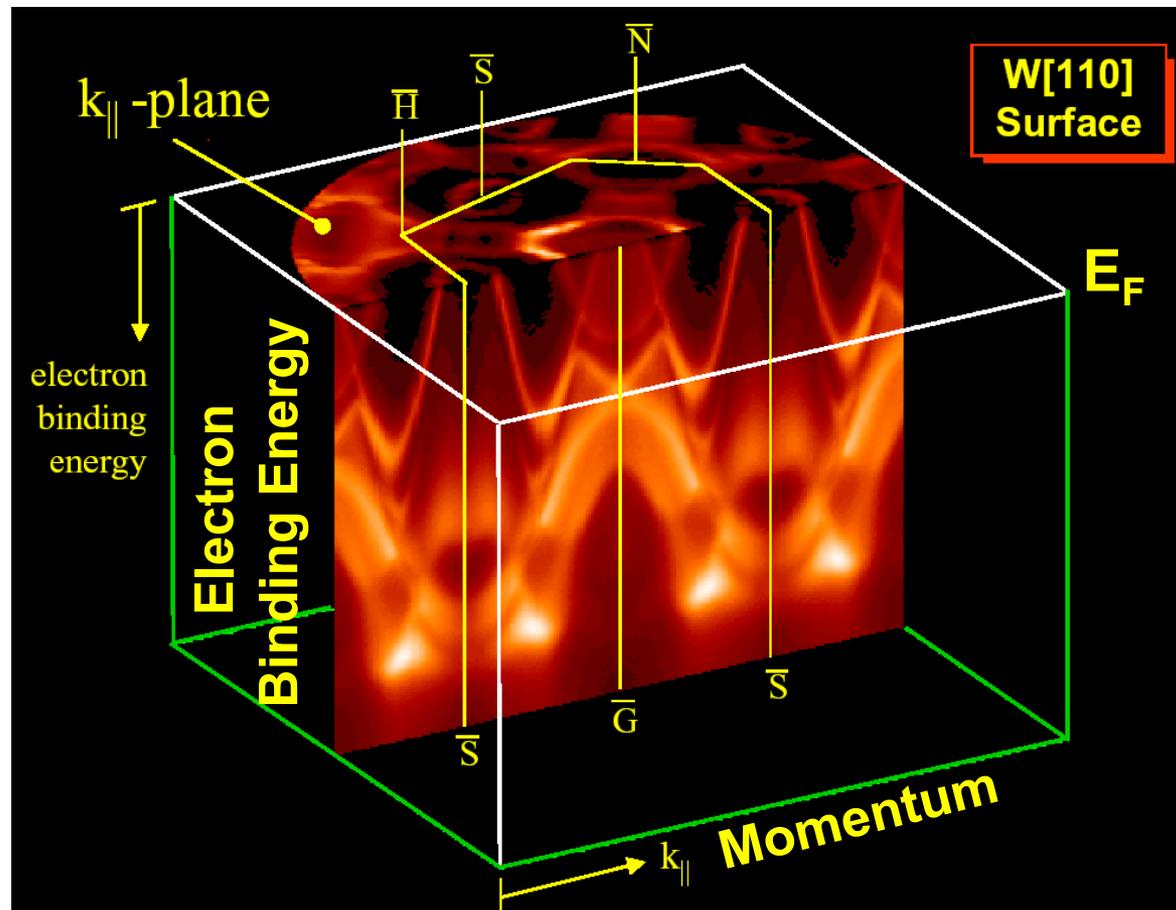
$A(k, \omega)$  spectral function

$M(k)$  Matrix element

$f(\omega)$  Fermi function

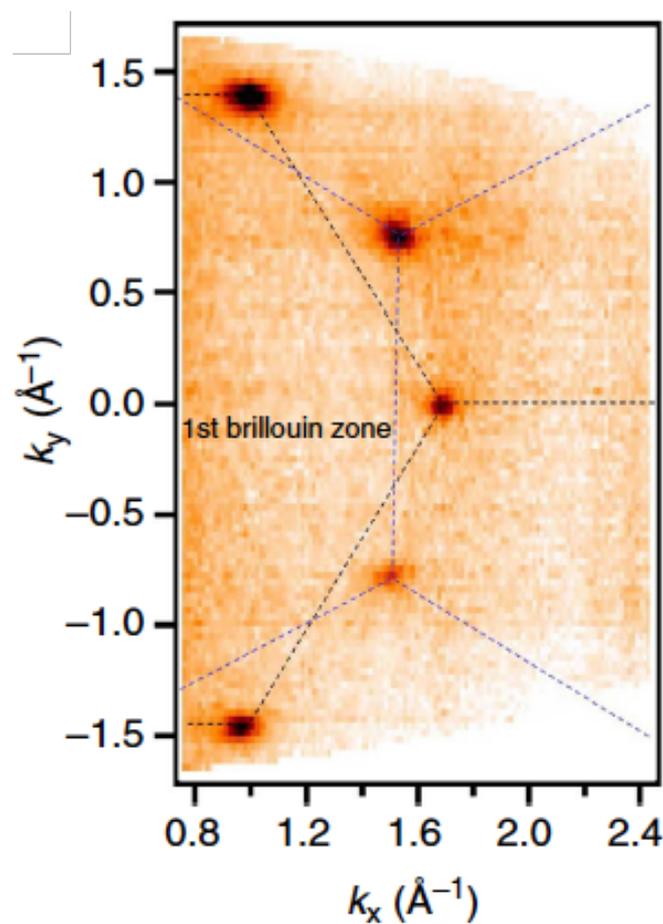
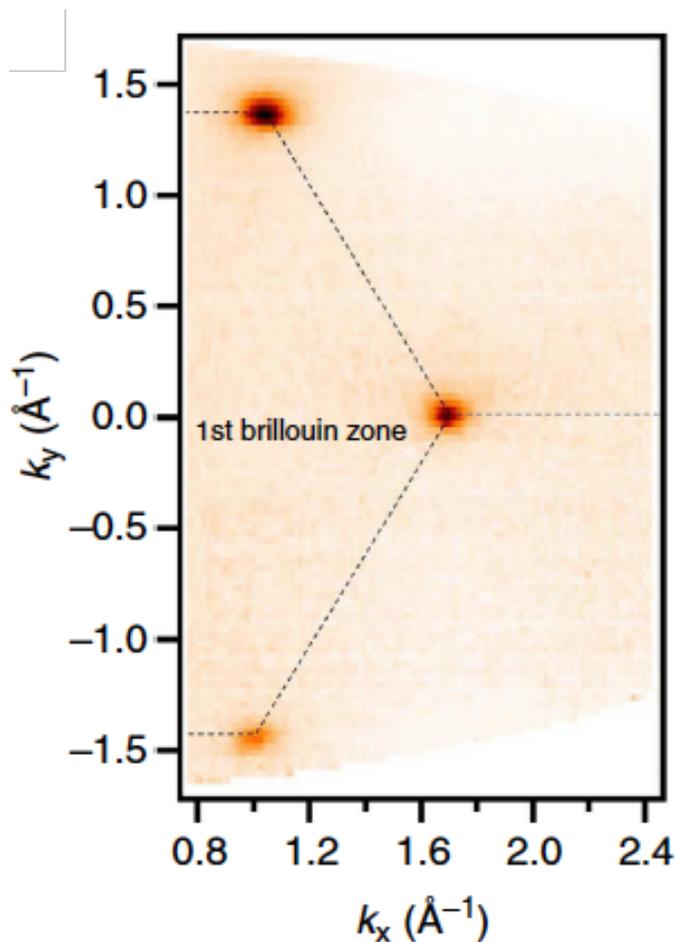
# What do we learn from momentum resolution?

- Energy vs. wave vector ( $k$ ) - Band structure
- Fermi surface
- Charge carrier density



# What do we learn from momentum resolution?

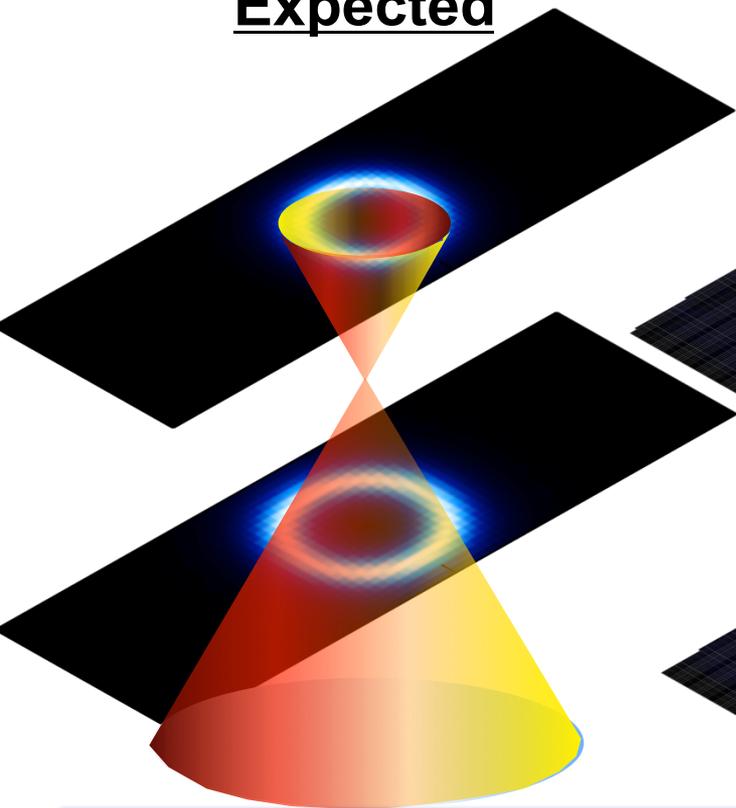
- Geometric structure



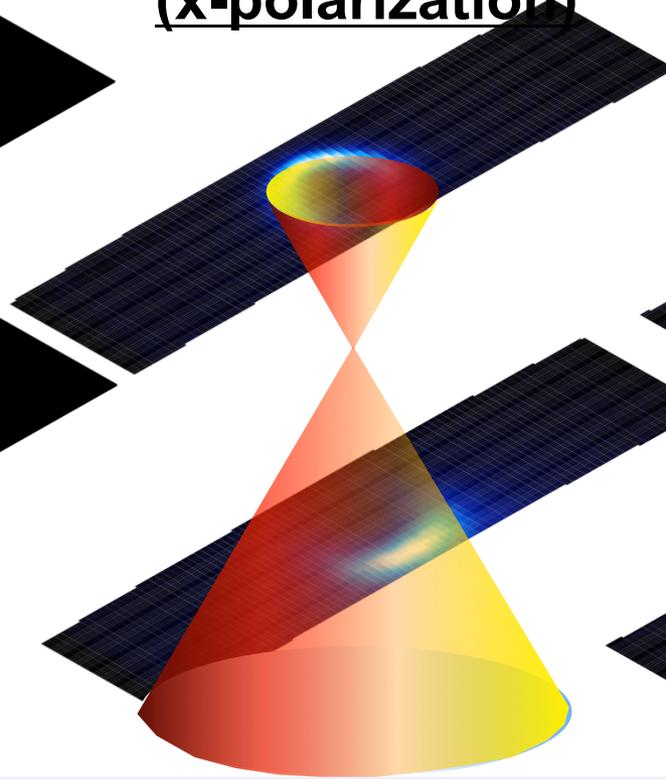
# What do we learn from momentum resolution?

**Graphene**

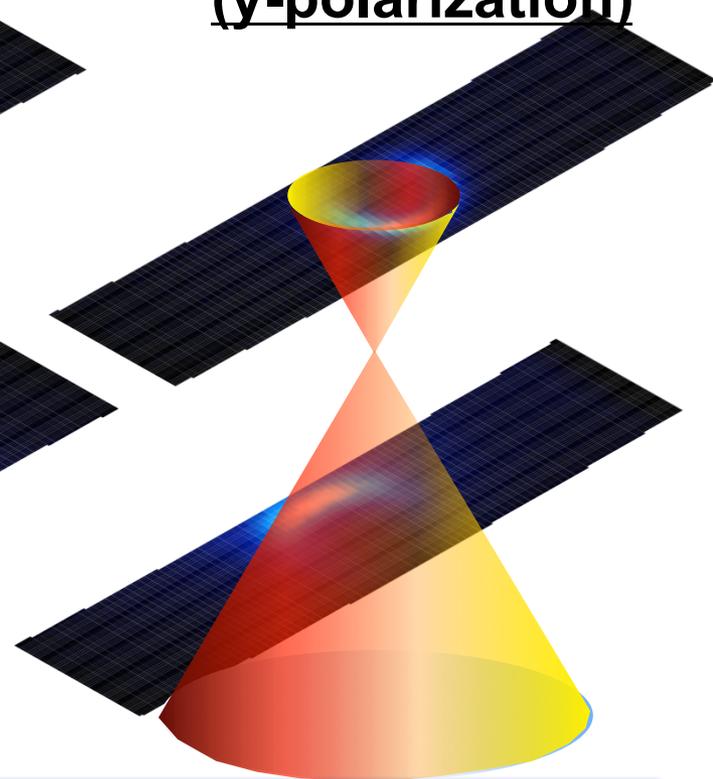
Expected



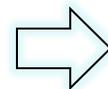
Measured  
(x-polarization)



Measured  
(y-polarization)

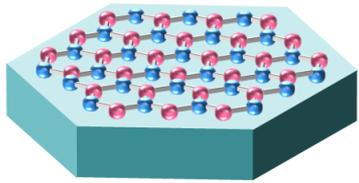


$\pi/n$  rotation of ARPES intensity

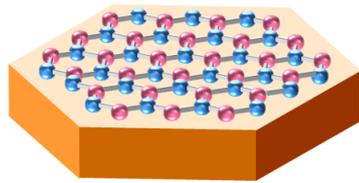


$n\pi$  Berry's phase

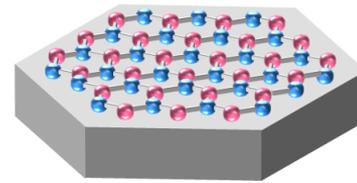
# Graphene on a dielectric substrate



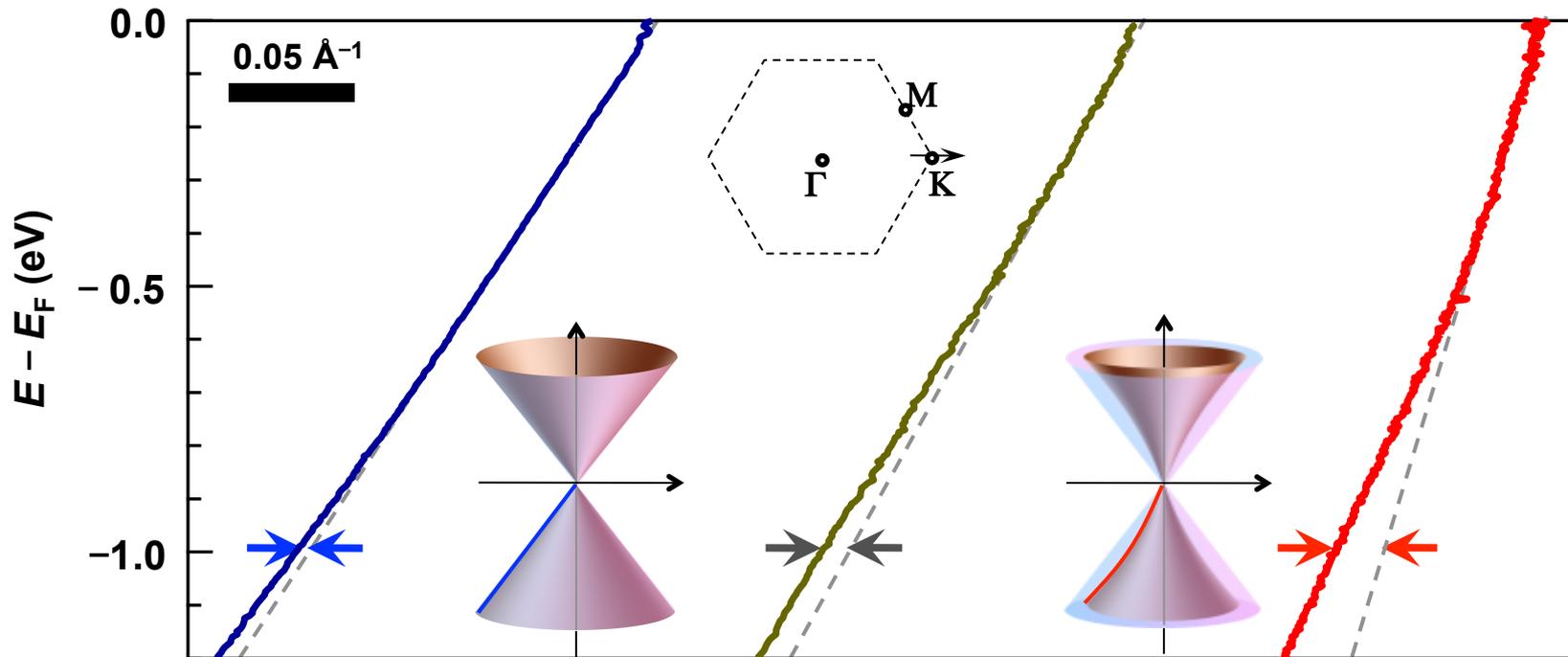
SiC(000 $\bar{1}$ )



h-BN



Quartz



momentum

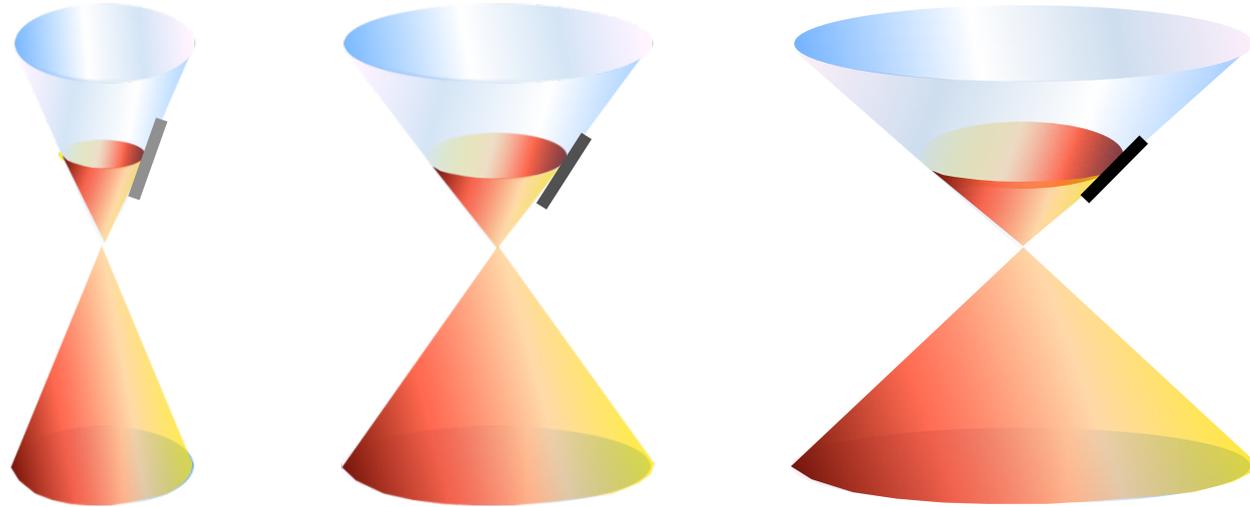
Hwang, Siegel (2011, 2012)

## 1. Departure from linearity

# Strong electron-electron interactions in graphene

Fermi liquid

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



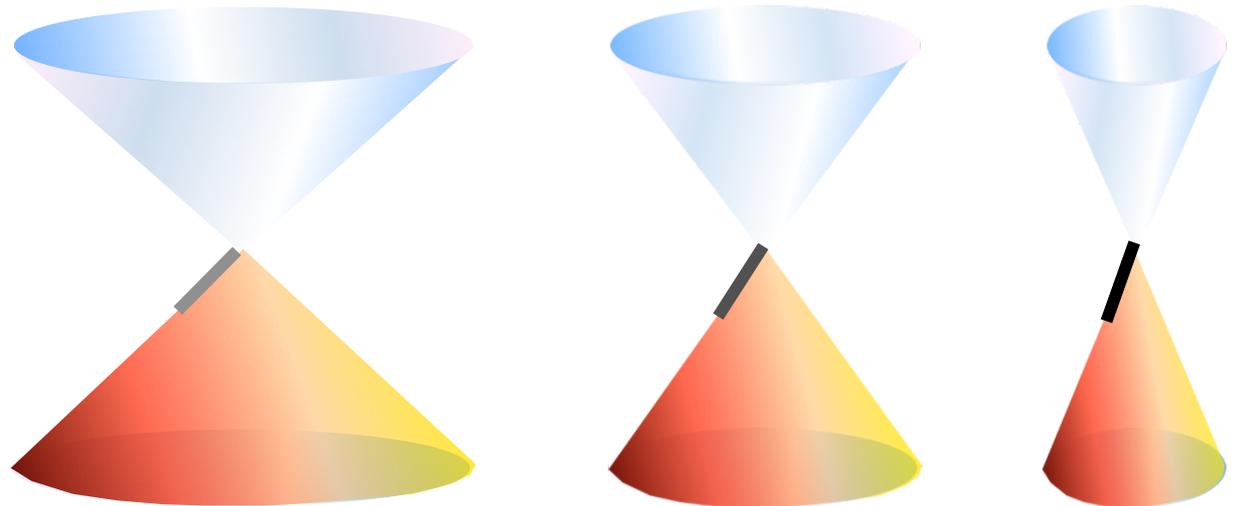
Increasing electron-electron interactions



Charge neutral graphene

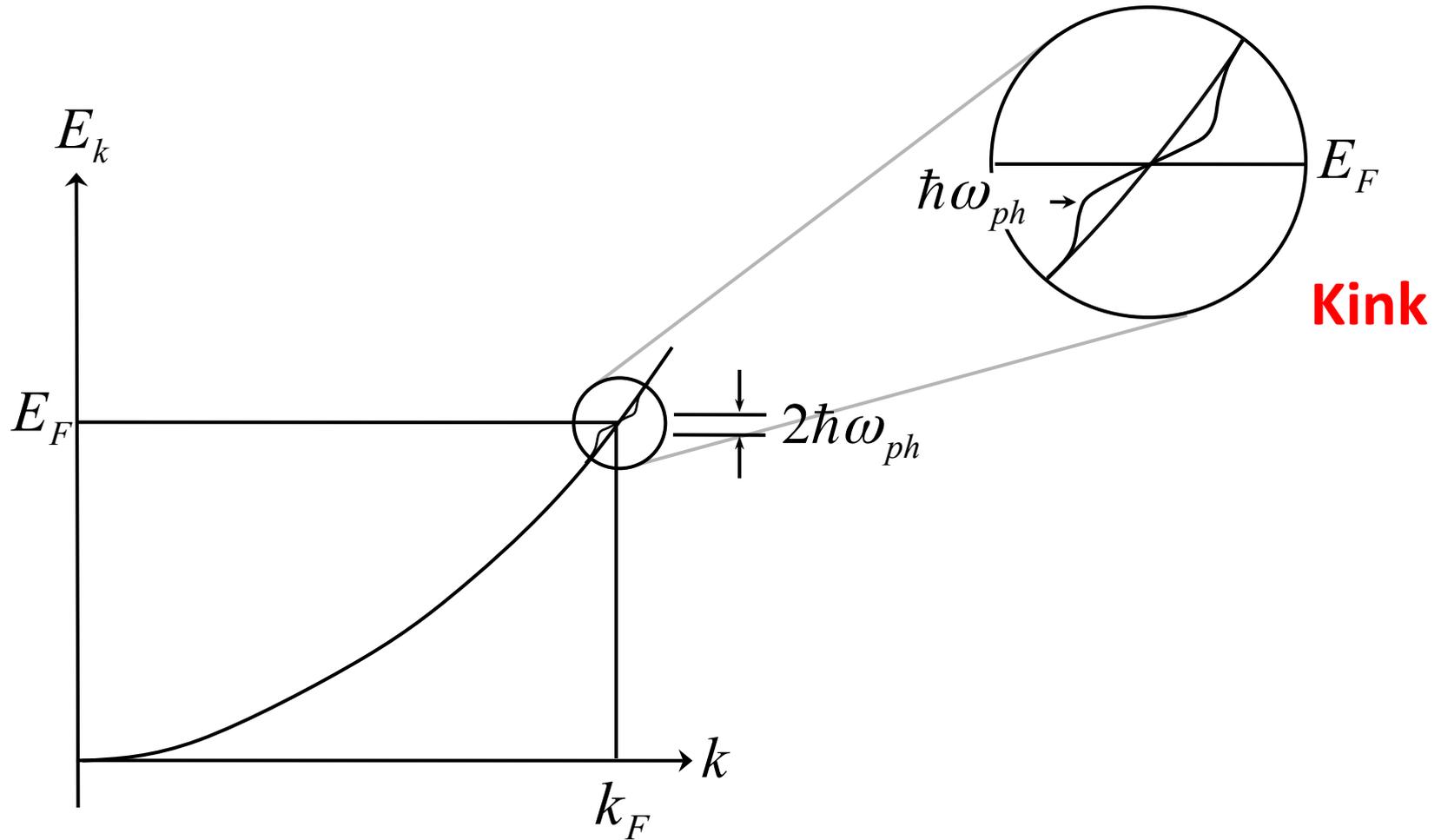
$$E = \hbar v_F^* k$$

**Non-Fermi  
liquid**

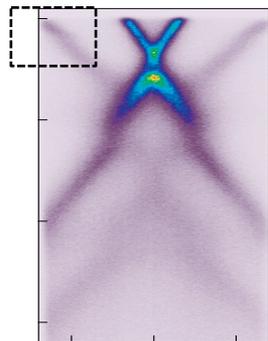


# Electron-phonon coupling

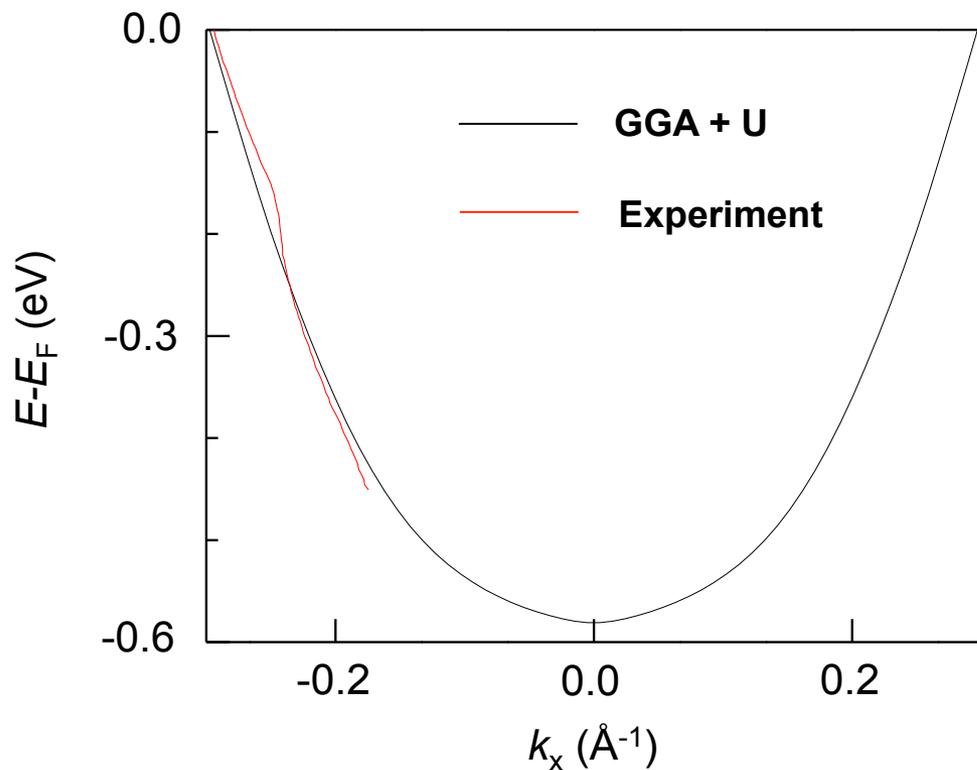
## Electron-phonon coupling



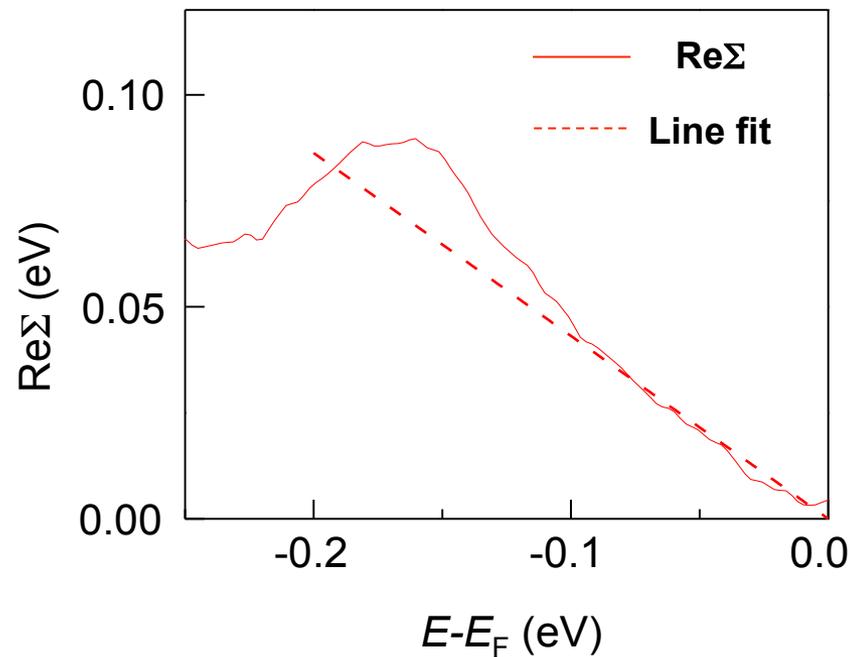
# Electron-phonon coupling in Yb/Graphene



Graphene + Yb



Self-energy by electron-phonon coupling



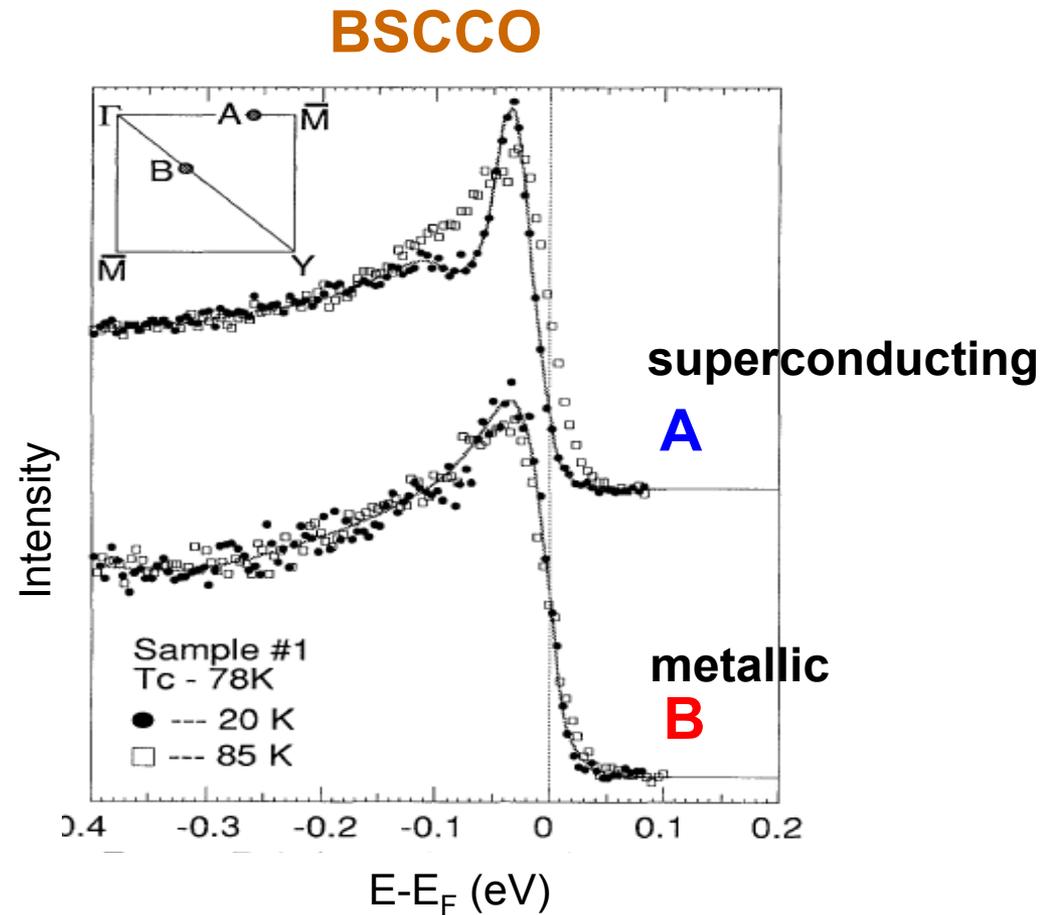
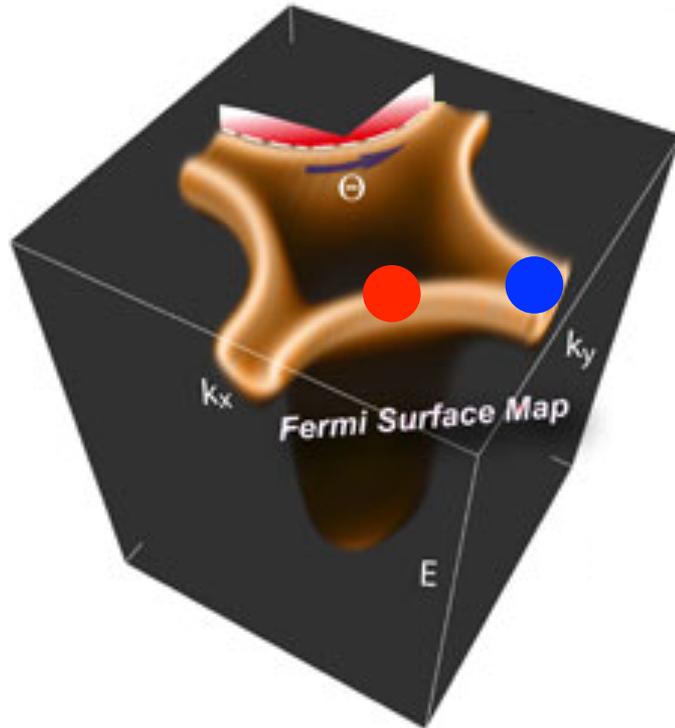
$$\lambda = \frac{d \operatorname{Re} \Sigma(E)}{d E}$$

$\lambda = 0.02 \sim 0.05$   
 $\lambda = 0.43 \sim 0.51$   
 $0.23 < \lambda < 1.12$

Hwang (2014)

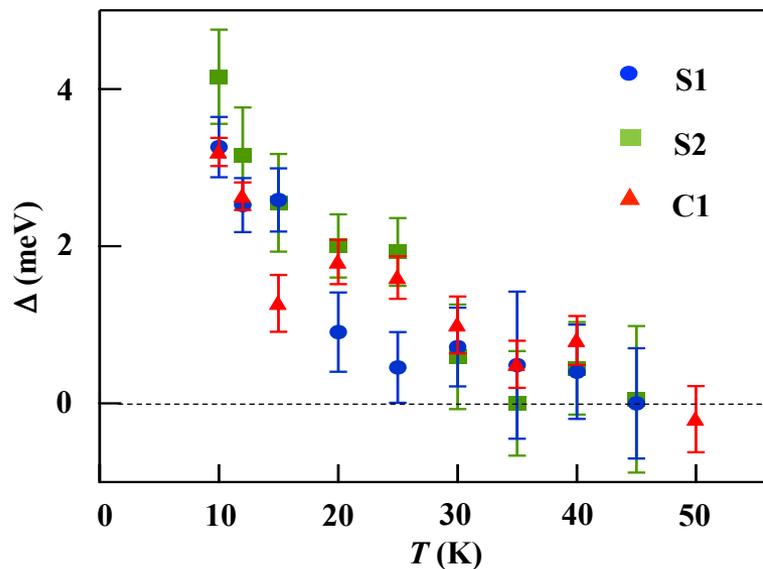
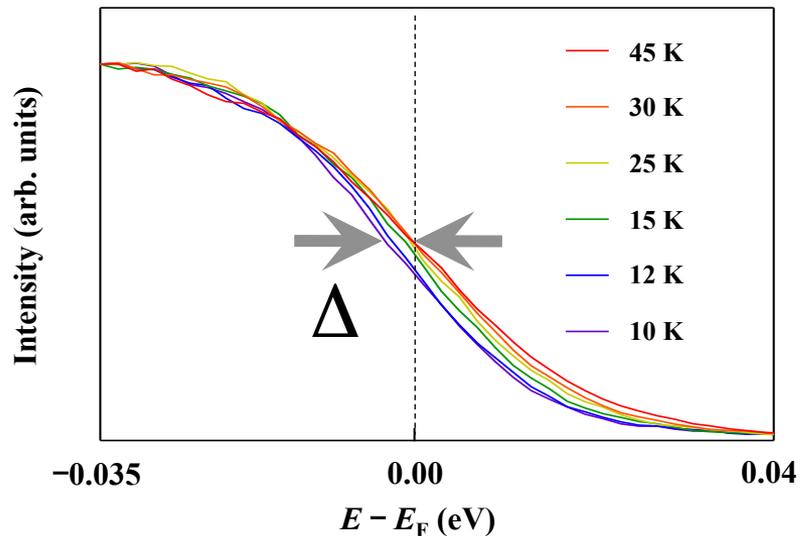
(Conventional superconductors)

# Anisotropic superconductivity

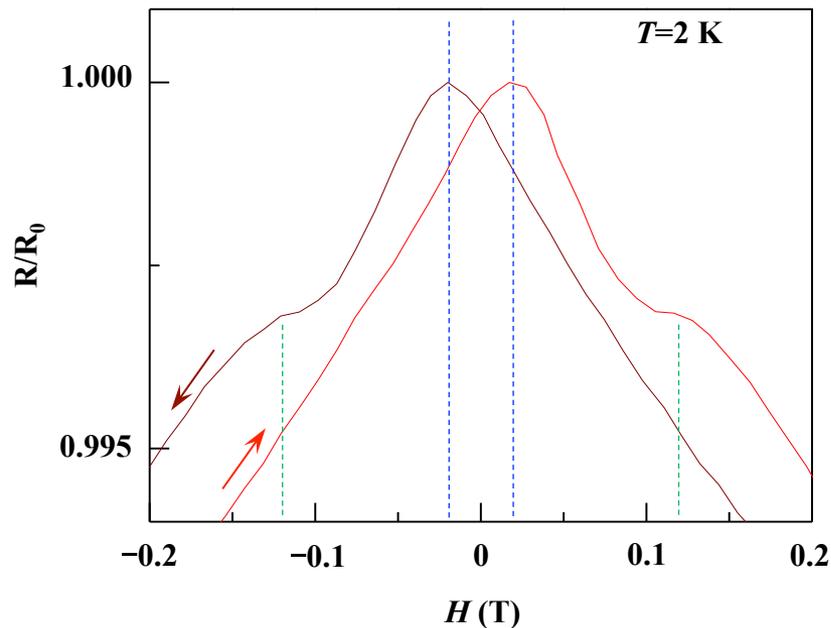


# Magnetism in S/Graphene

## Graphene + S



## Transport measurements



**Magnetic effects**  
directly tied to the  
**graphene band structure**

# Outline

## 1. What are the grand challenges

## 2. Angle-resolved photoemission spectroscopy (ARPES)

- From Einstein's photoemission to Present
- Conventional ARPES

## 3. Future directions of ARPES

- Time resolution
- Spin and time resolution

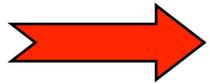
# The future of photoemission

What is missing? **Time** and **Spin**



**Time resolved photoemission**

*To study excitation and phase transitions*

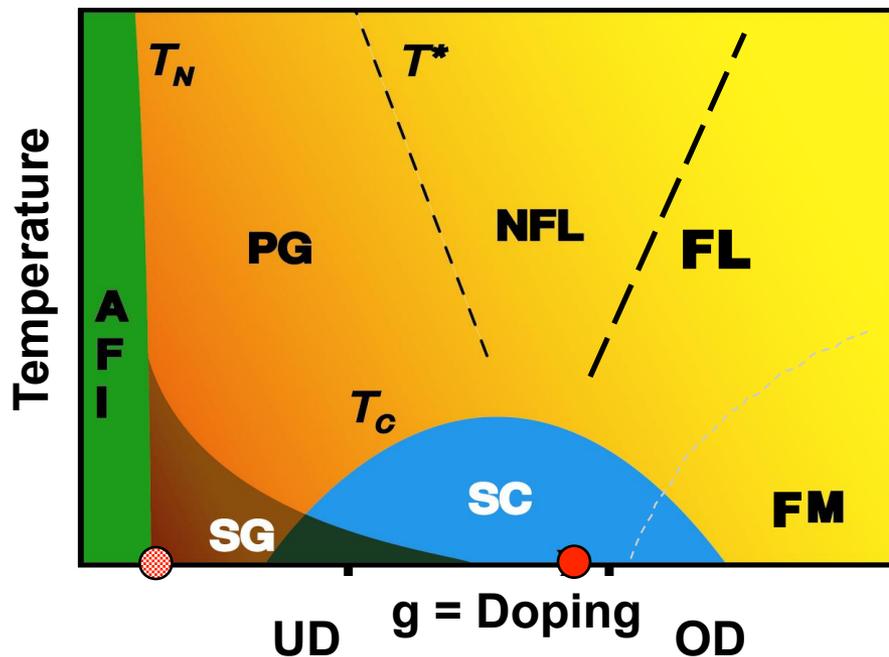


**Spin resolved photoemission**

*To access spin degrees of freedom*

# The mystery of high $T_C$ superconductivity

- Why do we need these additional resolutions?



1) Is there a Quantum Critical Point?

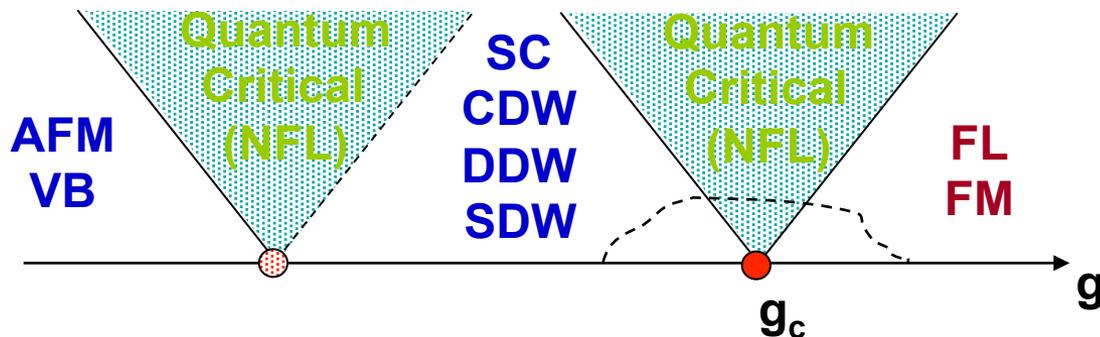
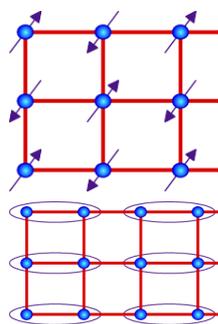
2) What is the nature of NFL region?

3) What defines the Pseudogap (PG) phase?

4) What are the basic excitations?

5) Is SC a consequence of QCP or is it hiding the QCP?

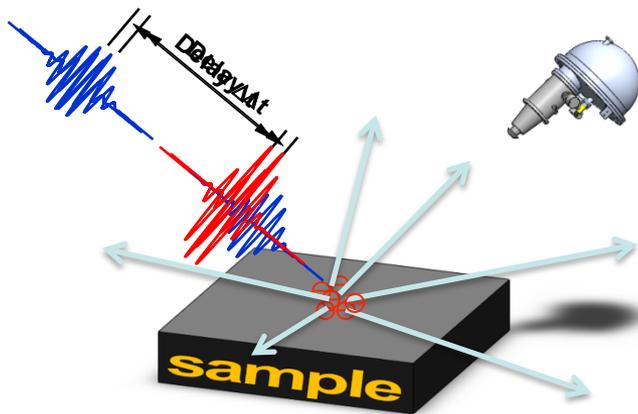
6) Are PG and SC state competing states of matter?



We need **time** and **spin** resolutions

# Time-resolved ARPES

## Pump-Probe, ultrafast ARPES:



### Possibilities:

Quasiparticle lifetimes

Ultrafast phase transitions

Closing/opening of gaps

Coherent oscillations

⋮

**Energy conservation**

$$E_B = \hbar\omega - E_K - \Phi_A$$

**Angle / momentum conservation**

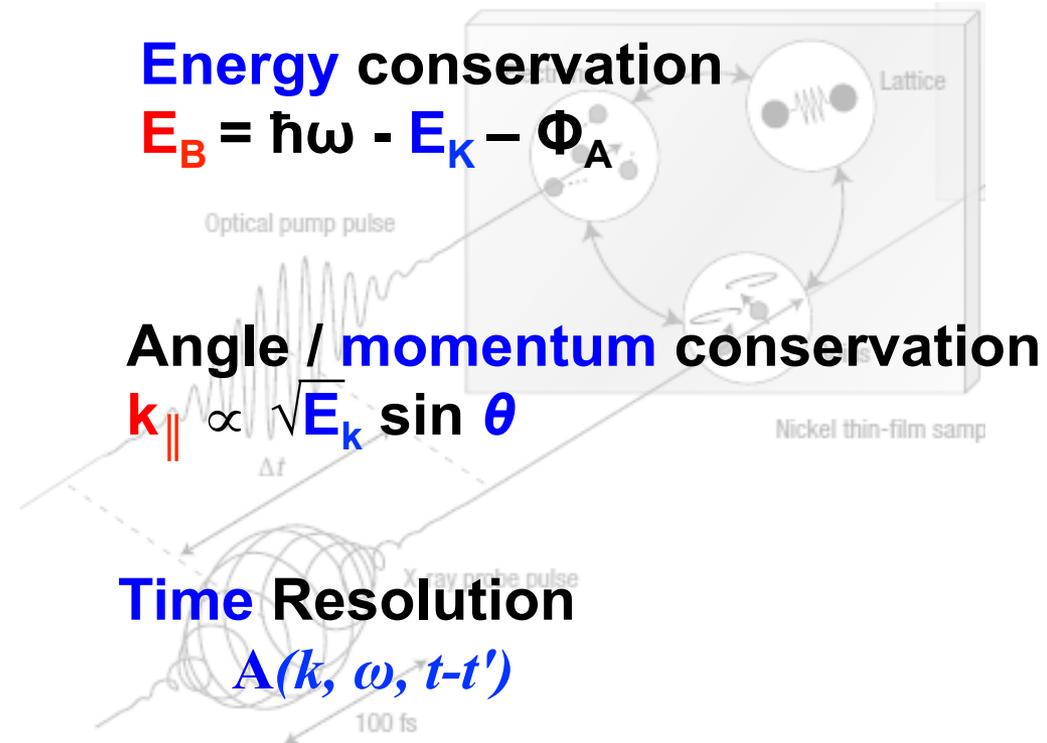
$$k_{\parallel} \propto \sqrt{E_K} \sin \theta$$

**Time Resolution**

$$A(k, \omega, t-t')$$

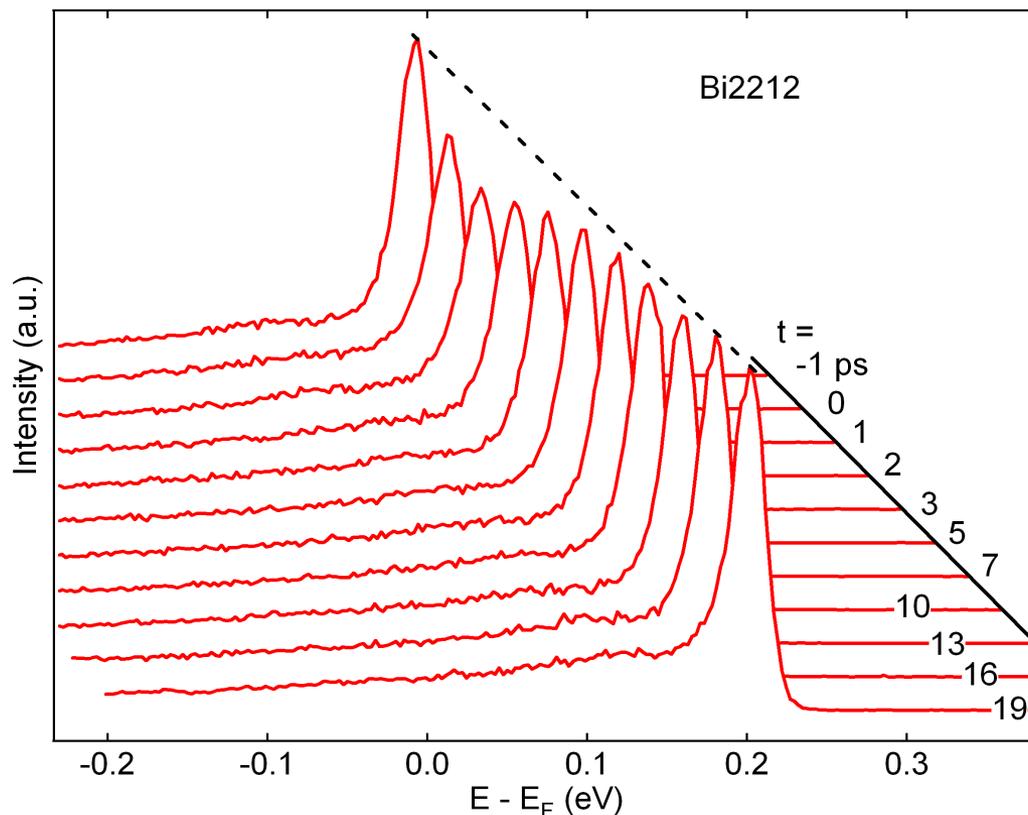
**Photoelectron intensity**

$$I(k, \omega) \propto |A(k, \omega, t-t')|^2 |M(k, \omega)|^2 f(\omega)$$



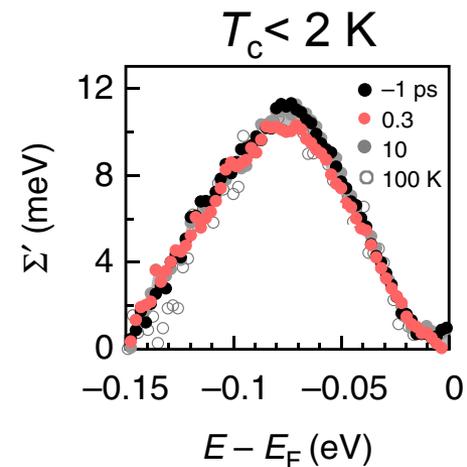
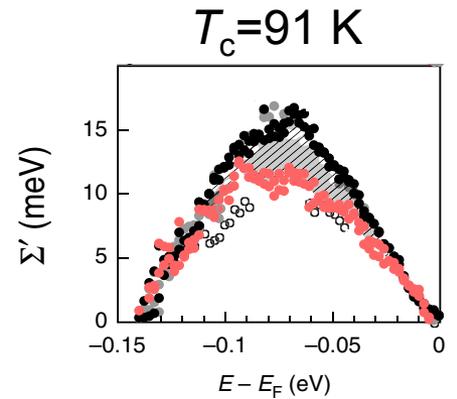
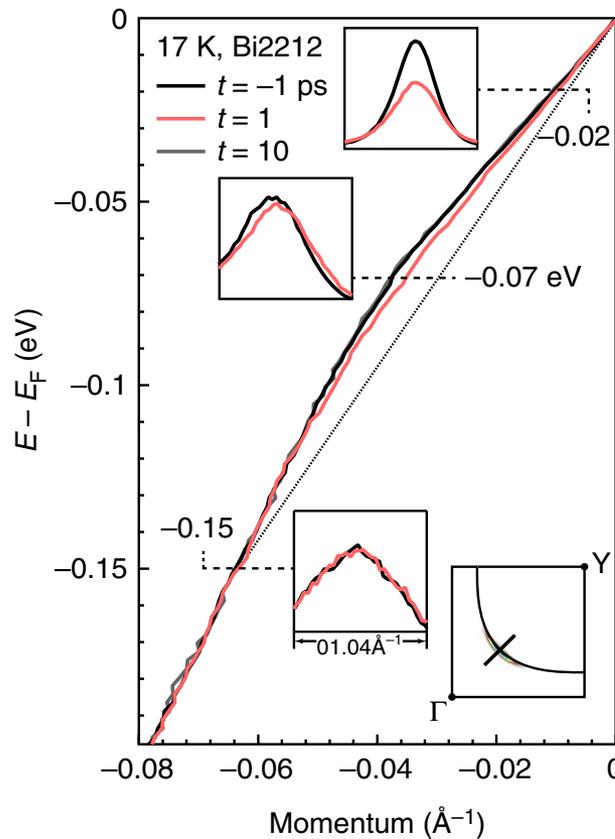
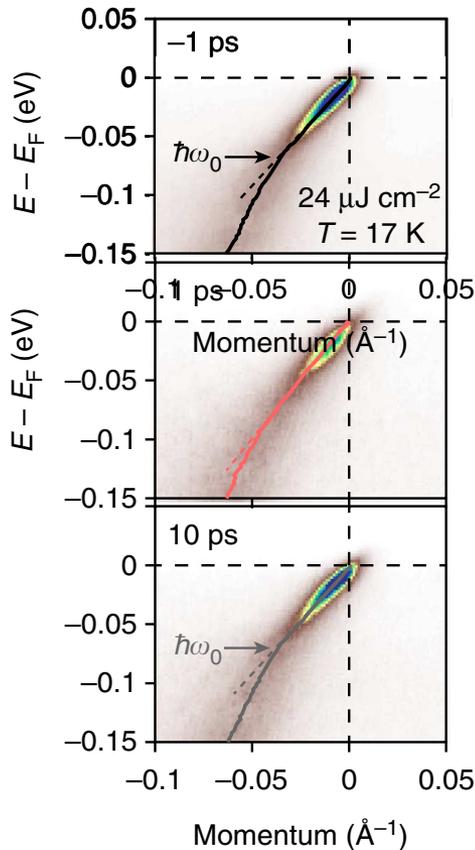
# What do we measure with time-resolved ARPES?

Directly measure electronic response / recovery dynamics  
- (non-equilibrium)



# The power of time + momentum resolution

## High- $T_c$ superconductors



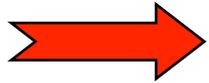
# The future of photoemission

What is missing? **Time** and **Spin**



**Time resolved photoemission**

*To study excitation and phase transitions*

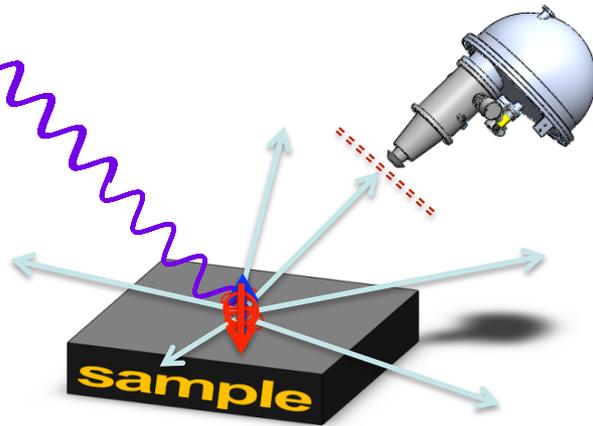


**Spin resolved photoemission**

*To access spin degrees of freedom*

# Spin + time-resolved ARPES

## Spin-ARPES



### Critical for:

- Fundamental magnetism
- Magnetic phase transitions
- Topological Insulators
- Colossal magnetoresistance
- FeAs superconductors
- ⋮

Energy conservation

$$E_B = \hbar\omega - E_K - \Phi_A$$

Angle / momentum conservation

$$k_{\parallel} \propto \sqrt{E_K} \sin \theta$$

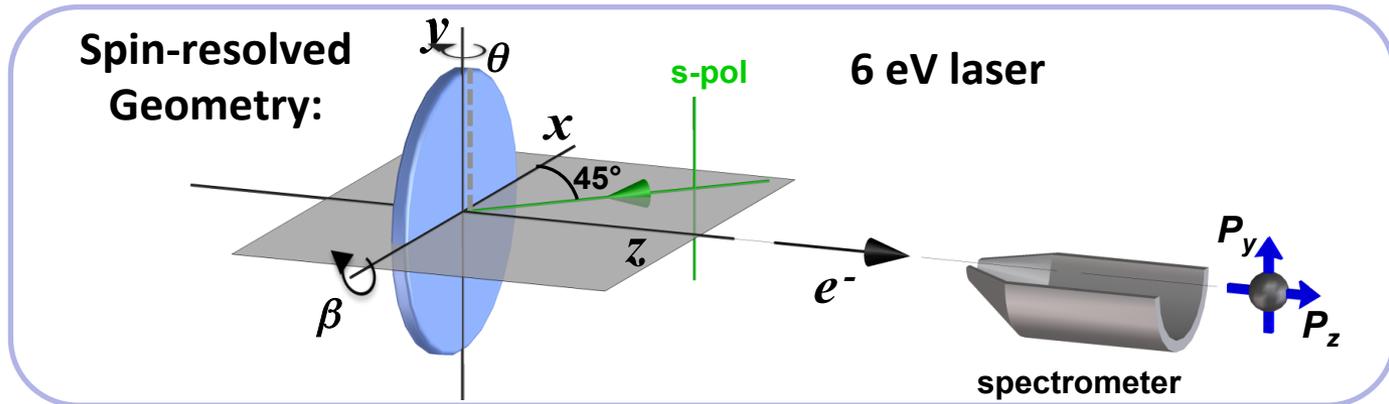
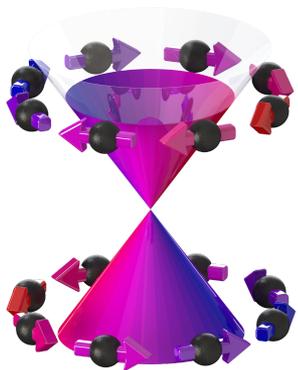
Time resolution

$$A(k, \omega, t-t')$$

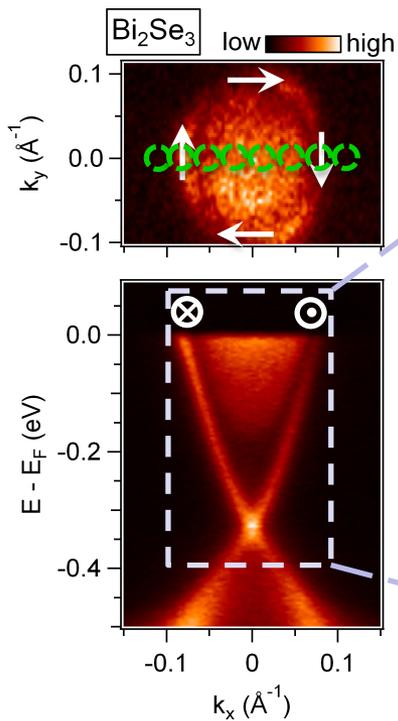
Spin resolution

$$P_x, P_y, P_z$$

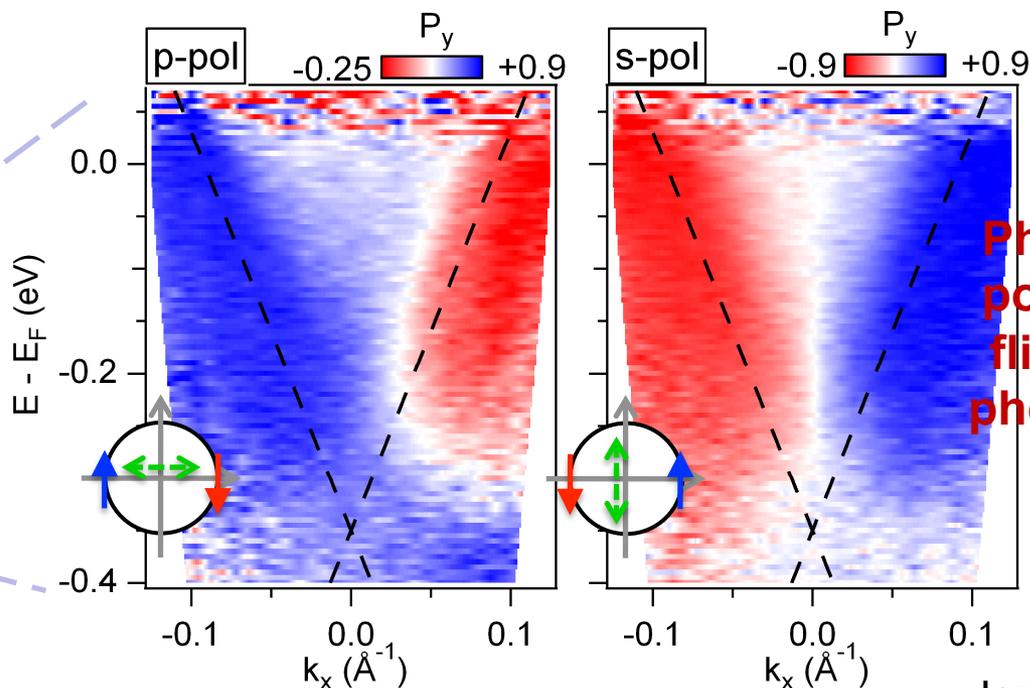
# Light-spin interactions in topological insulator



**Standard ARPES:**



**Full Spin Polarization Maps:**



**Photoelectron spin polarization is fully flipped by rotating photon polarization!**

# What have we learned?

## ARPES

A **powerful** tool to **understand electronic properties** of a material

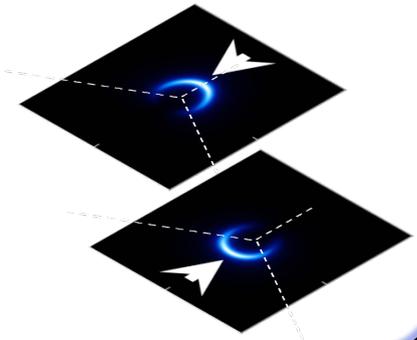
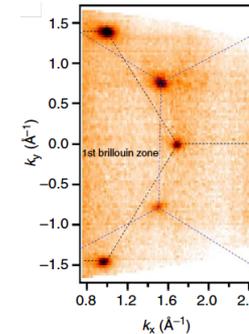
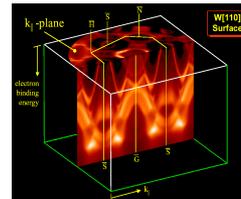
## Electron band structure

Fermi surface

Geometric structure

Quantum mechanical phases

Many-body effects



## Future of ARPES

**Time-resolved ARPES**

: *study excitation and phase transitions*

**Spin-ARPES**

: *fundamentals of magnetism*

