Bandstructure and Spectral Function of Single and Bilayer Graphene Measured by ARPES

Eli Rotenberg
Dr. Aaron Bostwick, Dr. Taisuke Ohta
Advanced Light Source
Lawrence Berkeley National Laboratory USA

Prof. Th. Seyller - Erlangen
Prof. K. Horn - FHI Berlin

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Outline

- Experimental technique
  - Angle-Resolved Photoemission Spectroscopy (ARPES)
  - Sample Preparation
- Bandstructure Determination of Graphene from 1 to 2 layers
  - new! 1 layer: Bostwick et al cond-mat/0609660.
  - bilayer: Ohta et al Science
- Spectral Function of 1-layer graphene
  - The lifetime of holes in *n*-doped graphene is determined by
    - electron-phonon coupling
    - electron-electron coupling
      - e-h pair generation
      - e-plasmon coupling
- Future Work
  - towards ARPES at 50 nm spatial resolution

Experimental

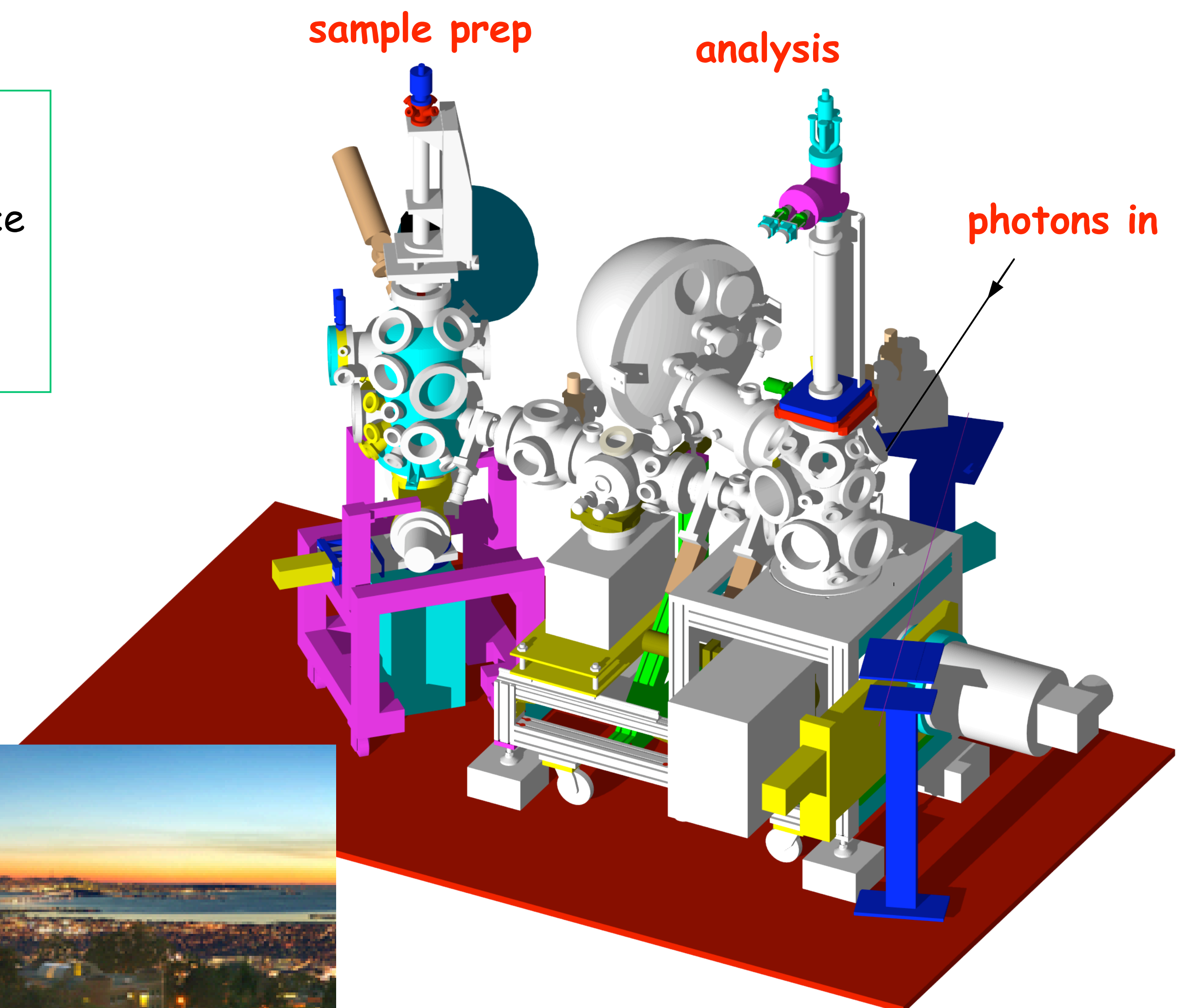
- **Substrate**
  - *n*-type (N) 6H-SiC(0001)
  - N=1.5±0.5 x 10^{18} cm^{-3}
- **Preclean**
  - anneal in hydrogen plasma
- **Graphetization**
  - anneal in ultra-high vacuum 1150C: longer = thicker [1,2]
  - P<1x10^{-10} T
- **Doping**
  - *n*-doping up to 6x10^{13} cm^{-2} by K deposition up to 0.04 ML
- **Measure**
  - P~2x10^{-11} T
  - T~20K

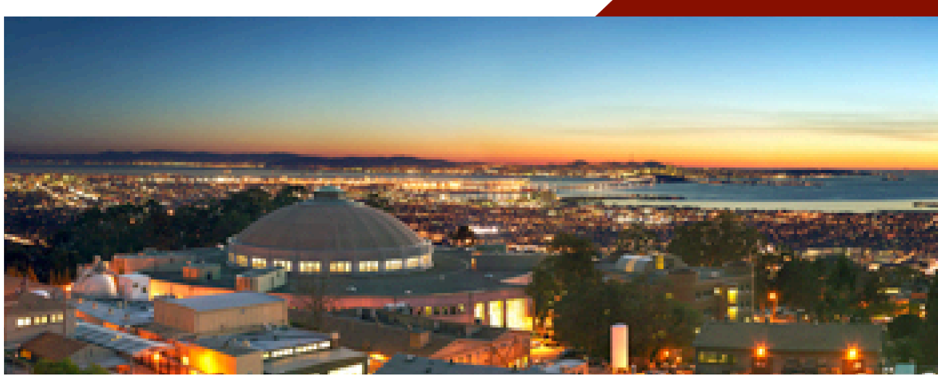
[1] Forbeaux, I., J.M. Themlin, and J.M. Debever. Phys. Rev. B, 1998. **58**(24): p. 16396-406
[2] Berger, C., et al., Science, 2006. 312: p. 1191-6.

ESF - the Electronic Structure Factory

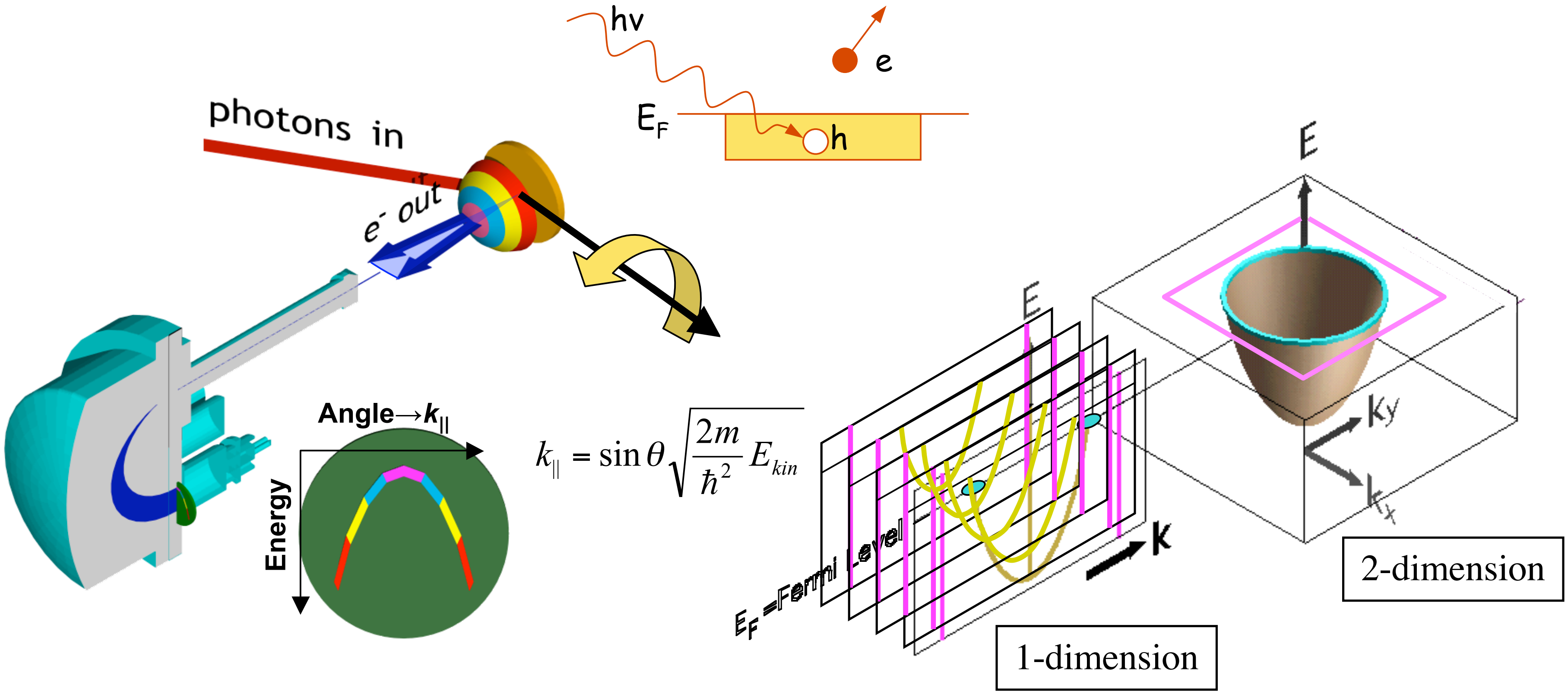
An international
user facility at the
Advanced Light Source

Lawrence Berkeley
Natl. Laboratory





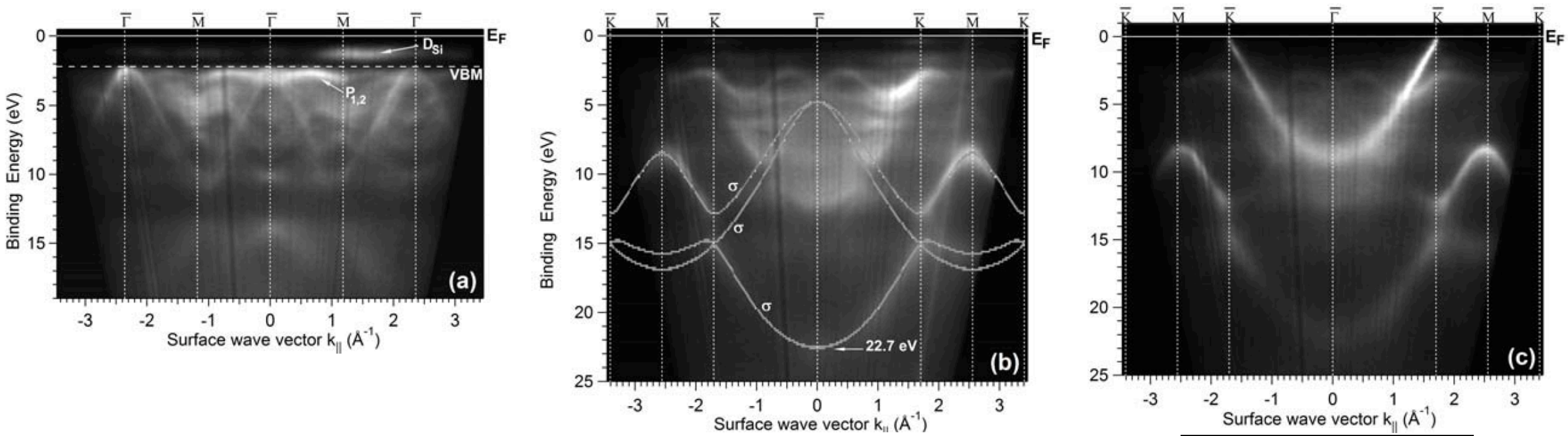
Measurement of Electronic Band Structure Using ARPES

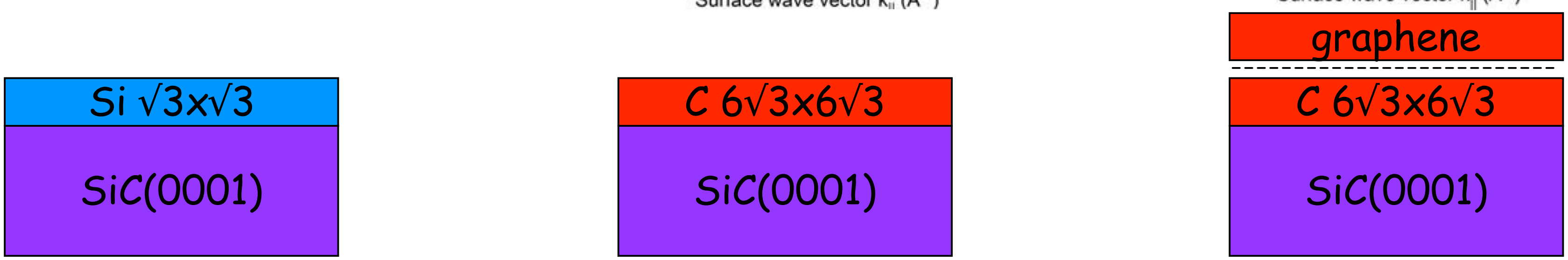


- Photon source: Beamline 7.01, ALS
  - *hv*=95eV, Energy resolution 25-30meV
- Electron analyzer: Scienta R4000
  - Angular resolution 0.1° (0.01Å^{-1})

Formation of first graphene layer

K.V. Emtsev, Th. Seyller, F. Speck, L. Ley, P. Stojanov, J.D. Riley, R.G.C. Leckey, cond-mat 0609383



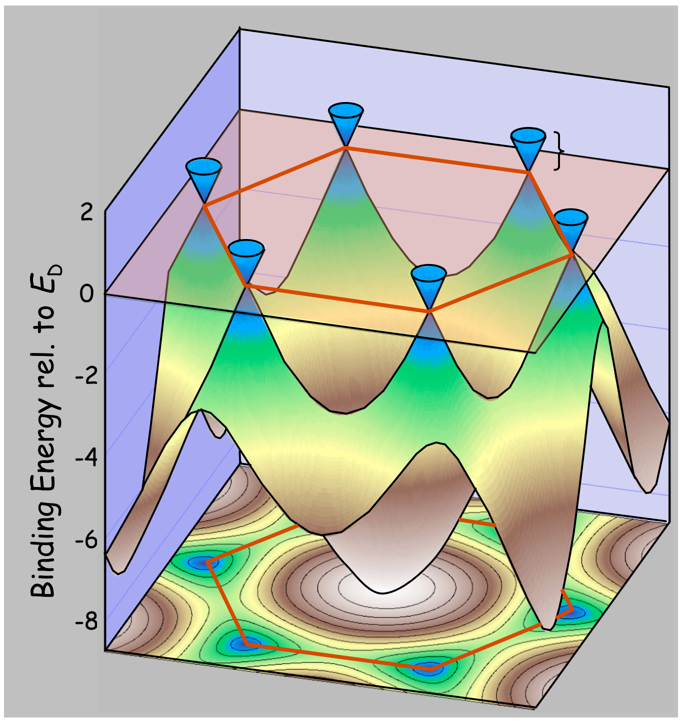


*sp*²-bonded
p_z hybr. with SiC

*sp*²-bonded
p_z derived band
Van Der Waals
Bonding

graphene bandstructure

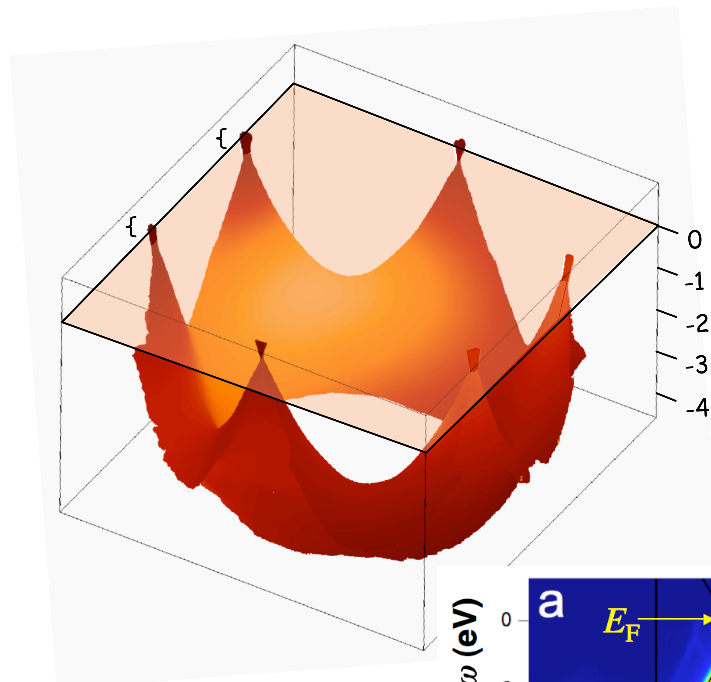
tight-binding model

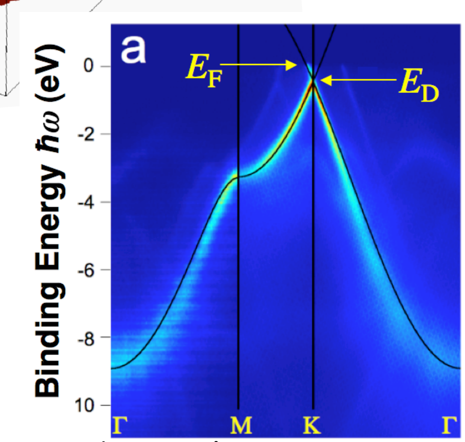


R. Saito, G. Dresselhaus, M. S. Dresselhaus Physical properties of carbon nanotubes, Imperial College Press, 1998

t = 2.82 eV

expt





$E_F = E_D + 0.435$ eV

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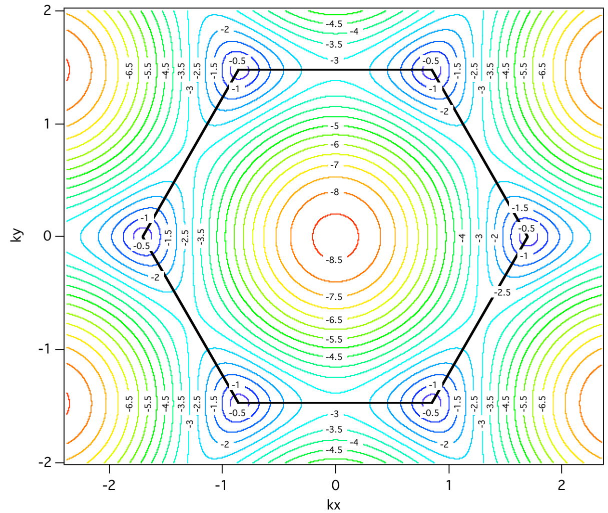
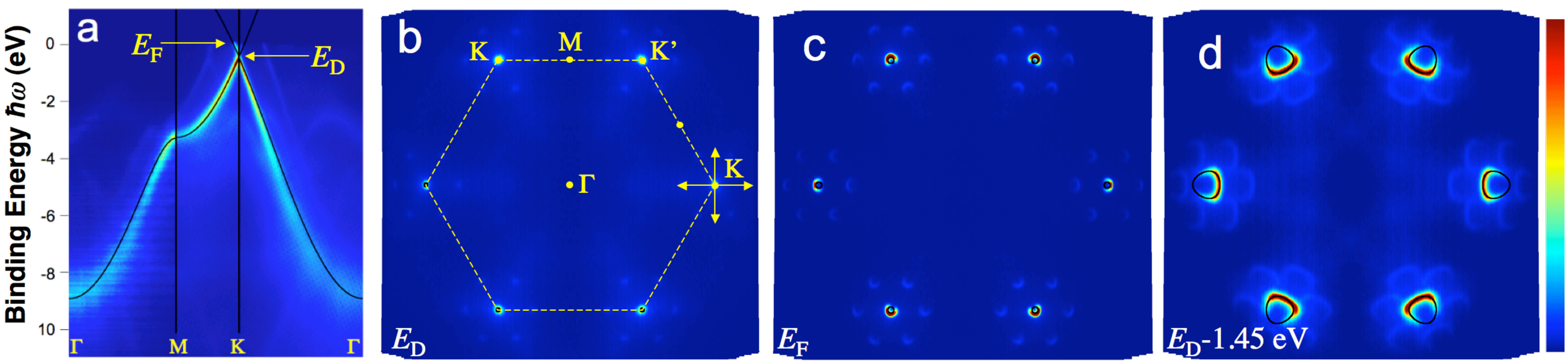
Graphene: TB vs Expt. Data



t = 2.82 eV

Momentum resolution
0.012Å^{-1} = 0.7% of ΓK



- expt. bands generally more anisotropic than model
- spectral peak widths are limited by sample lifetime

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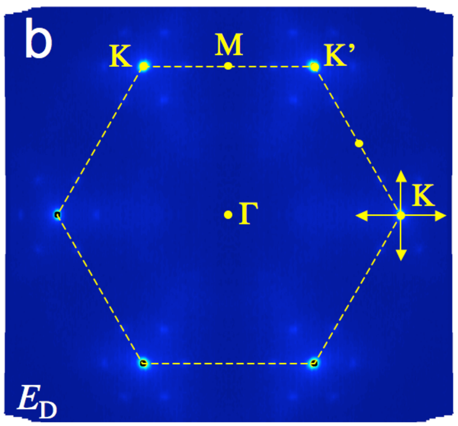
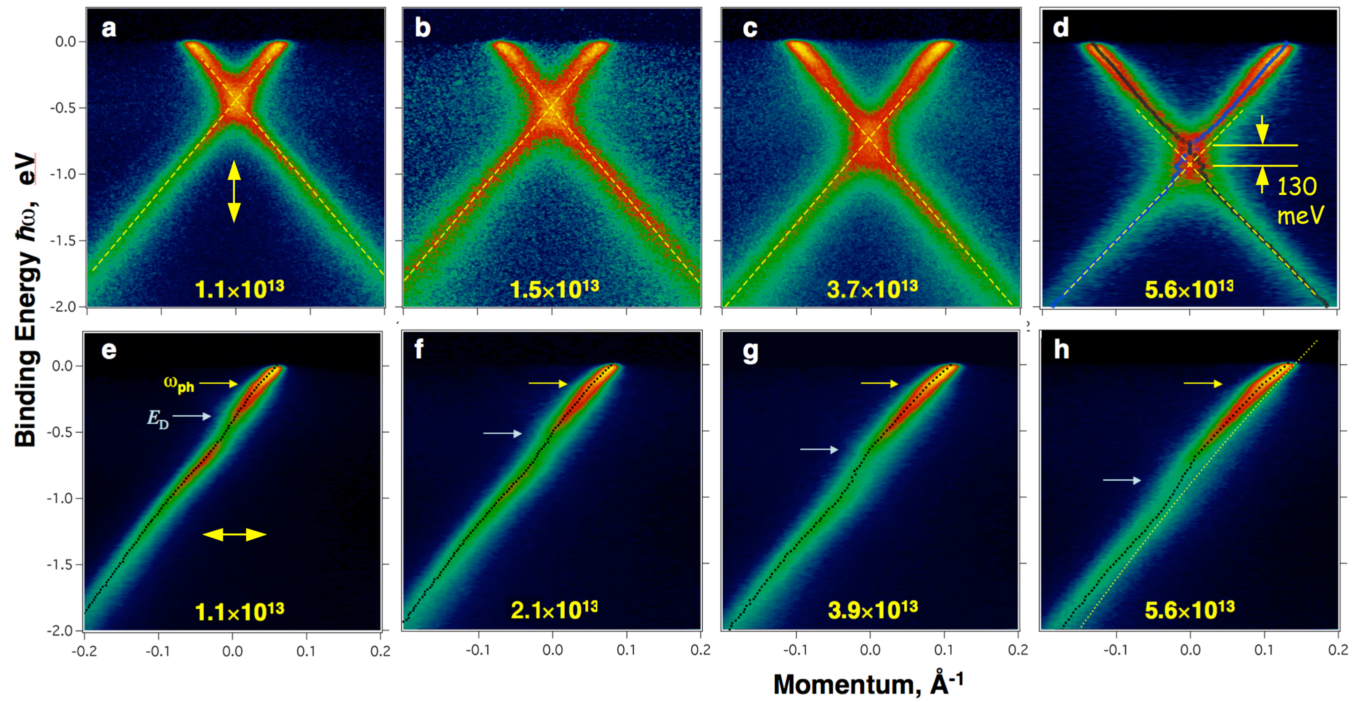
Spectral Function of Graphene



As grown

Increased doping by K deposition ⟶



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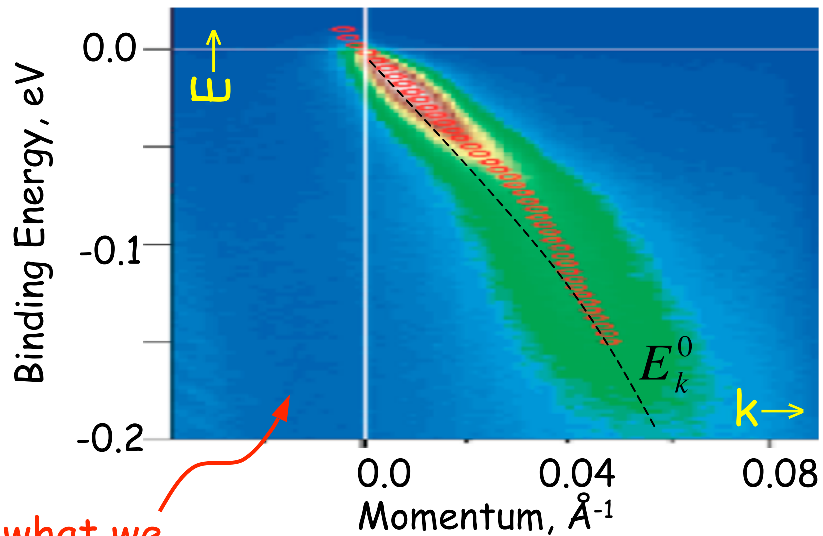
Many body effects and photoemission

"Kinkology"

BSCCO Superconductor Results [1]

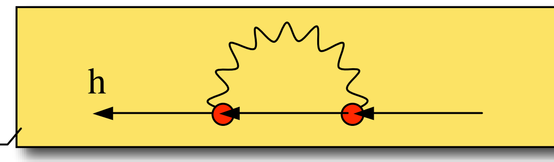


what we measure

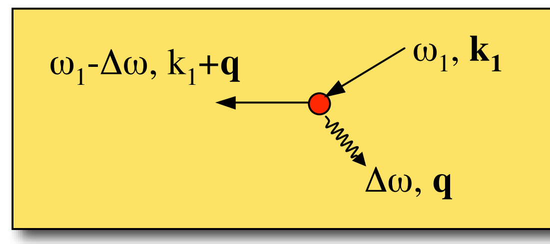
$$\overbrace{A(k,E)}^{\text{what we measure}} = \frac{1}{\pi}\frac{\mathrm{Im}\,\Sigma(k,E)}{\left[E - E_k^0 - \mathrm{Re}\,\Sigma(k,E)\right]^2 + \left[\mathrm{Im}\,\Sigma(k,E)\right]^2}$$

$$\Sigma(k,E) = \underbrace{\mathrm{Re}\,\Sigma(k,E)}_{\text{energy shift}} + i\underbrace{\mathrm{Im}\,\Sigma(k,E)}_{\text{lifetime}}$$

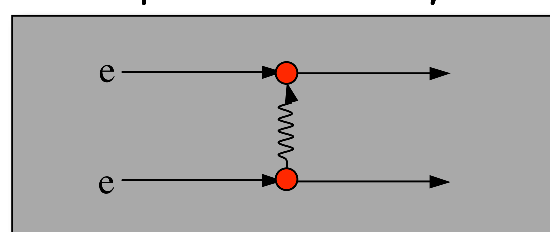
mass renormalization



quasiparticle decay

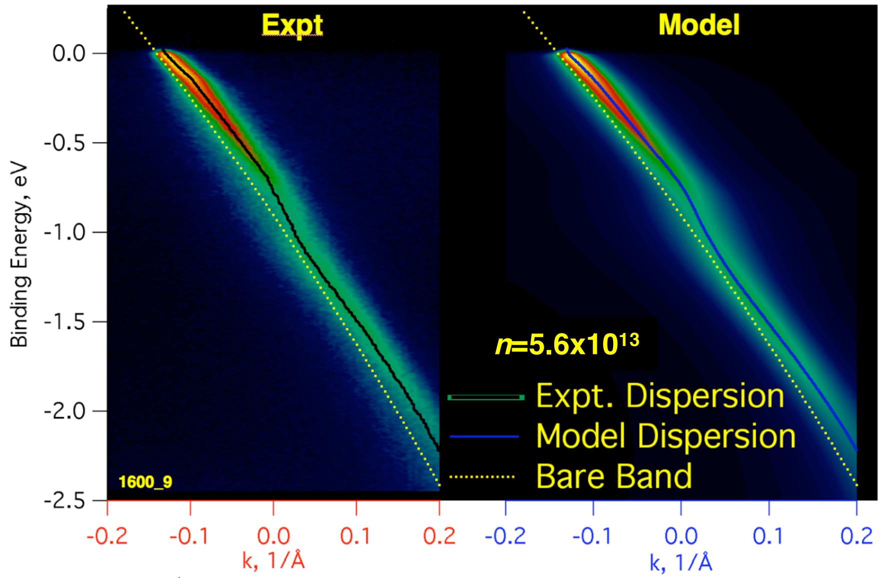


superconductivity



[1] Koralek et al, Phys. Rev. Lett. 96, 017005 (2006)

Is the Quasiparticle picture valid for graphene?



what we measure

Calculated A(k,E) Using only ImΣ (expt) and ReΣ (calc)

$$\overbrace{A(k,E)} = \frac{1}{\pi}\frac{\mathrm{Im}\Sigma(k,E)}{\left[E - E_k^0 - \mathrm{Re}\Sigma(k,E)\right]^2 + \left[\mathrm{Im}\Sigma(k,E)\right]^2}$$

$$\Sigma(k,E) = \underbrace{\mathrm{Re}\Sigma(k,E)}_{\text{energy shift}} + i\underbrace{\mathrm{Im}\Sigma(k,E)}_{\text{lifetime}}$$

Kinks are due to many-body interactions, not details of the single-particle bandstructure

(i.e. not a consequence of strain, coupling to substrate, etc)

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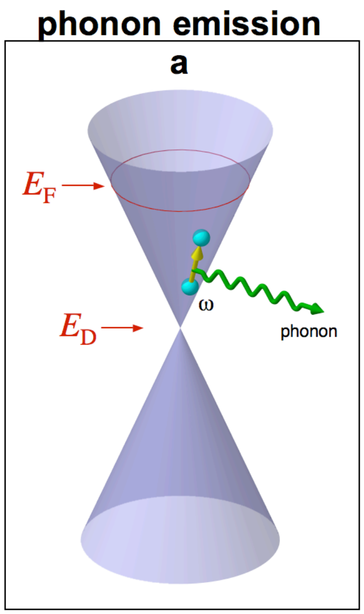
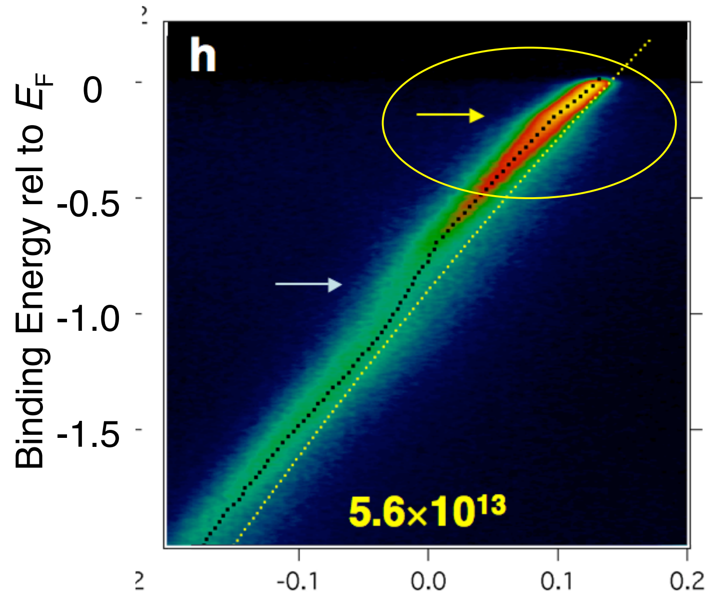
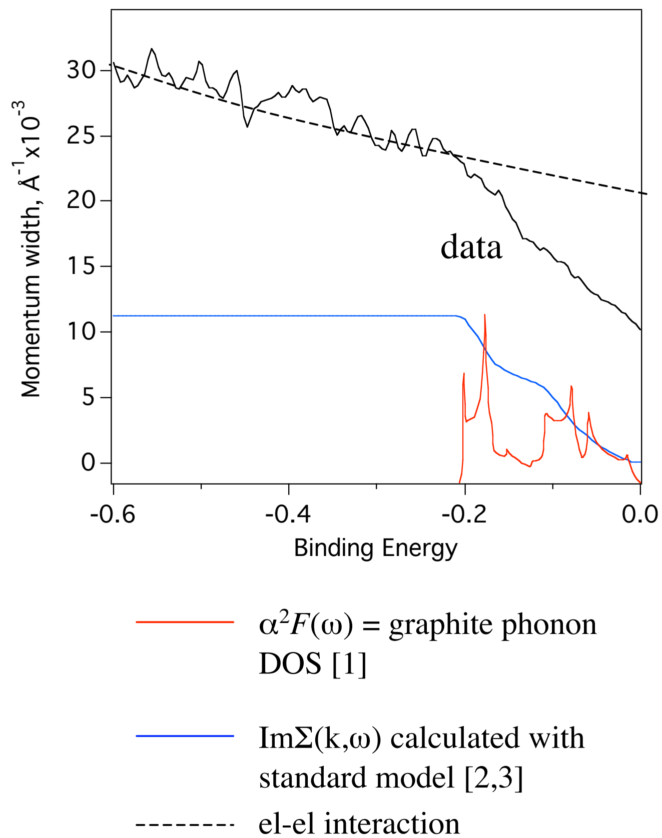
Electron Phonon Coupling







λ=0.3

[1] Vitali, L., et al., *Phonon and plasmon excitation in inelastic electron tunneling spectroscopy of graphite.* Phys. Rev. B, 2004. 69(12): p. 121414.
[2] Grimvall, G., The Electron-Phonon Interaction in Metals. 1981, Amsterdam: North Holland Publishing Company.
[3] Also seen for graphite, Zhou et al.Annals of Physics 2006

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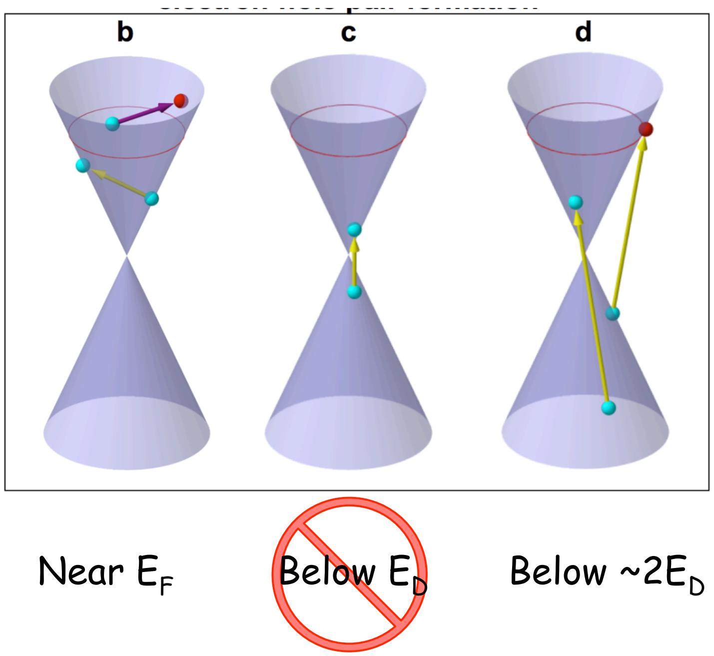
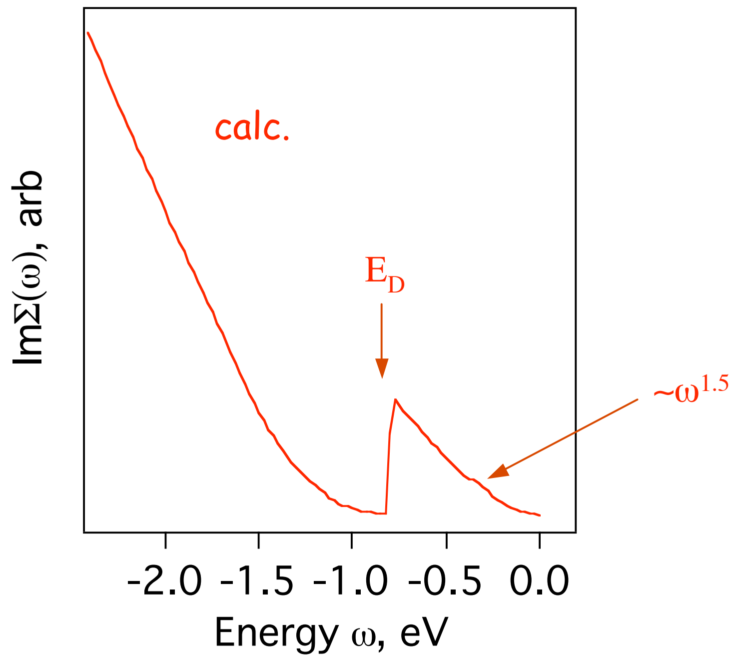
Electron-Electron Coupling

Fermi-liquid-like decay by electron-hole pair formation





Departure from the usual <u>monotonic</u> Fermi liquid behavior (~ω^2)

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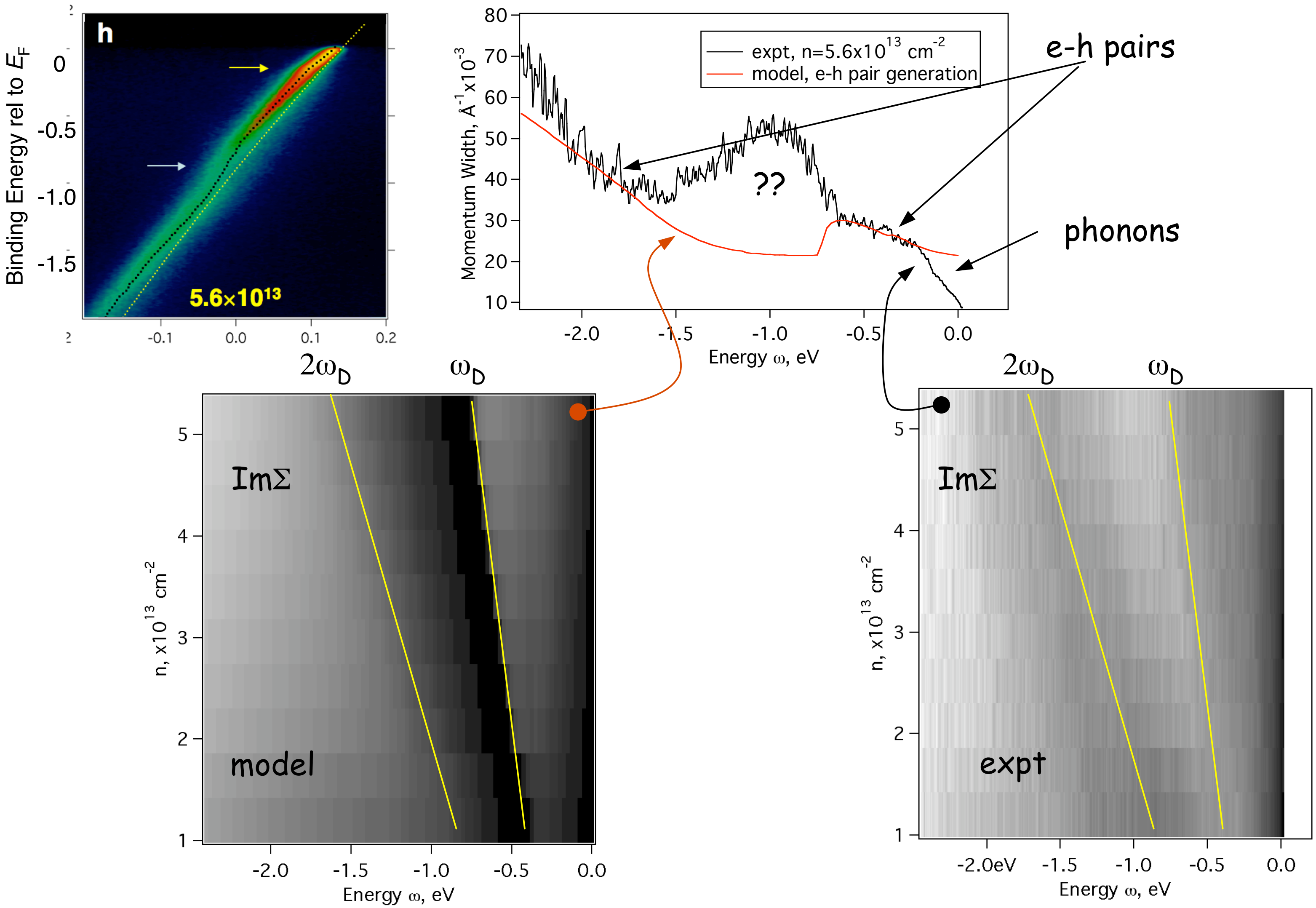
Im Σ due to *e-h* pair generation



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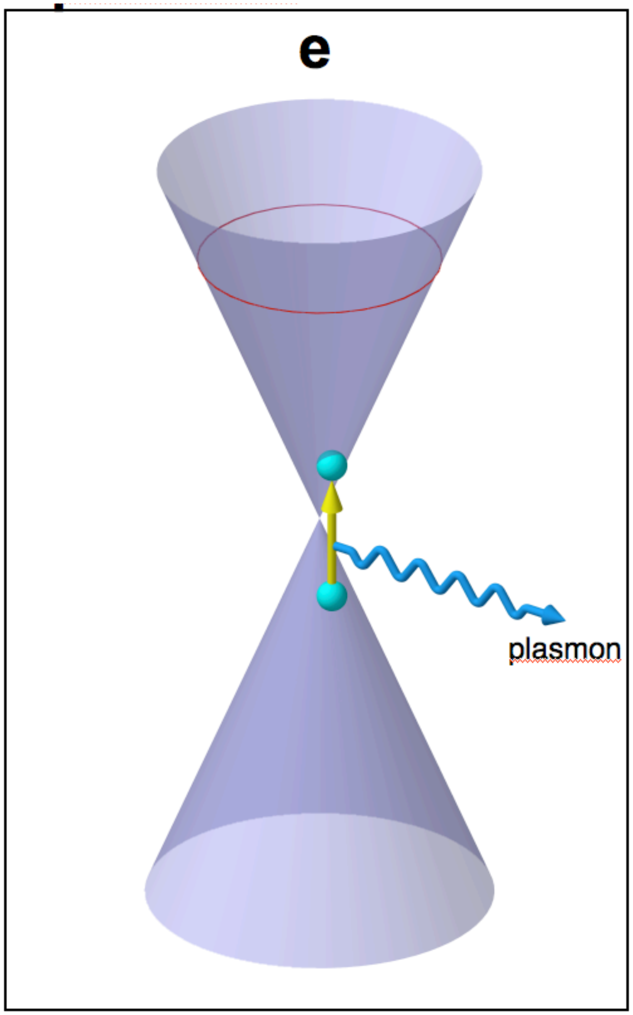
Plasmon model



we need to couple to a mode
with large ω and small q

--> plasmons

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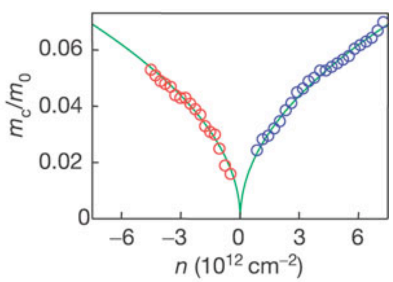
A little bit more about the plasmon spectrum

Ordinary 2-dimensional dispersion function

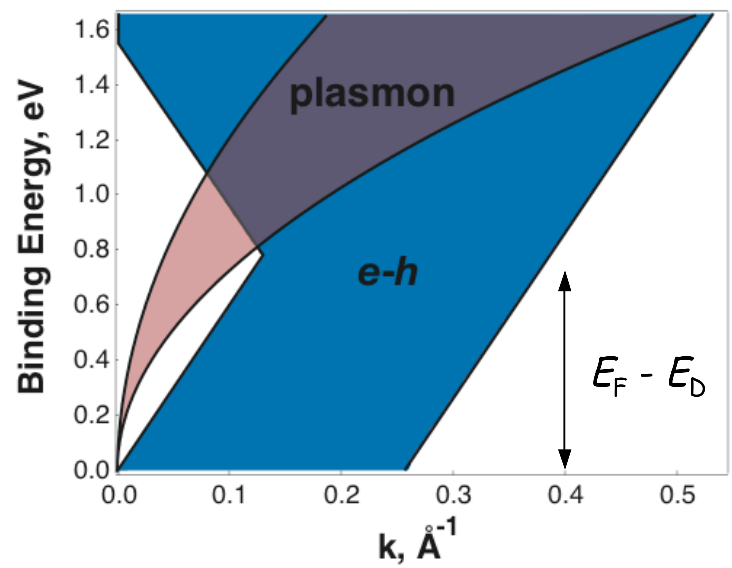
$$\omega_{pl}(q) = \sqrt{4\pi n e^2 q / m_c (1+\varepsilon)}$$

Carrier mass ~0.1 m_e extrapolated from the transport msmts. of Novoselov *et al*

Screening constant. We use from 3-10



Novoselov, K. S. et al. Two-dimensional gas of massless Dirac fermions in graphene. Nature 438, 192-200 (2005).



We expect a peak in ImΣ which scales in width and size with (E_F - E_D)

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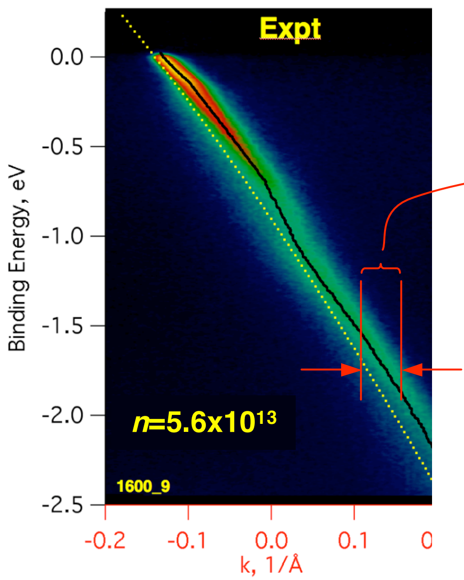
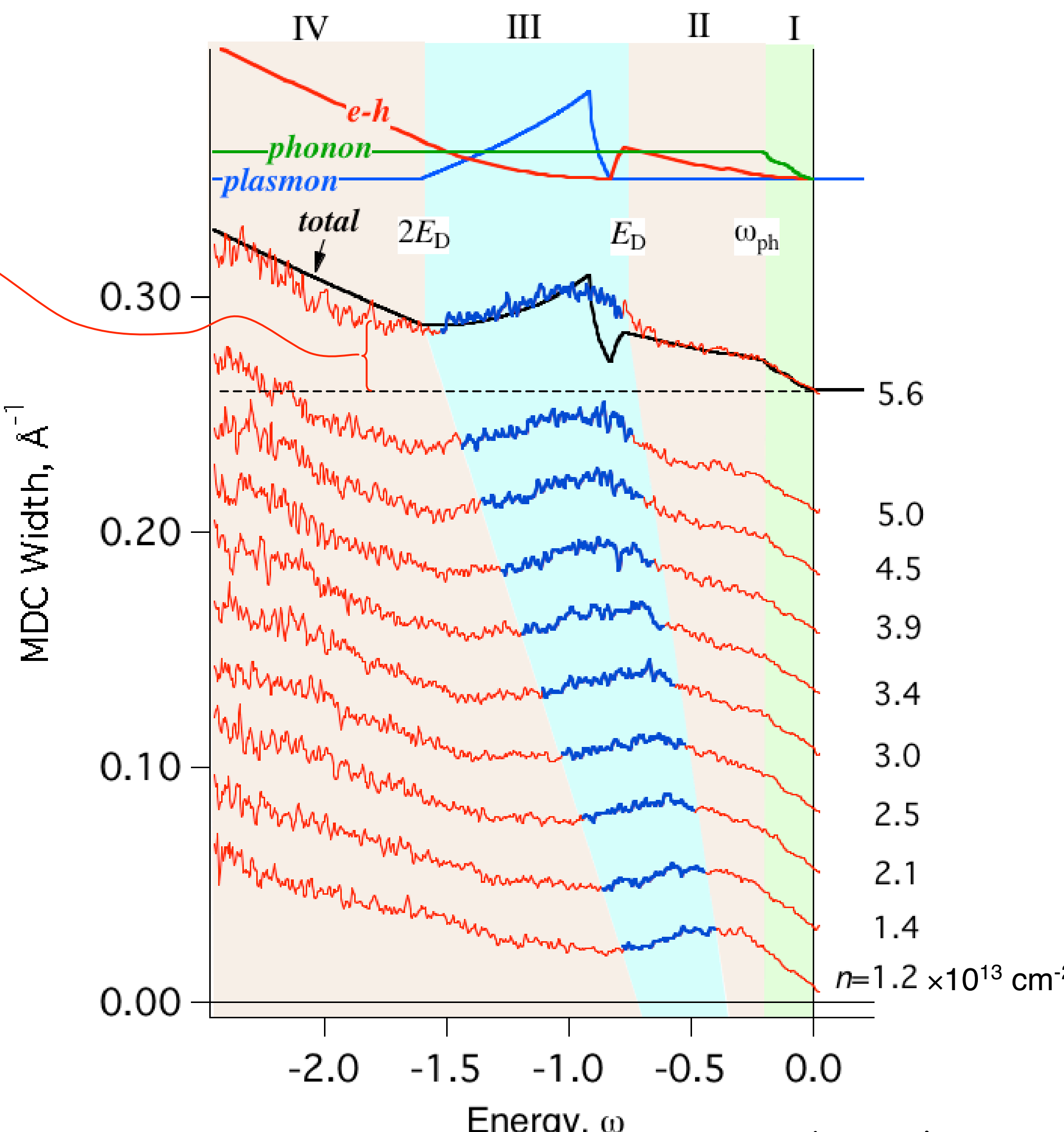
Quantitative Comparison to Data





only 4 free parameters used:
- λ[ph]
- scale factors for e-h & e-pl coupling
- screening const. ε

<u>expt inputs:</u>
shape of bands
eff. mass vs *n*[1]

[1] Novoselov, K. S. et al. Two-dimensional gas of massless Dirac fermions in graphene. Nature 438, 192-200 (2005).

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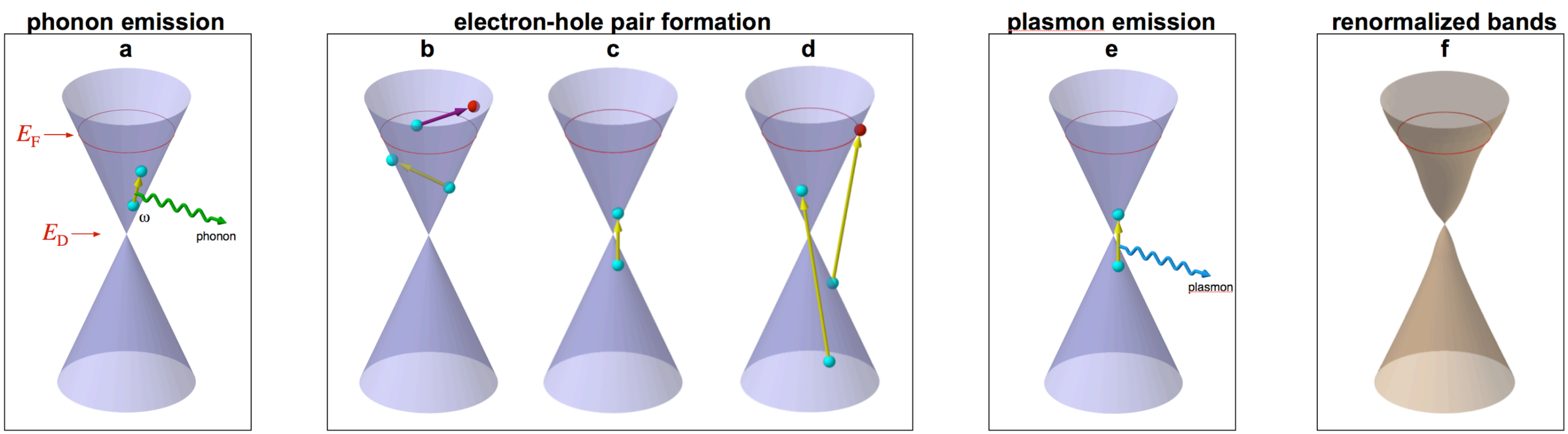
Summary: QP lifetime in graphene



Near E_F | Near E_F | Near E_D | Below E_D | Near E_D

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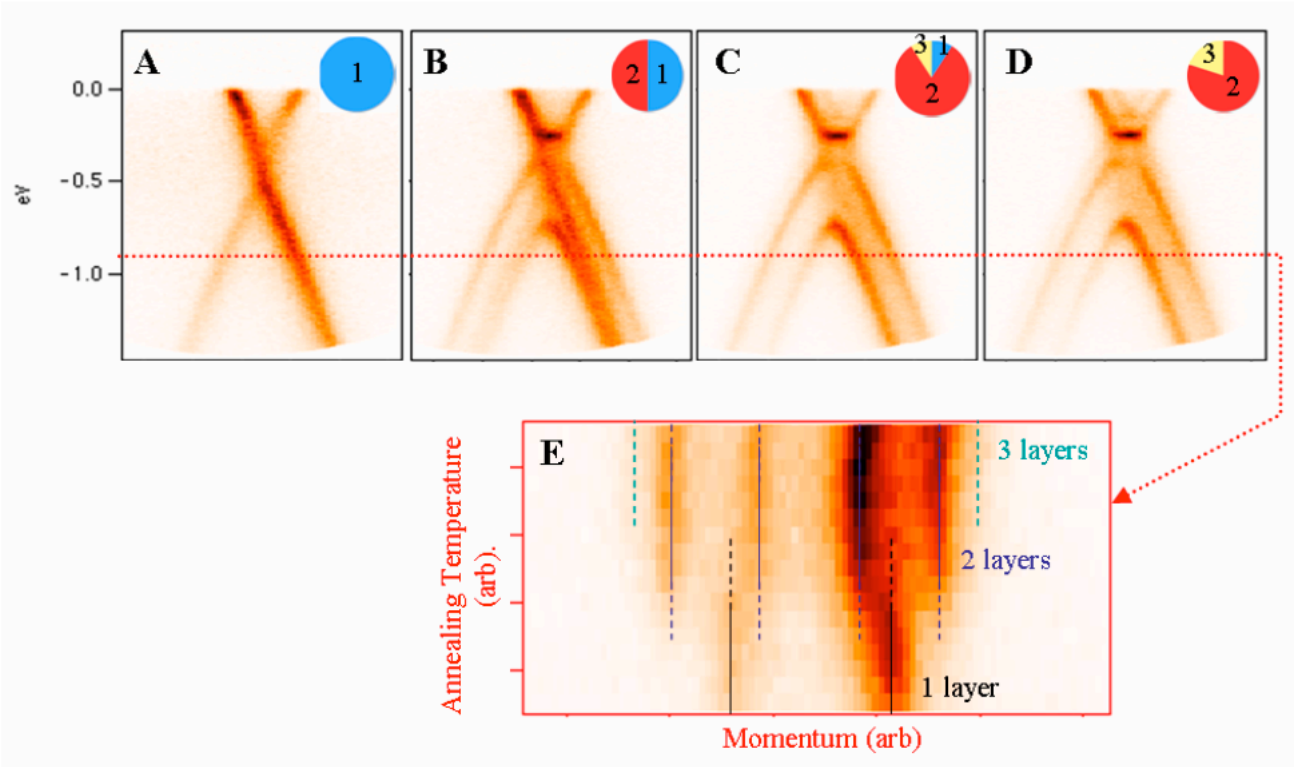
Determining the number of layers

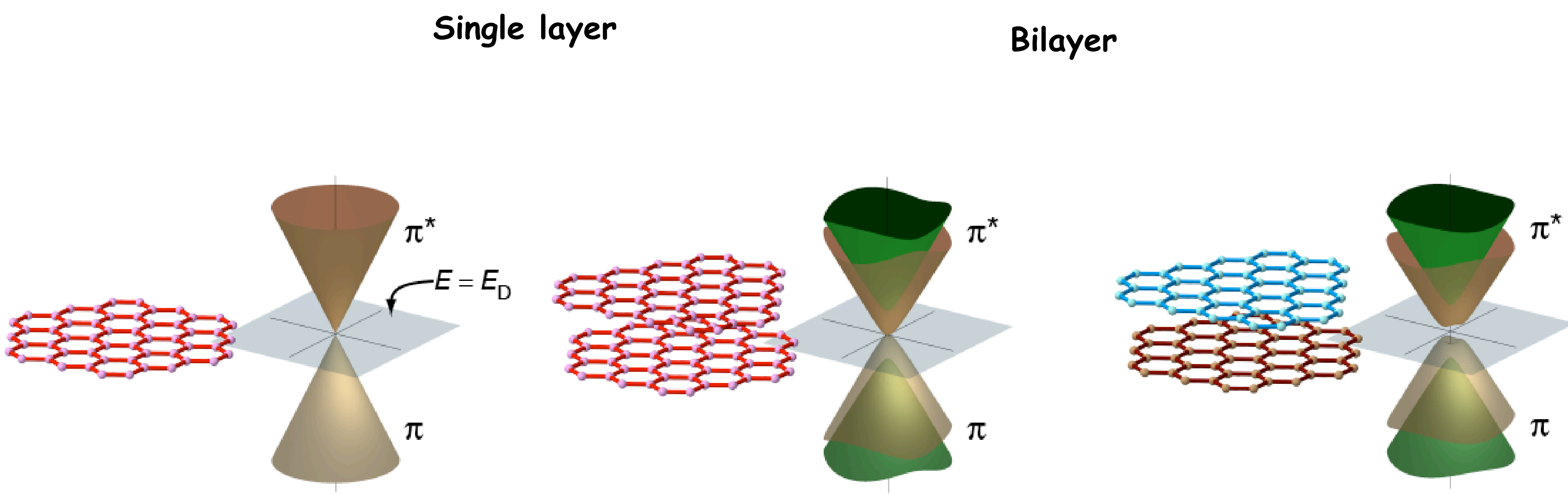
We count the number of π states



Ohta, T., Bostwick, B., Seyller, T., Horn, K. & Rotenberg, E.
Controlling the Electronic Structure of Bilayer Graphene. Science 313, 951-954 (2006).

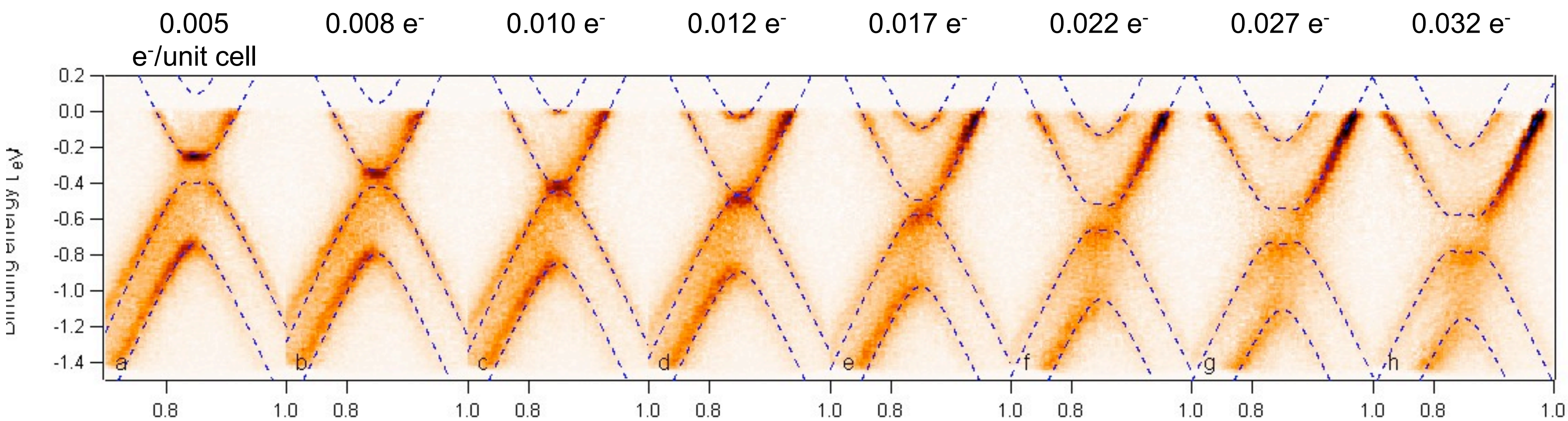
⅄ Controlling gap between π and π* bands in bilayer graphene [1]

Evolution of the Bandstructure [2]



[1] Ohta, T., Bostwick, B., Seyller, T., Horn, K. & Rotenberg, E.
Controlling the Electronic Structure of Bilayer Graphene. Science 313, 951-954 (2006).
[2] McCann, E. and V.I. Fal'ko, *Landau-level Degeneracy and Quantum Hall Effect in a Graphite Bilayer.* Phys. Rev. Lett., 2006. **96**: p. 086805.

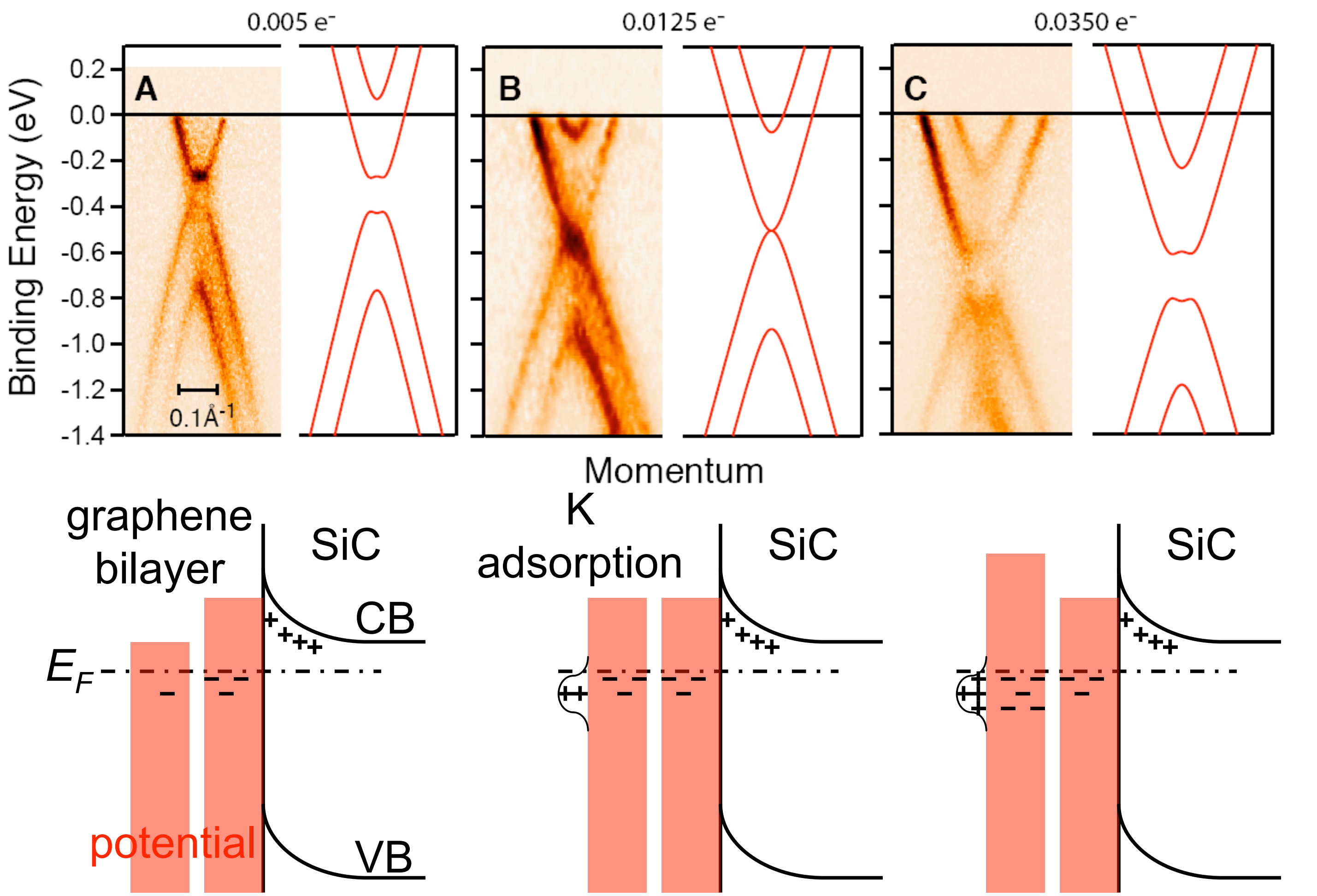
Evolution of π bands on surface doping



- **Deposition of potassium**
- **Shift of π band due to increased total carrier density**
- **Continuous closing/reopening of the gap**

Ohta, T., Bostwick, B., Seyller, T., Horn, K. & Rotenberg, E.
Controlling the Electronic Structure of Bilayer Graphene. Science 313, 951-954 (2006).

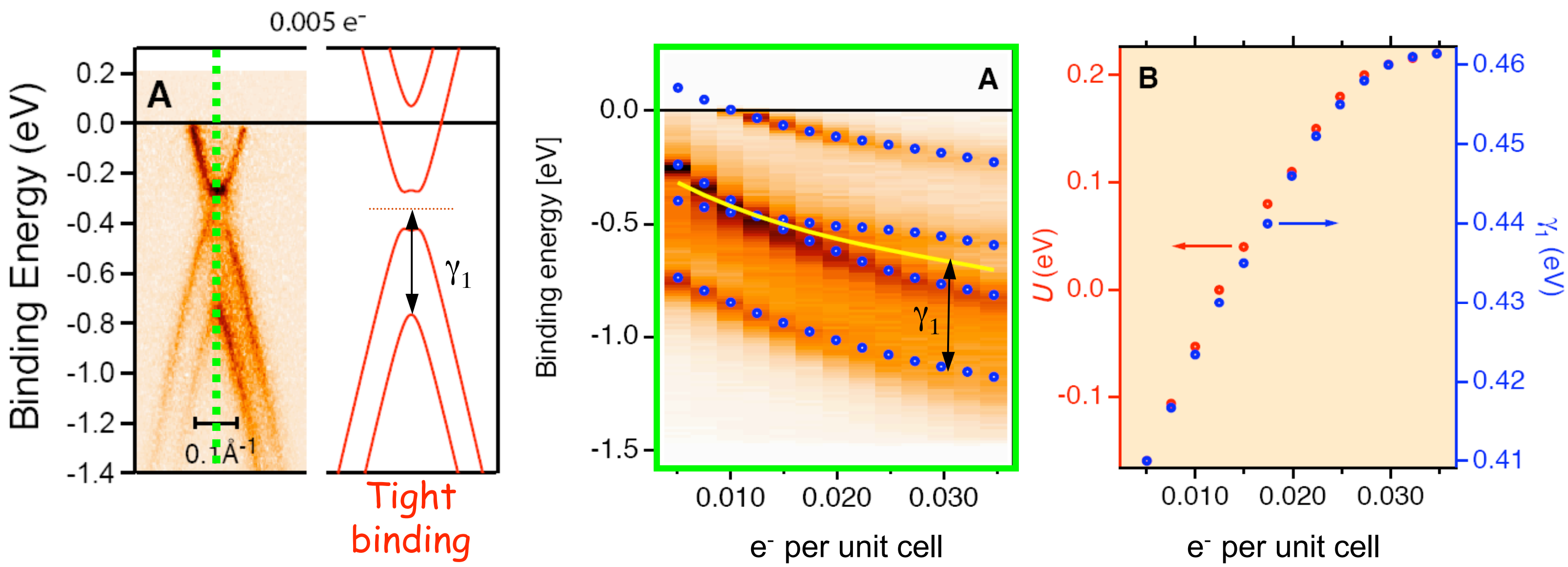
Closing and re-opending of the gap between π and π* band



Non-equal charge distribution due to short interlayer screening length

Ohta, T., Bostwick, B., Seyller, T., Horn, K. & Rotenberg, E.
Controlling the Electronic Structure of Bilayer Graphene. Science 313, 951-954 (2006).

Evolution of π and π* bandgap and tight binding parameters



U: on-site Coulomb energy
γ1: NN interlayer hopping integral

- **π orbital overlap between adjacent layers**
  →γ_1 increases at higher electron density
  - smaller interlayer distance caused by a shorter screening length

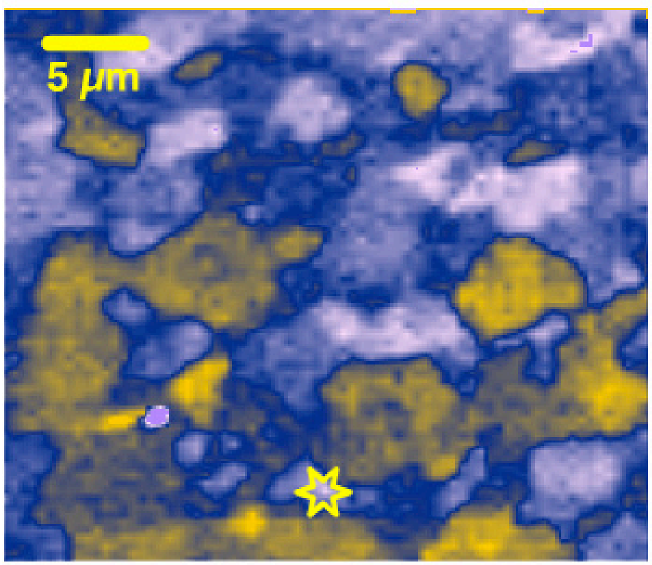
Ohta, T., Bostwick, B., Seyller, T., Horn, K. & Rotenberg, E.
Controlling the Electronic Structure of Bilayer Graphene. Science 313, 951-954 (2006).

TB: McCann and Fal'ko
Phys. Rev. Lett. 96, 086805 (2006).

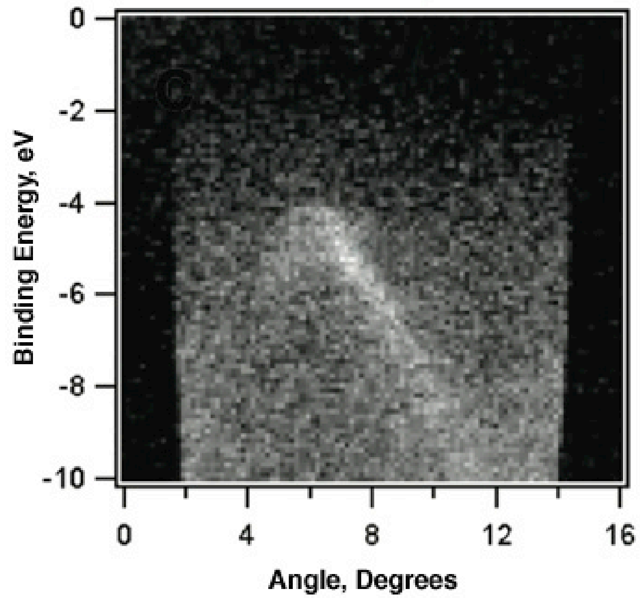
The future: towards 50nm spatial resolution

We succeeded to measure ARPES of graphite using 300 nm probe size

HOPG Graphite
Imaged with Valence Band Contrast



Valence Band



We are heading towards 50 nm spot size to measure bandstructure of
- graphene and multilayers under bias conditions
- individual CNTs?
- etc

Summary

- One-layer: The lifetime of the carriers is determined by
  - Electron-phonon coupling
  - Electron-hole pair generation
  - Electron-plasmon coupling

- These many body effects
  - profoundly distort the bands over energy scale $2(E_F - E_D)$
  - become inseparable at lower doping

- Electron-phonon coupling constant λ=0.1 to 0.3 for n=1-6 x10^{13} cm^{-2}

- Two-layer: Controlling the electronic structure of graphene layers through out-of-plane symmetry
  - Relative potential in bilayer Controls the gap between π and π* states