Out-of-equilibrium electron dynamics in photo-excited topological insulators studied by TR-ARPES

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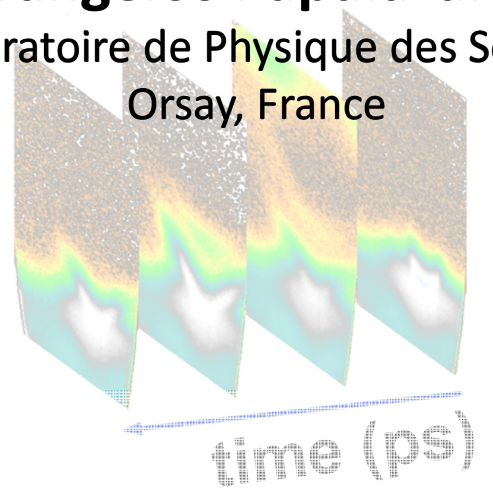
Orsay, France



Acknowledgements

Experimental team

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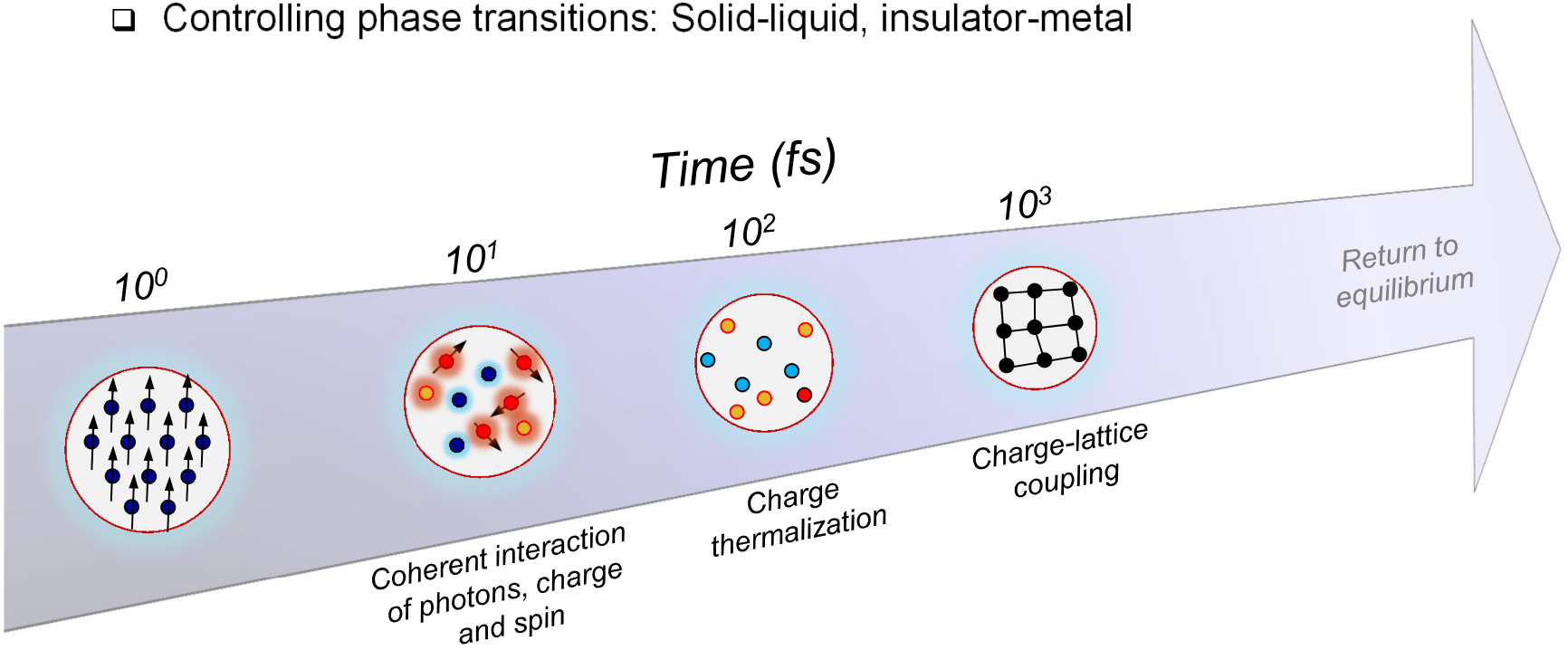
Outline

- Time- and angle-resolved photoelectron spectroscopy
  - *Basic principles*
  - The FemtoARPES setup
- Surface carrier (electrons and holes) relaxation dynamics in Bismuth chalcogenide compounds (n- and p-type Bi_2Te_3, Bi_2Te_2Se)
- Tuning a Schottky barrier in a photo-excited topological insulator with transient electron-hole asymmetry in Dirac-like surface states

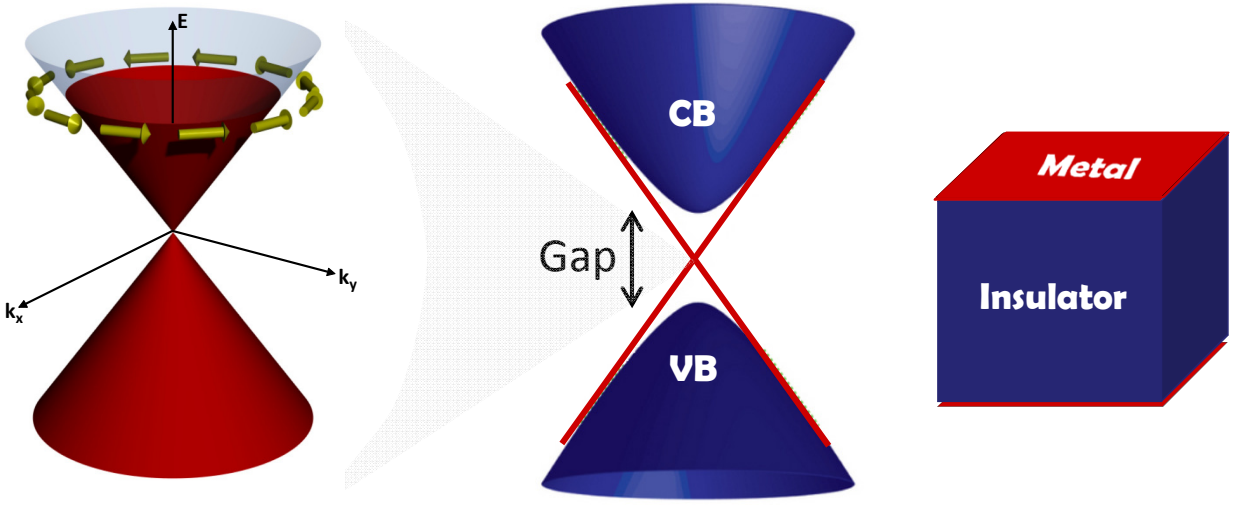
Why ultrafast spectroscopy?

Studying matter out-of-equilibrium

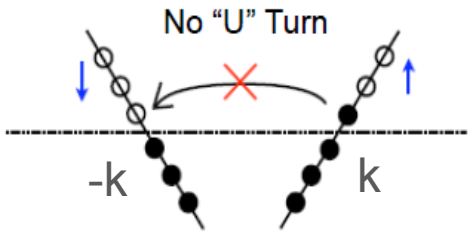
- Resolves non-equilibrium dynamics of fundamental times and spatial scales of electronic and nuclear motion.
- Understanding the relaxation mechanisms: electron-phonon coupling
- Determine the key mechanism leading to photo-induced phase transitions
- Controlling phase transitions: Solid-liquid, insulator-metal



3D Topological insulator: A novel quantum material



Destructive Interference!



A new type of 2DEG with unusual properties:

- A Z2 topological invariant (single Dirac cone, massless Dirac fermions) ⇒ Linear energy dispersion
- Helical spin texture

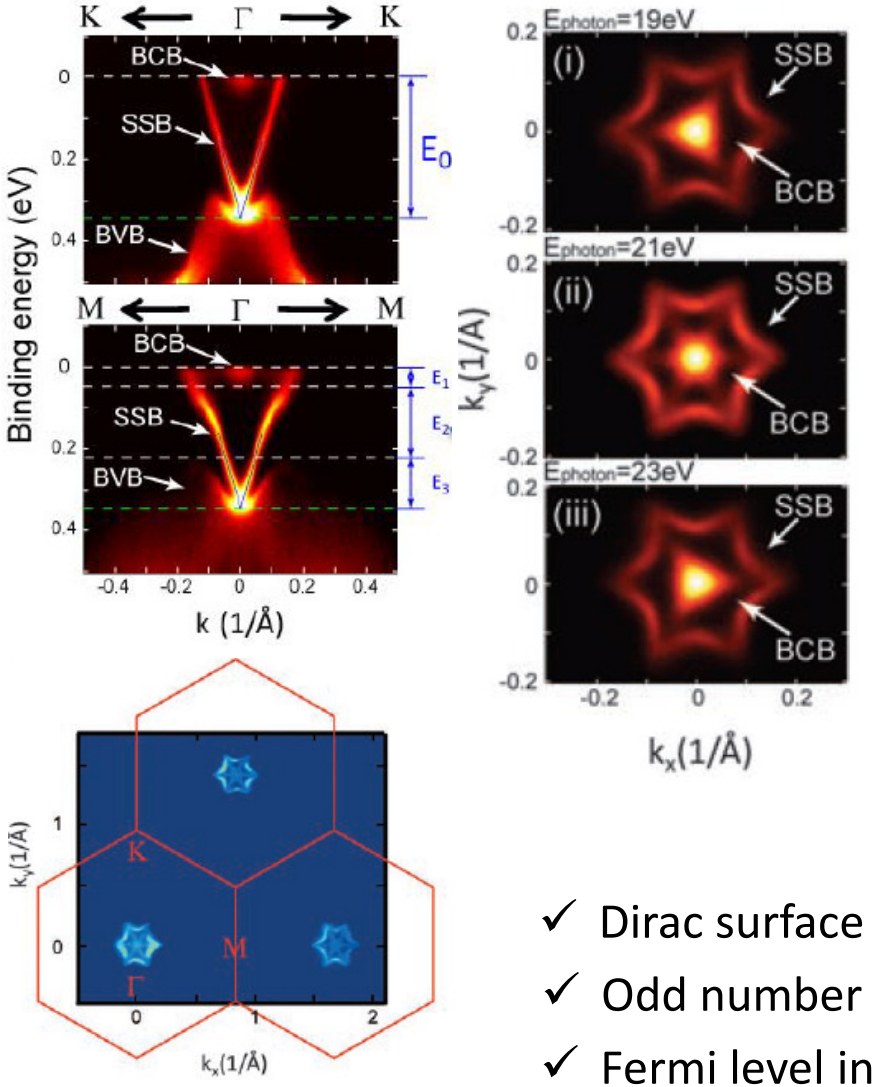
  For in-gap Dirac fermions their spin lies in-plane and perpendicular to its wave-vector
- Quasiparticle back-scattering with non-magnetic impurities is suppressed

  Time-inversion symmetry protects fermions from backscattering

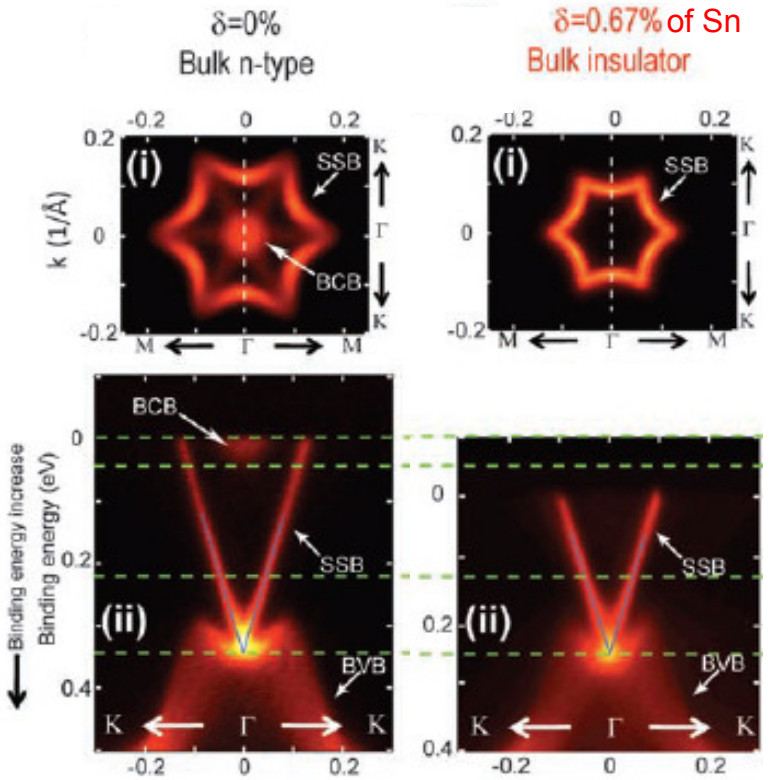
Hsieh *et al.*, Nature (2009)

ARPES study of the topological insulator Bi_2Te_3

Single Dirac cone in the Brillouin zone



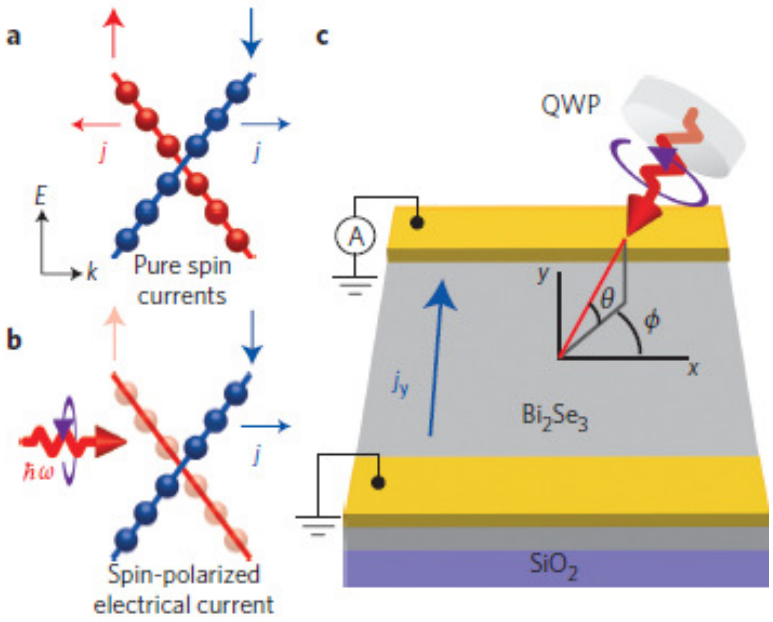
Tuning Fermi level with non-magnetic impurities

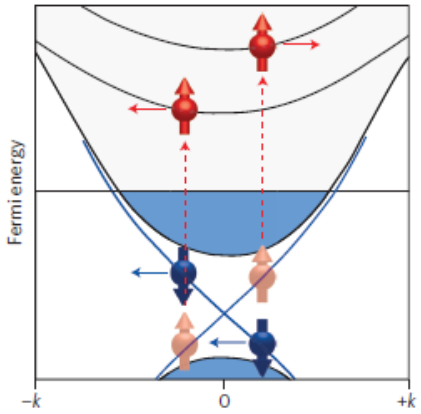


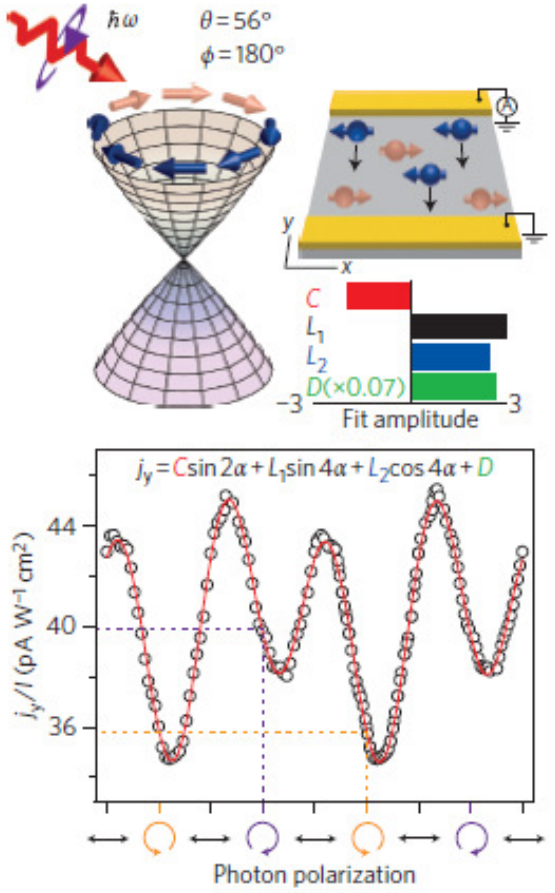
Chen *et al.*, Science (2009)

- ✓ Dirac surface states
- ✓ Odd number of Dirac cones in the Brillouin zone
- ✓ Fermi level in the gap

Towards “topotronics” ?







Polarization-dependent photocurrents that stem from the helical nature of Dirac surface states coexists with a bulk photocurrent

McIvan *et al.*, Nature Nano (2011)

Junk *et al.*, Phys. Rev. B (2013)

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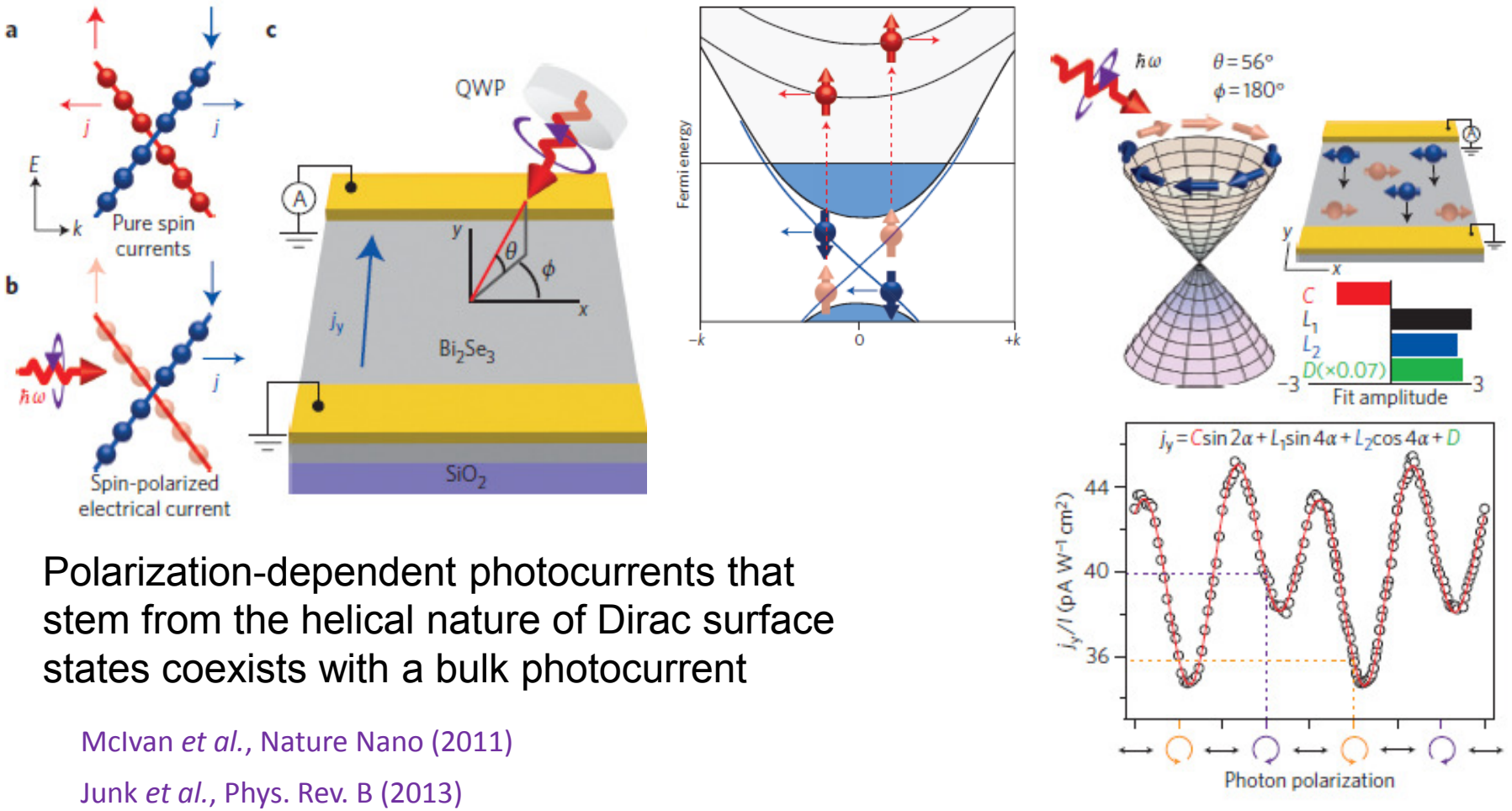


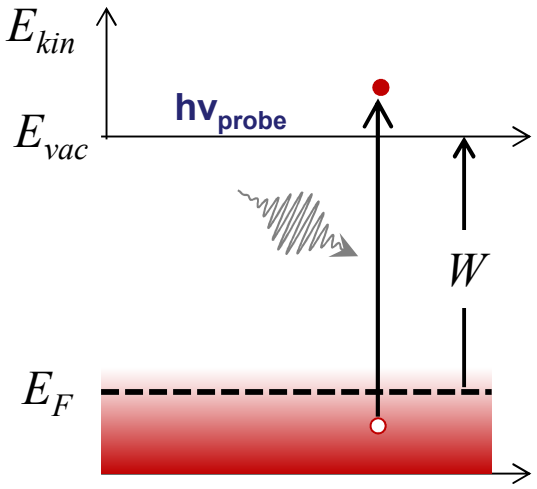
Polarization-dependent photocurrents that stem from the helical nature of Dirac surface states coexists with a bulk photocurrent

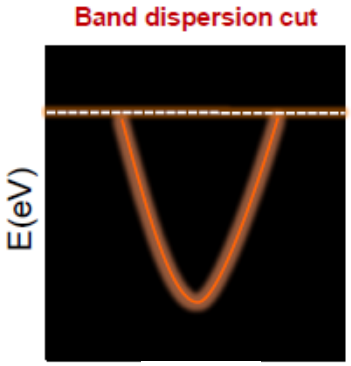
McIvan *et al.*, Nature Nano (2011)

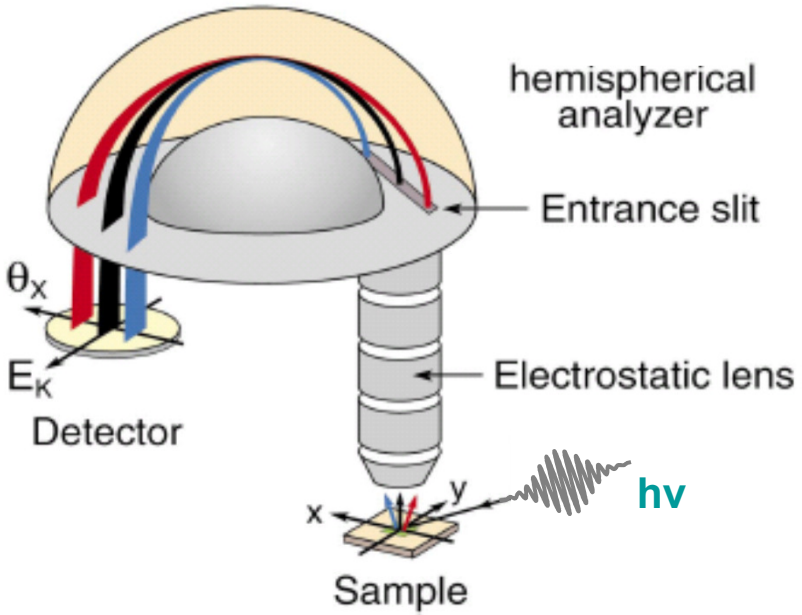
Junk *et al.*, Phys. Rev. B (2013)

- ❑ What is the interband and intraband scattering rate of topologically protected surface states?

Angle-resolved photoelectron spectroscopy (ARPES)

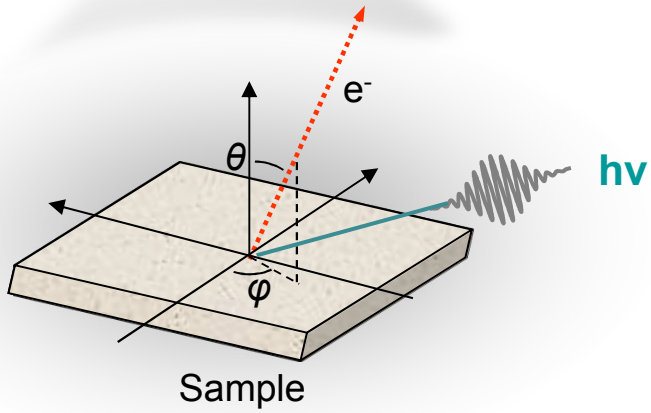






$$E_B = h\nu - W - E_{kin}$$

$$\boldsymbol{k}_{\parallel} = \frac{1}{\hbar}\sqrt{2mE_{kin}} \cdot \sin\vartheta$$

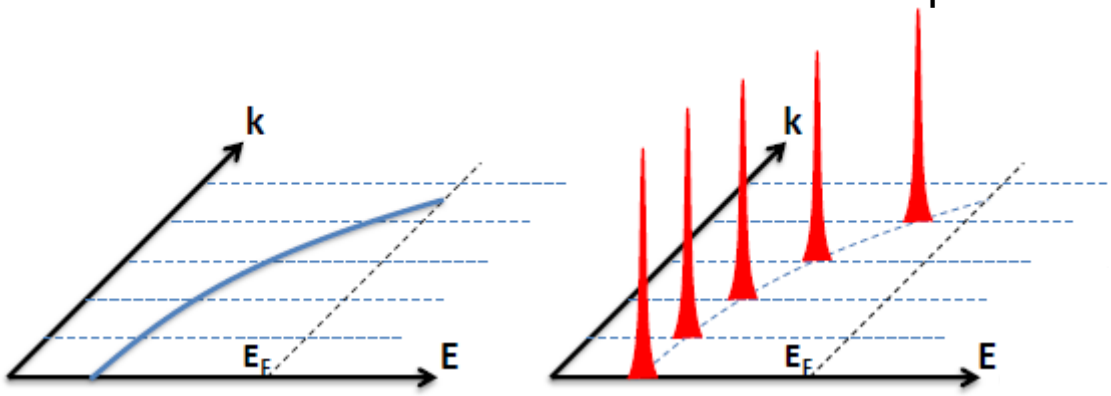


Measure of photoelectron intensity as a function of E_{kin} and θ

- Direct access to electronic population and its density of states N(E)

Basic principle of ARPES

Electron band structure ARPES spectrum



Band dispersion Non-interacting system

The measured photoelectron intensity $I(k, E_{kin})$ in the sudden approximation limit:

$$I(k, E_{kin}) \propto f_{FD}(h\nu, T) \cdot \delta(h\nu - E_f - E_i) \cdot \left|\langle \psi_{f,N} | H_{int} | \psi_{i,N} \rangle\right|^2$$

Fermi-Dirac distribution

$$|\psi_{i,N}\rangle = |\varphi_{i,k}\rangle |\psi_{i,k,N-1}\rangle$$

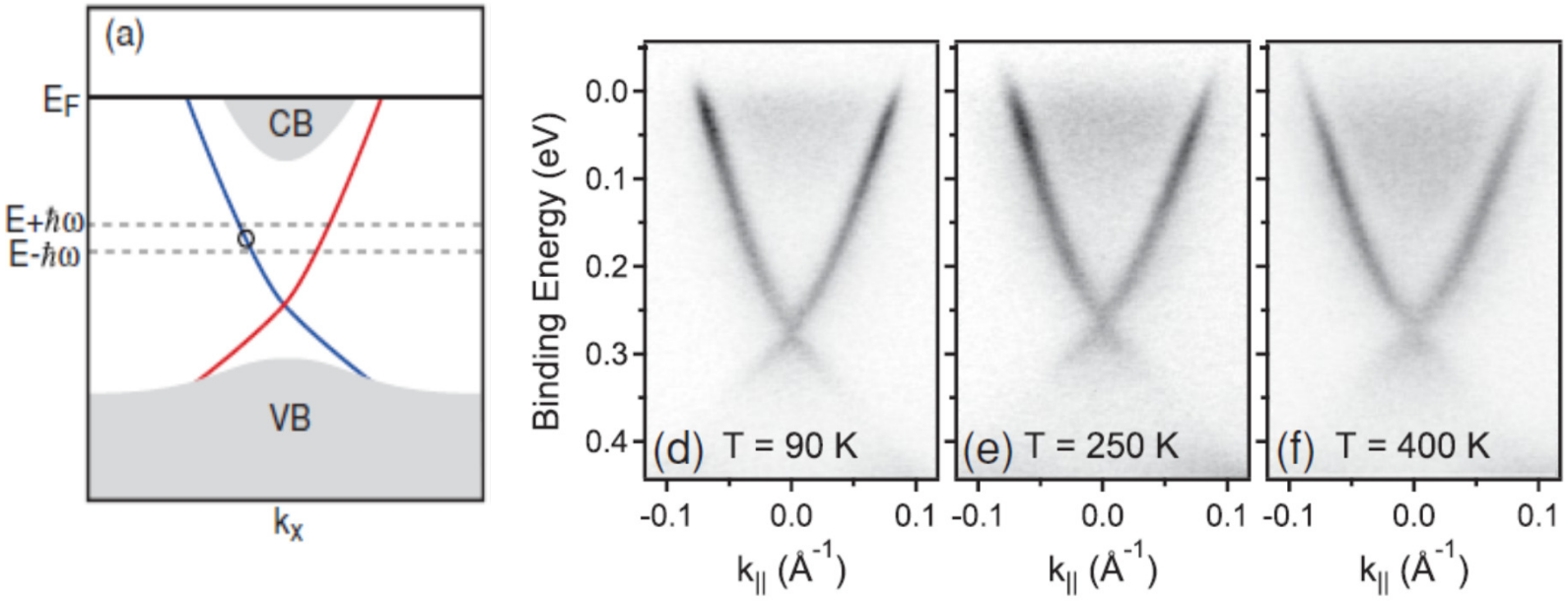
$$|\psi_{f,N}\rangle = |\varphi_{f,k,h\nu}\rangle |\psi_{i,k,N-1}\rangle_s$$

1 electron state — N-1 electron system

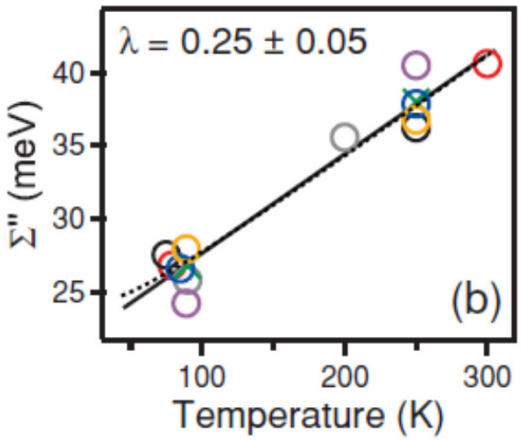
$$I(k, E_{kin}) \propto f_{FD}(h\nu, T) \cdot \left|\langle \varphi_{f,k} | H_{int} | \varphi_{i,k} \rangle\right|^2 \cdot \sum_s \delta(E_{i,N-1} - E_{f,s,N-1} - h\nu) \cdot \left|\langle \psi_{f,s,(N-1)} | \psi_{i,k,N-1} \rangle\right|^2$$

one-particle spectral function

ARPES determination of electron-phonon strength

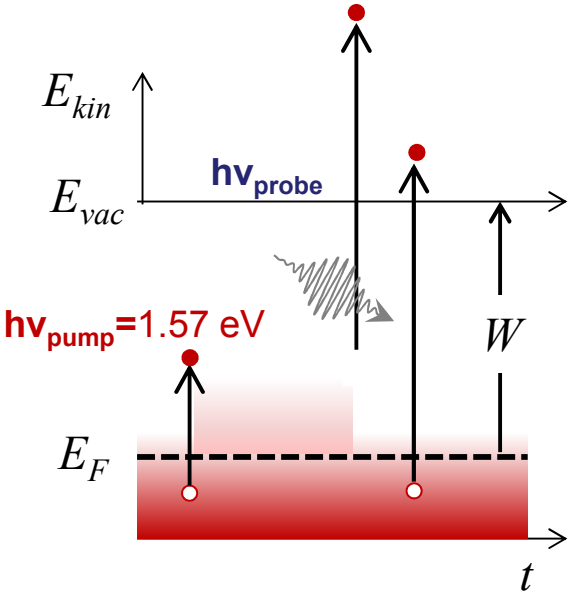


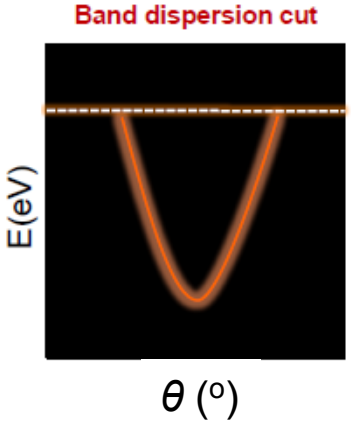
- Typical ARPES based on synchrotron radiation they can only attain a resolution of about 5 meV
- Energy resolution limits the detection energy range
  ➔ impractical to explore low energy interactions

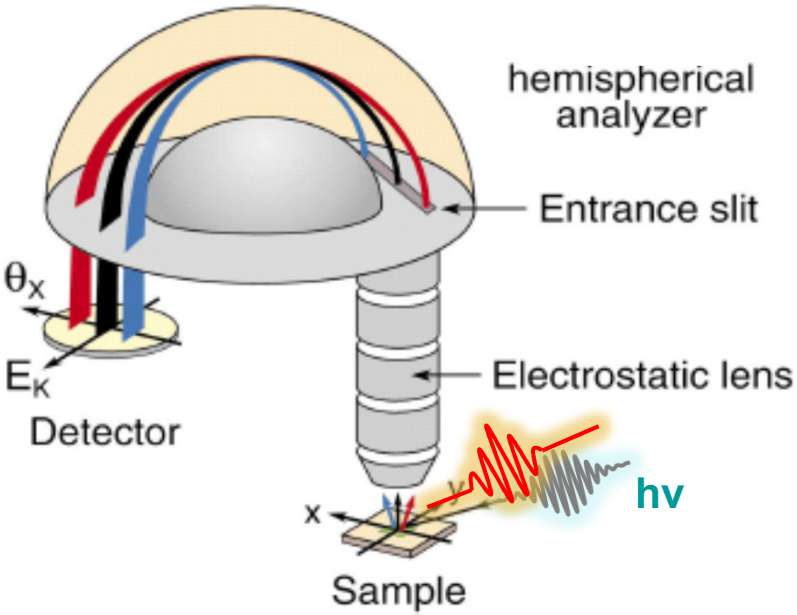


R. C. Hatch *et al.*, PRB 83, 241303 (2011)

Time-resolved UV photoelectron spectroscopy (TR-ARPES)

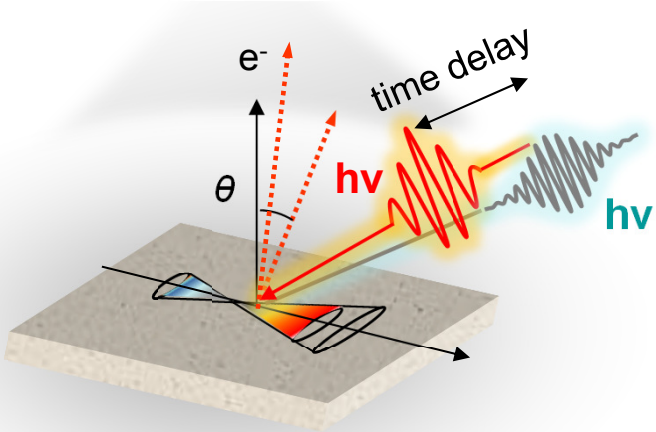






$$E_B = h\nu - W - E_{kin}$$

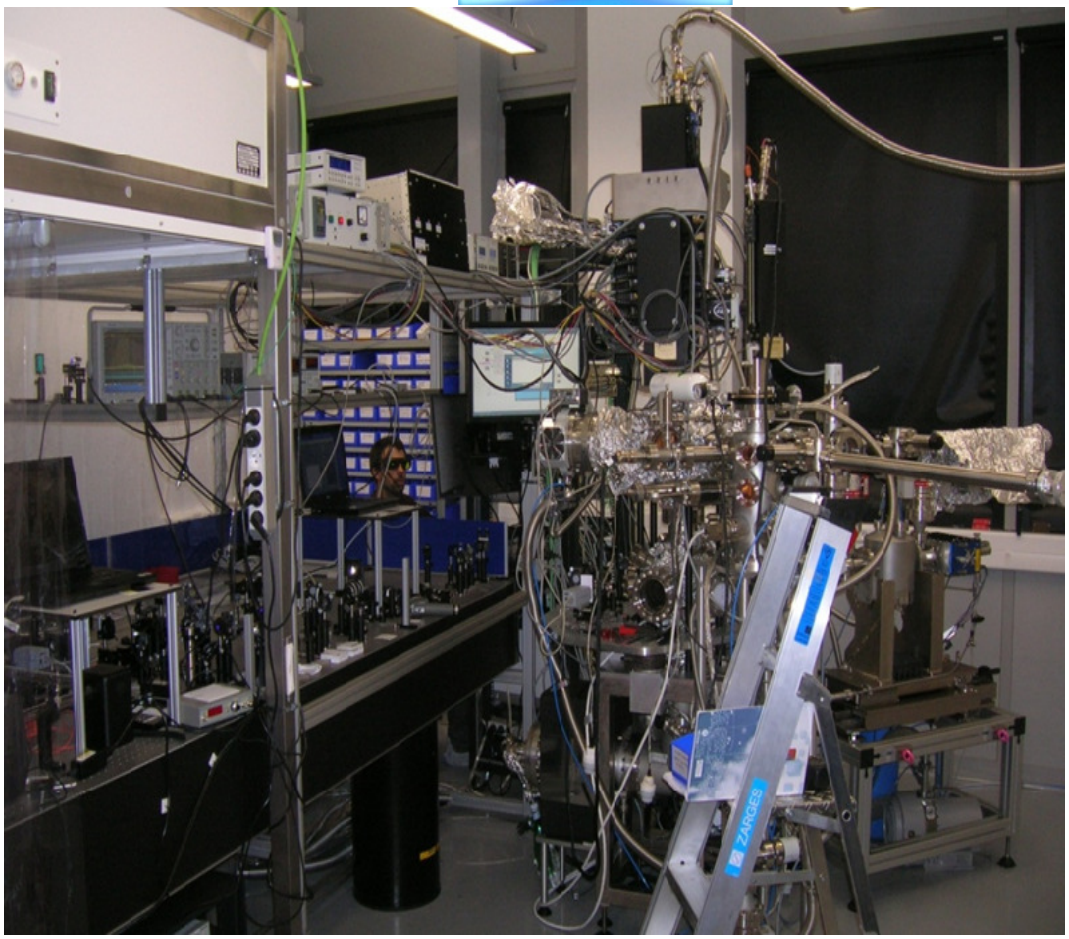
$$k_{\parallel} = \frac{1}{\hbar}\sqrt{2mE_{kin}} \cdot \sin\vartheta$$



Photoelectron intensity as a function of E_{kin} and θ

- Direct access to transient electronic population and its density of states

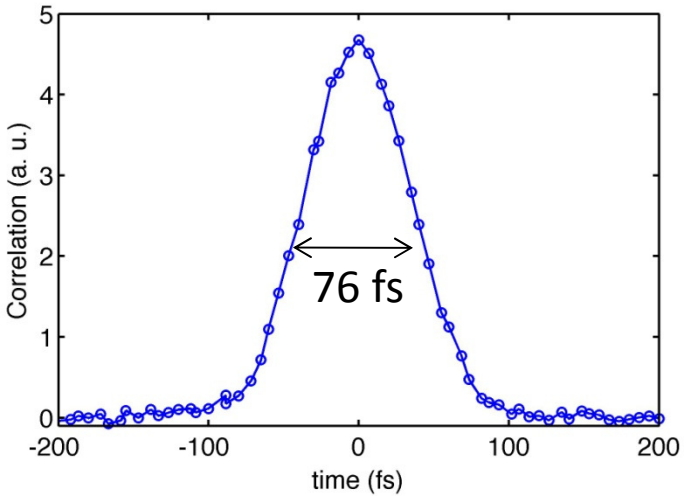
The FemtoARPES set-up

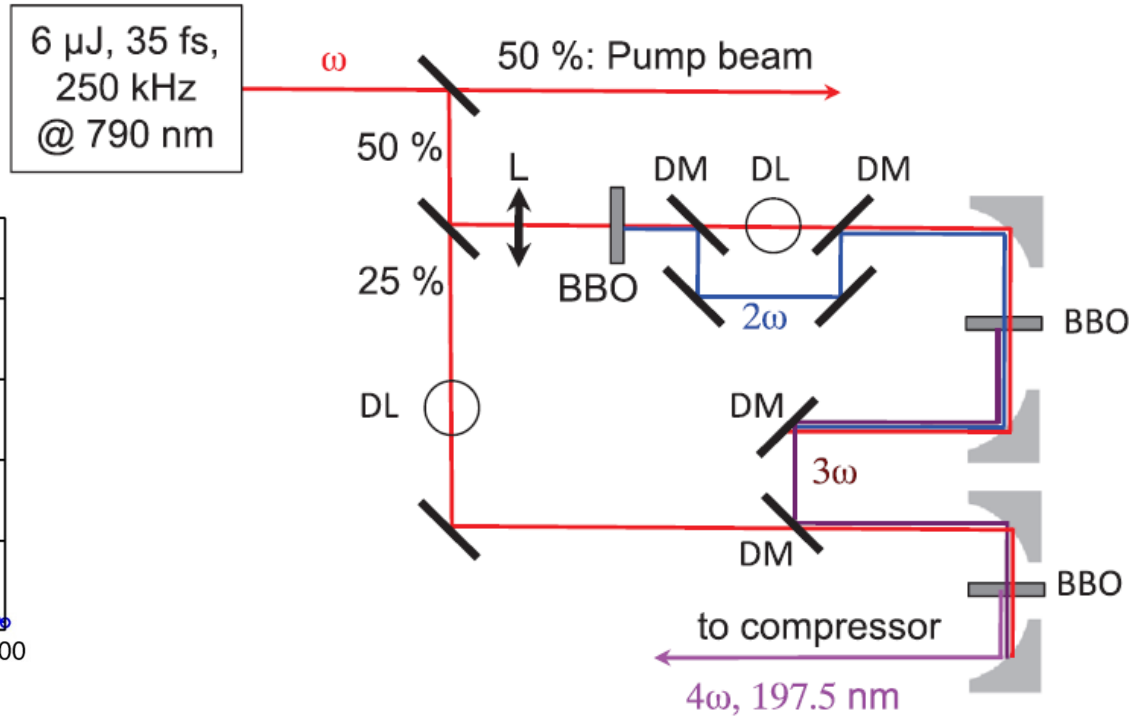


ARPES

- 5-axis manipulator
- Temperature range ~30-800 K
- SPECS Phoibos 150 analyzer
- energy resolution : 80 meV
- angle resolution : < 0.15°
- UHV ~7.10^{-11} mbars

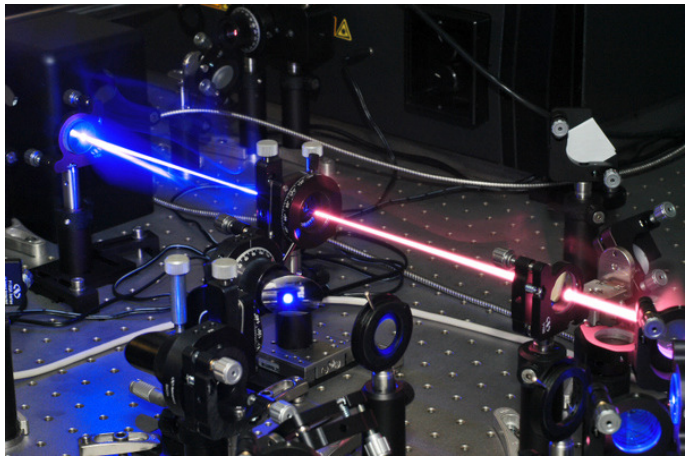
Optical pump – UV probe



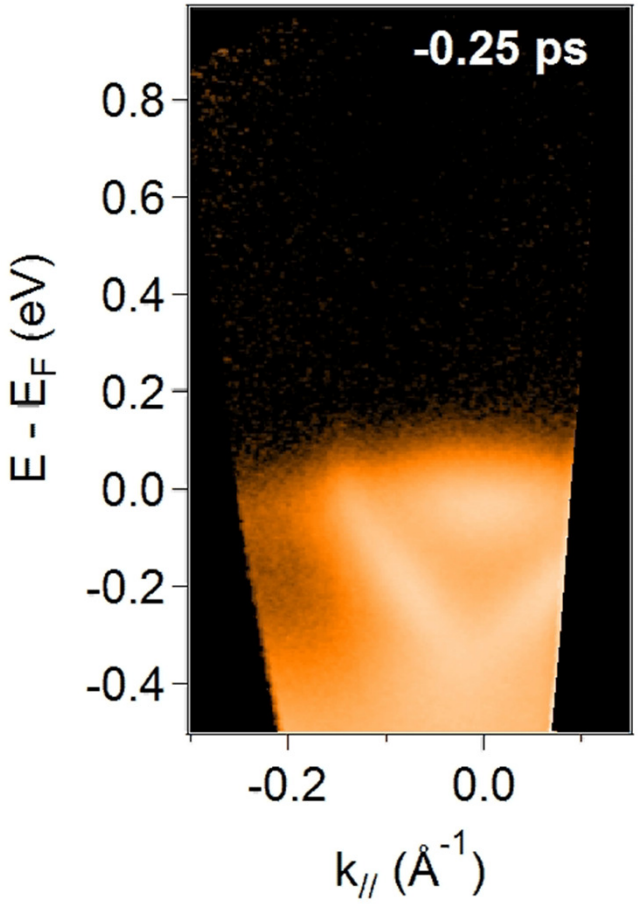


- Rep. rate = 250 kHz
- 4HG UV pulses:
  - 40 pJ per pulse
  - ~10^{13} photons/s
  - Pulse width ~50 fs
  - Beam-spot in focus ≈ 50 μm²

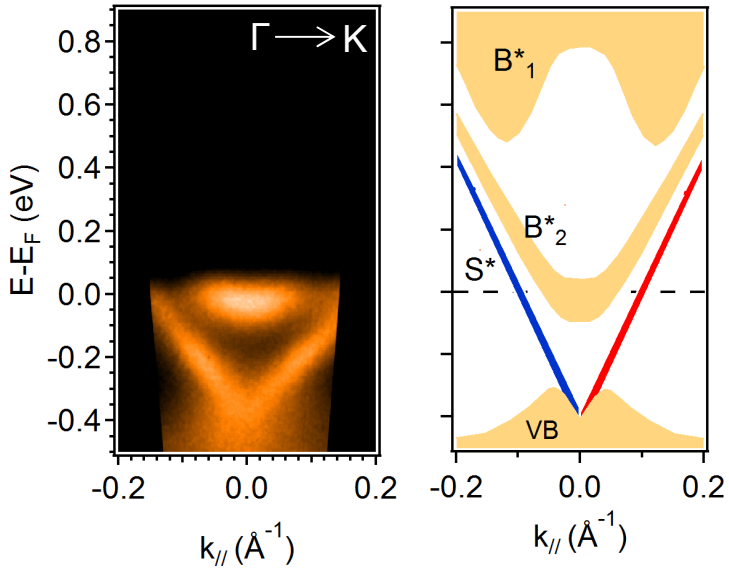
Faure *et al*. RSI 83 (2012)

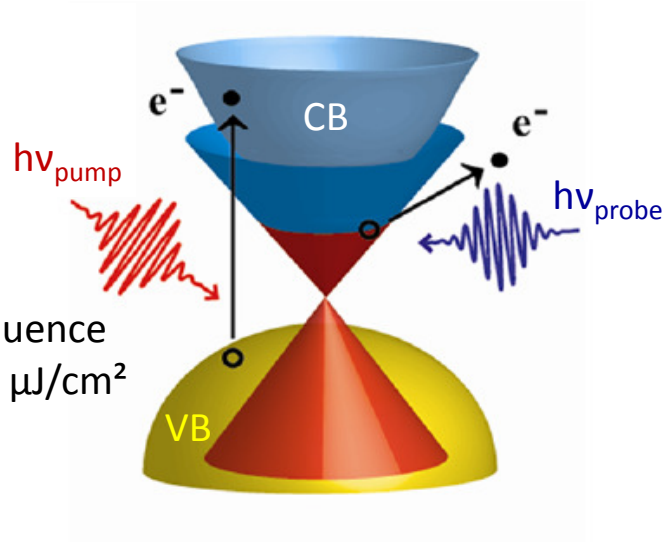


TR-ARPES in n-doped Bi_2Te_3



1PPE with 6.28 eV

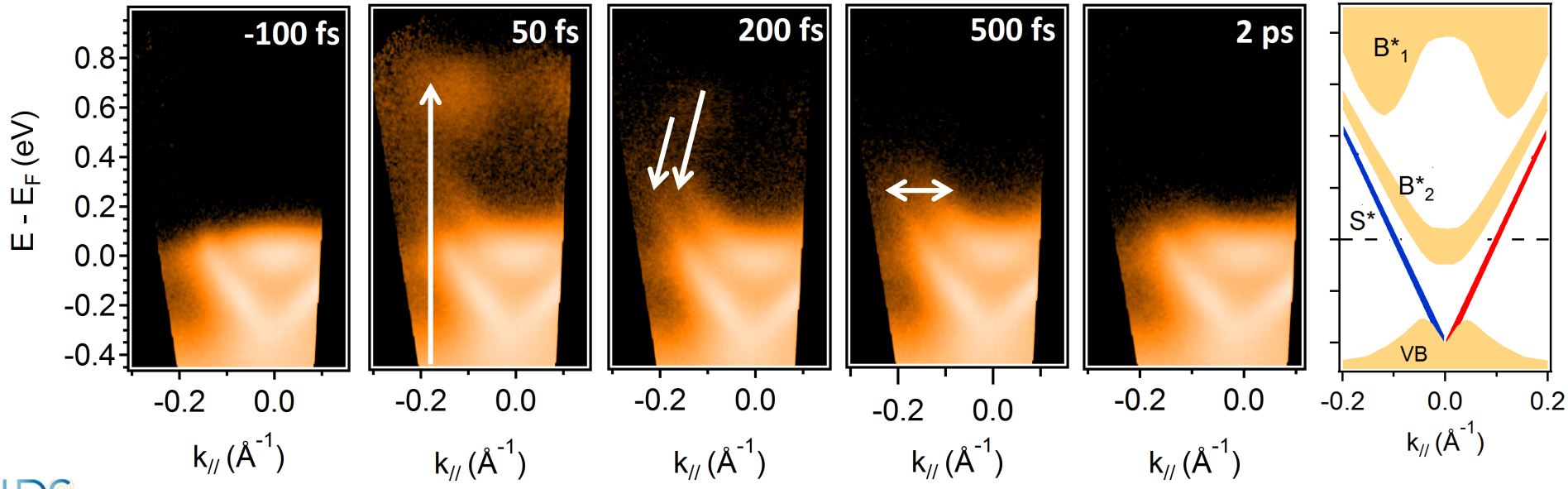




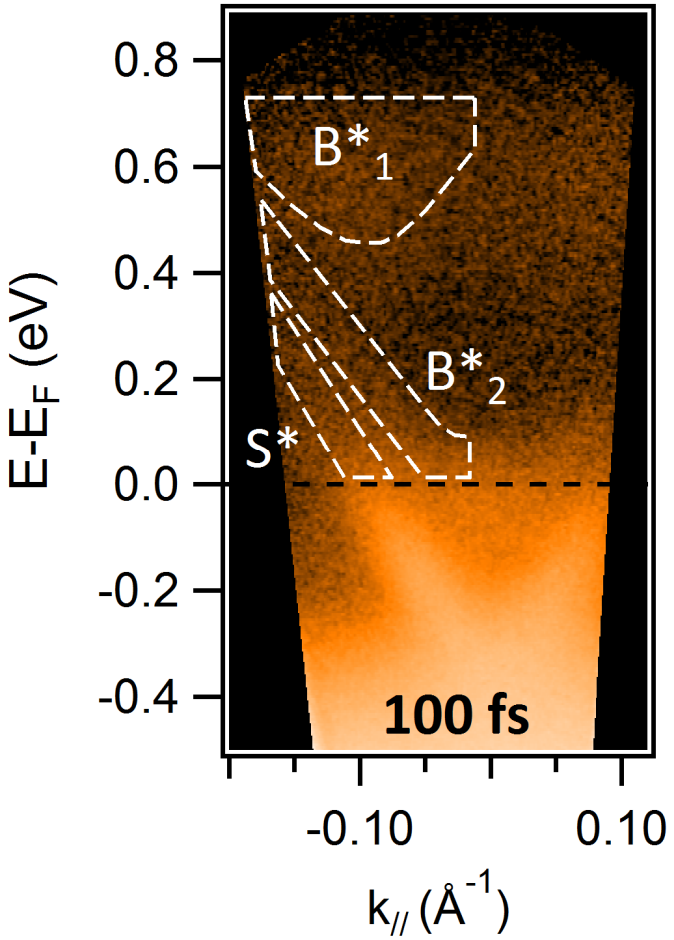
Pump fluence
150 ± 10 µJ/cm²

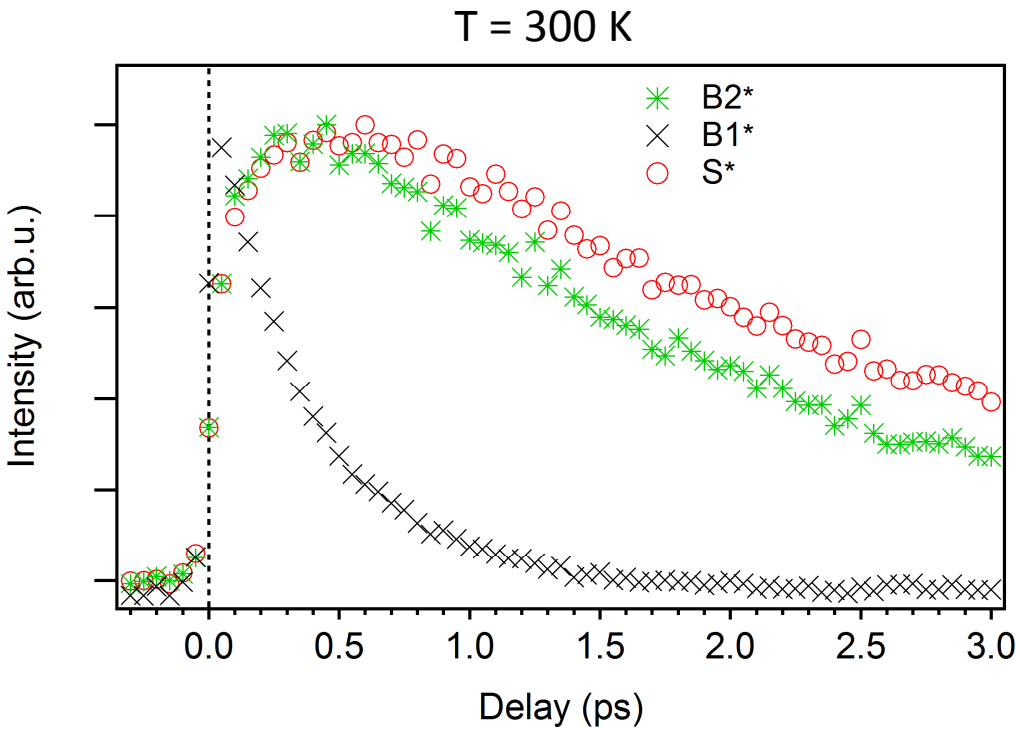
TR-ARPES in n-doped Bi_2Te_3

- Direct excitation of electron population in higher excited states, *i.e.* band B^*_1
- B^*_1 scatters to B^*_2 and S* through interband scattering processes
- Thermalization and relaxation through interband and intraband scattering processes
- Recombination of surface and bulk states



Evolution of charge carrier population in n-doped Bi_2Te_3





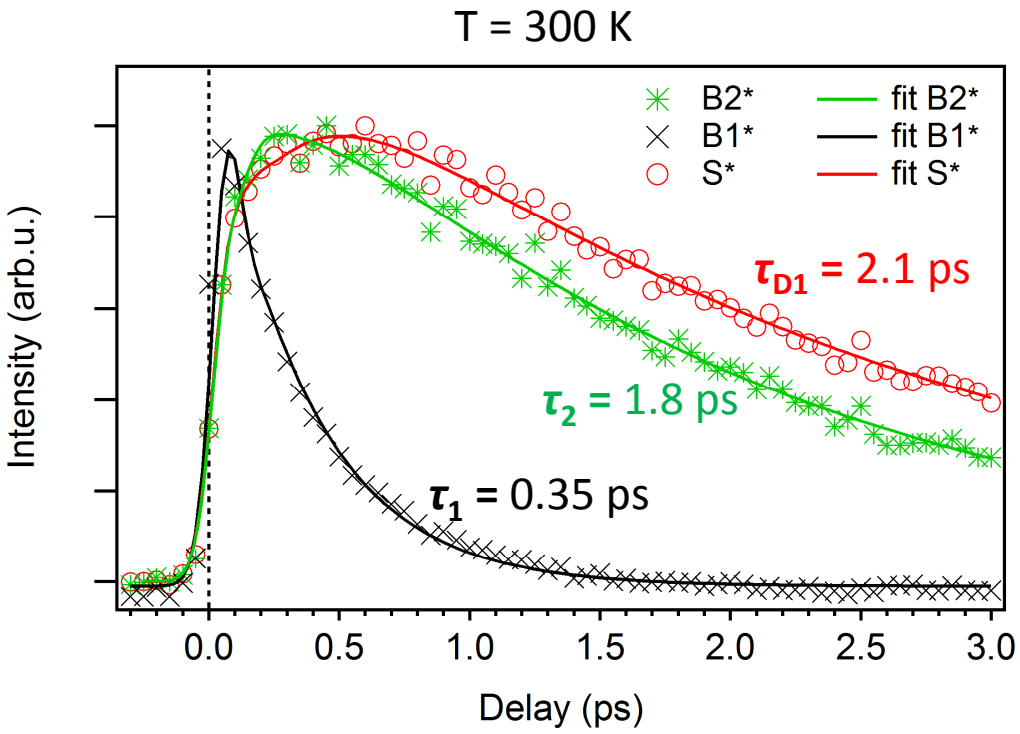
Hajlaoui *et al.*, Nano Lett. (2012)

Evolution of charge carrier population in n-doped Bi_2Te_3

$$\frac{dB_1^*}{dt} = G_1(t) - \frac{B_1^*(t)}{\tau_1}$$

$$\frac{dB_2^*}{dt} = G_2(t) + \alpha\frac{B_1^*(t)}{\tau_1} - \frac{B_2^*(t)}{\tau_2}$$

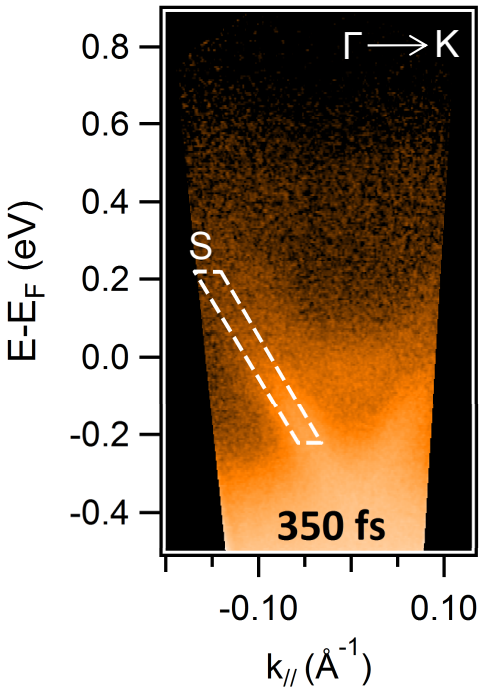
$$\frac{dS^*}{dt} = G_{S^*}(t) + \beta\frac{B_1^*(t)}{\tau_1} - \frac{S^*(t)}{\tau_{D1}}$$

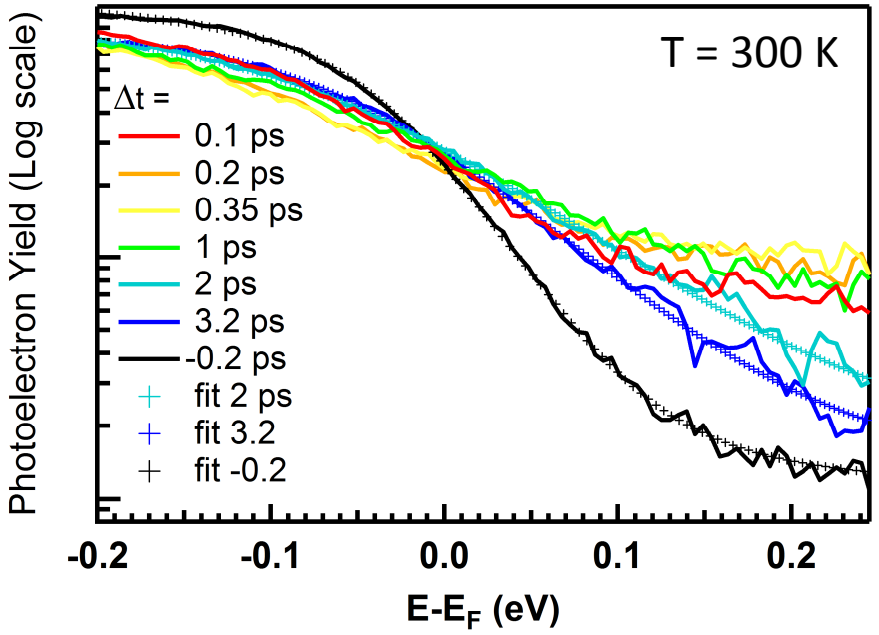


Hajlaoui *et al.*, Nano Lett. (2012)

- Relaxation dynamics fitted with single exponential.
- The very similar values of τ_{D1} and τ_2 indicates that S*(t) and B*$_2$(t) present a parallel evolution ⇒ no explicit insert of a scattering term between them in the rate equations.
- Similarity indicates that an effective carrier exchange takes place between the two bands S*(t) and B*$_2$(t).

Transient Dirac states in n-doped Bi_2Te_3





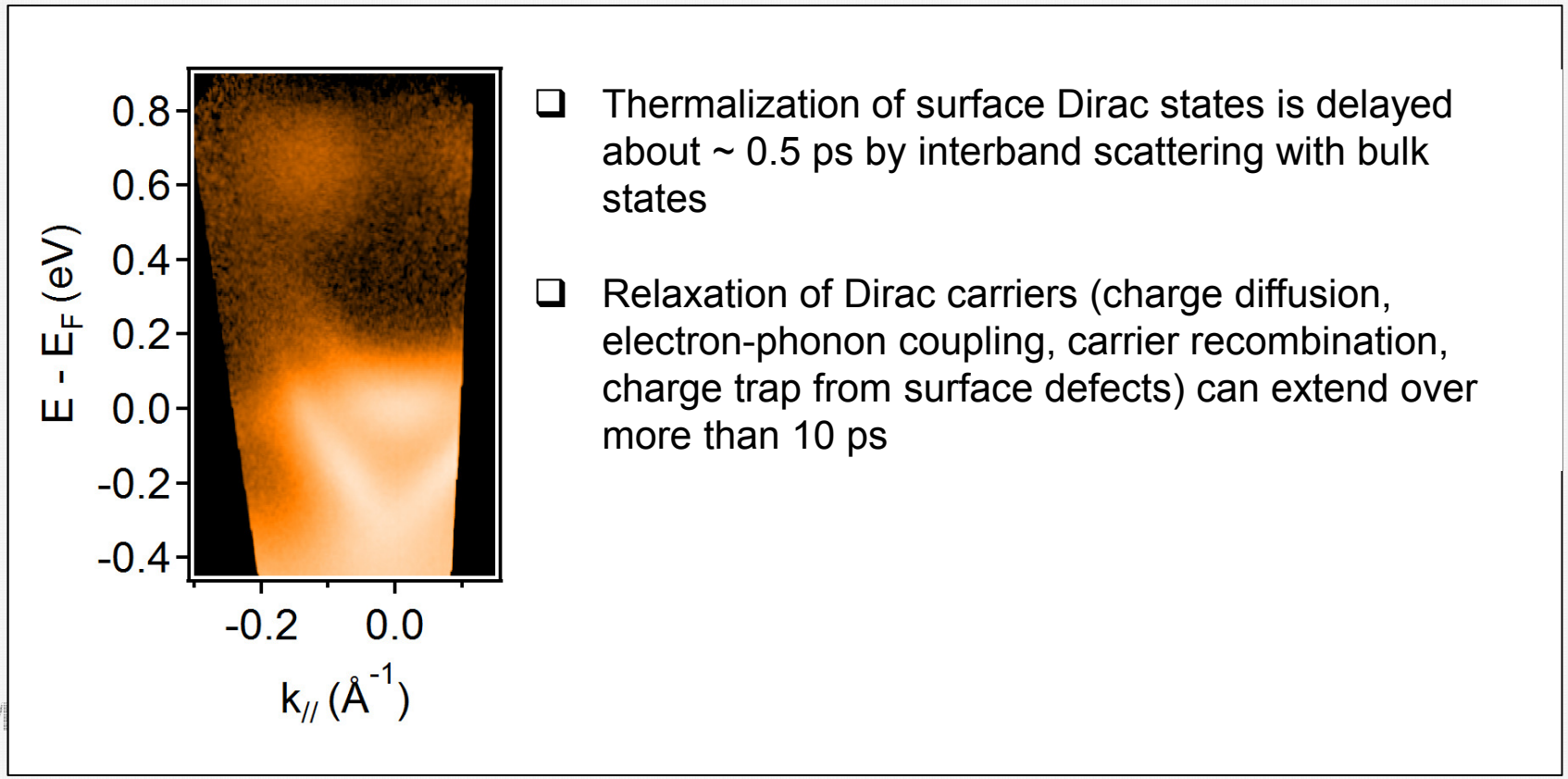
Measured transient k-integrated photoelectron intensity: $I(\epsilon, t) = D_1(\epsilon) * f_{FD}(\epsilon, T(t))$

Thermalization of hot Dirac electrons takes ~0.5 ps :

- Theoretically thermalization of Dirac electrons ~20-40 fs
- Strong presence of high energy electrons.
- The population of S* increases due to flow of charge carriers from the bulk bands.

Scattering distance of Dirac electron ~ $\tau_C\, v_F$ = (0.5 ps)(0.36 m/μs) = 180 nm

Transient Dirac states in n-doped Bi_2Te_3



- Thermalization of surface Dirac states is delayed about ~ 0.5 ps by interband scattering with bulk states
- Relaxation of Dirac carriers (charge diffusion, electron-phonon coupling, carrier recombination, charge trap from surface defects) can extend over more than 10 ps

Thermalization of hot Dirac electrons takes ~0.5 ps :

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- The population of S* increases due to flow of charge carriers from the bulk bands.

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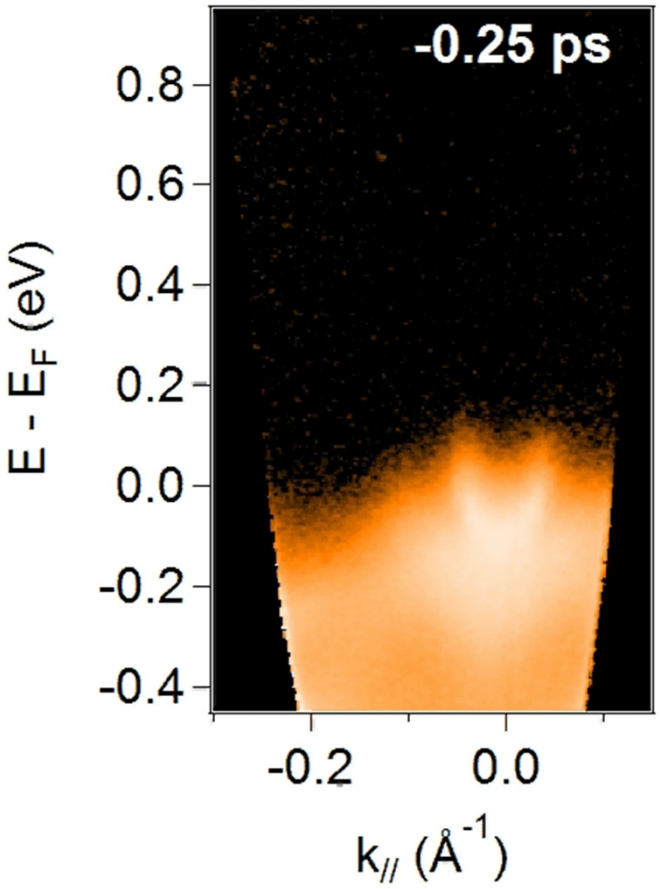
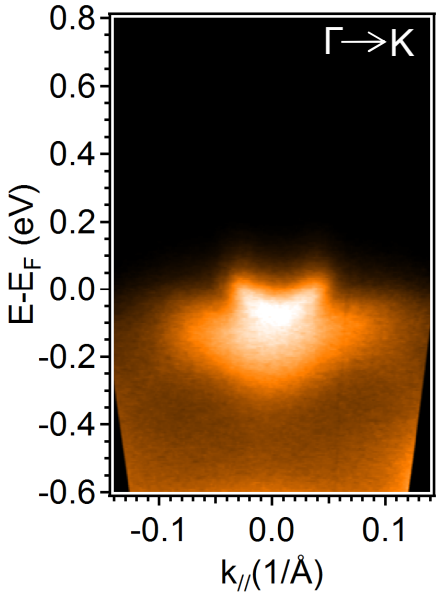
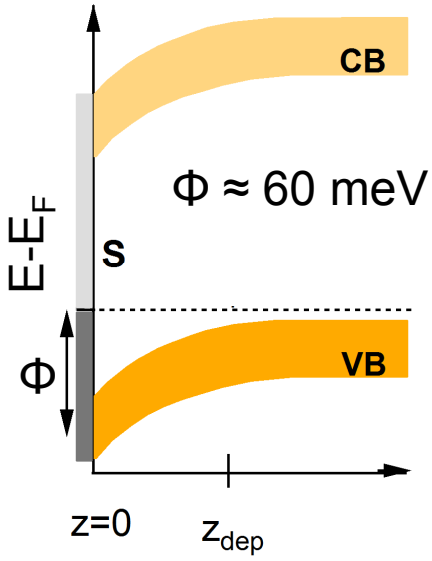
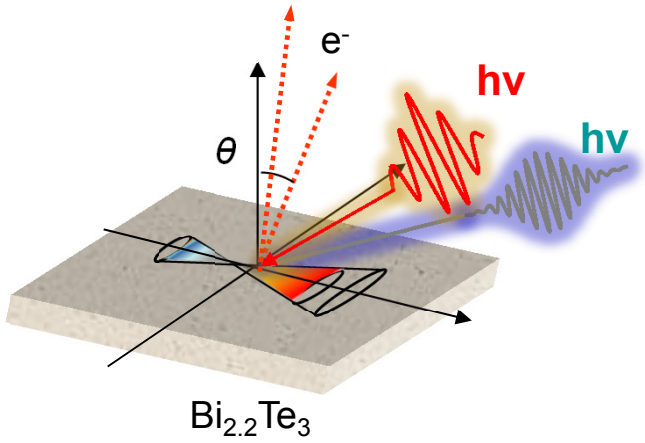
TR-ARPES in $Bi_{2.2}Te_3$ (p-doped)

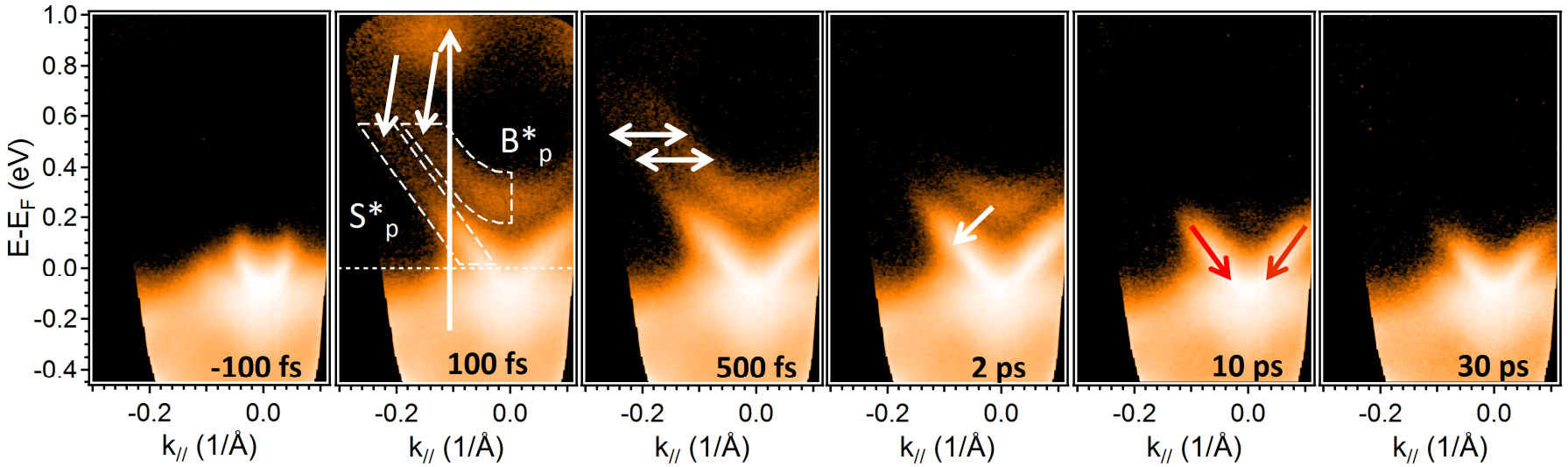






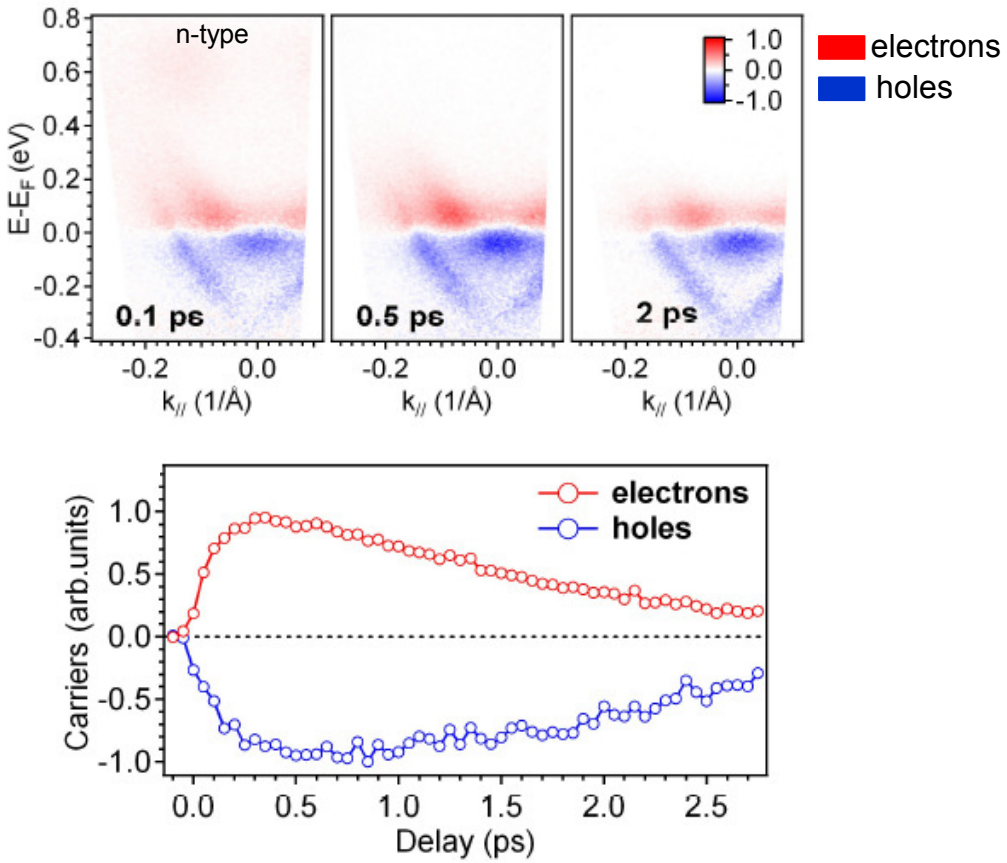


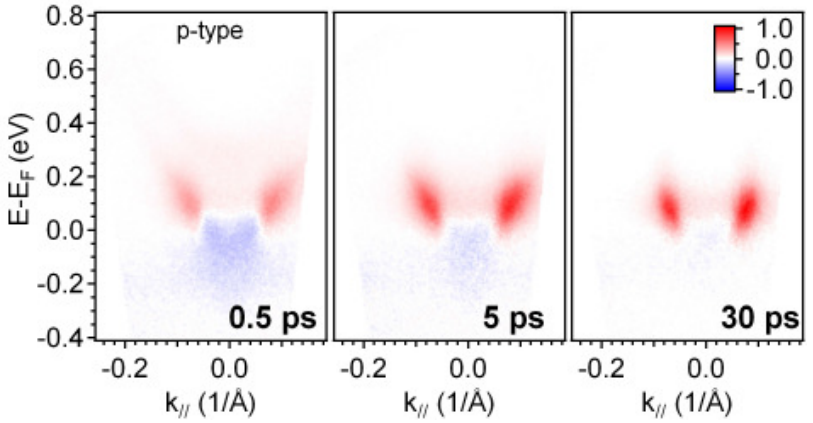
TR-ARPES in $Bi_{2.2}Te_3$ (p-doped)

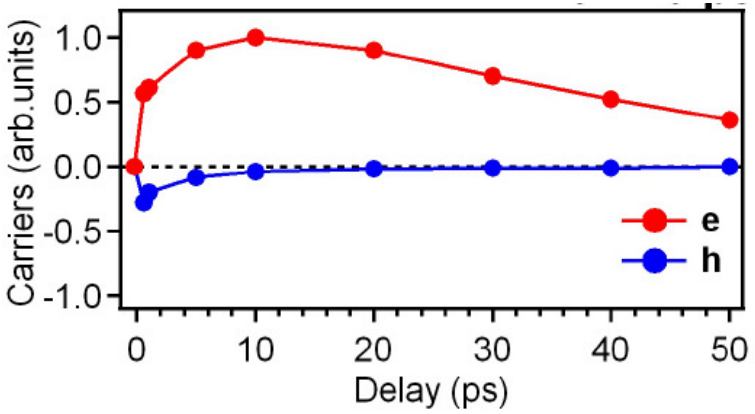


- Direct excitation to higher energy states B*
- Higher states scatters to B^*_p and S^*_p through interband scattering processes
- Thermalization and relaxation through interband and intraband scattering processes
- Slow relaxation of Dirac electrons (extremely weak electron-phonon coupling)
- No carriers can be detected at the bottom of the CB (B^*_p) for time delays > 10 ps

Time evolution of excess electron and hole populations







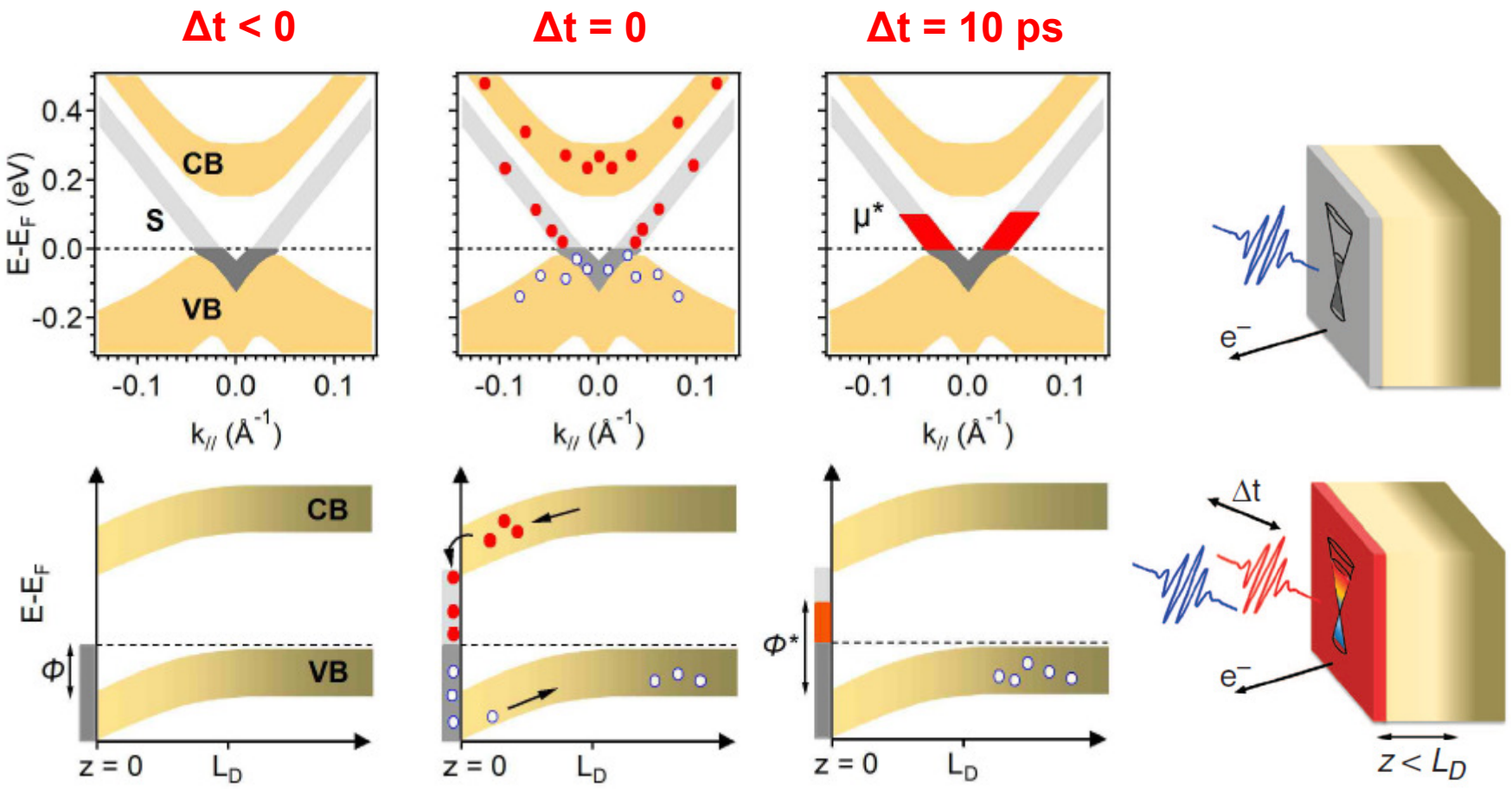
- Nearly flat-band system:
  - The electron and hole populations are well-balanced at all time delays

- System with downward band bending:
  - Asymmetry of transient excess carriers: hot electrons accumulate in the Dirac surface states

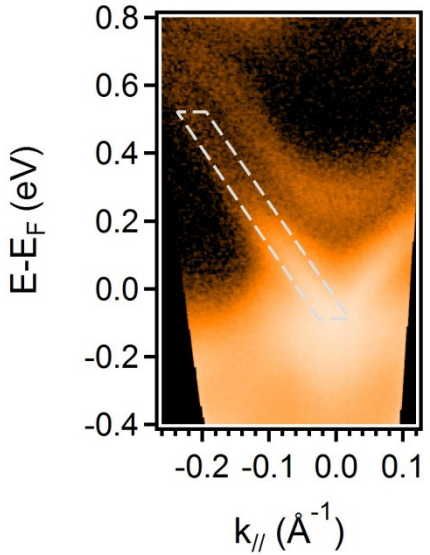
Hajlaoui *et al.*, Nature Comm (2013)

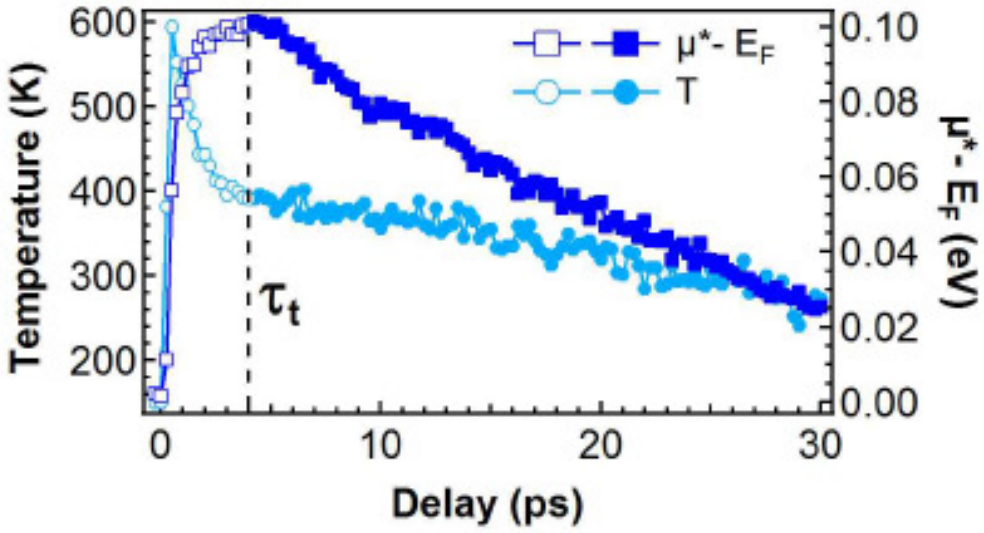
Formation of a transient hot metallic layer at the surface

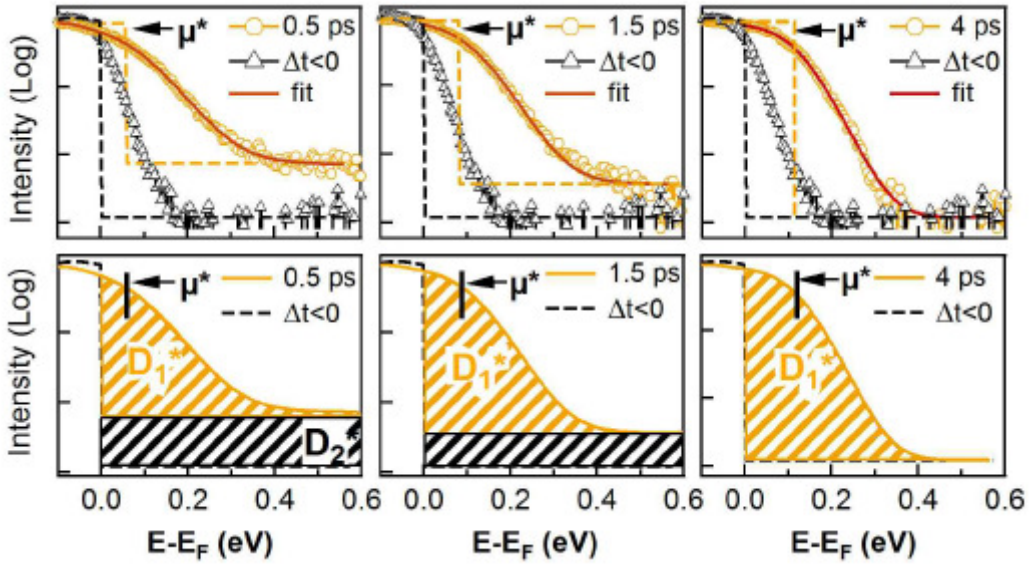
The lack of holes in the surface and subsurface region acts as the effective bottleneck for the recombination of excess Dirac electrons

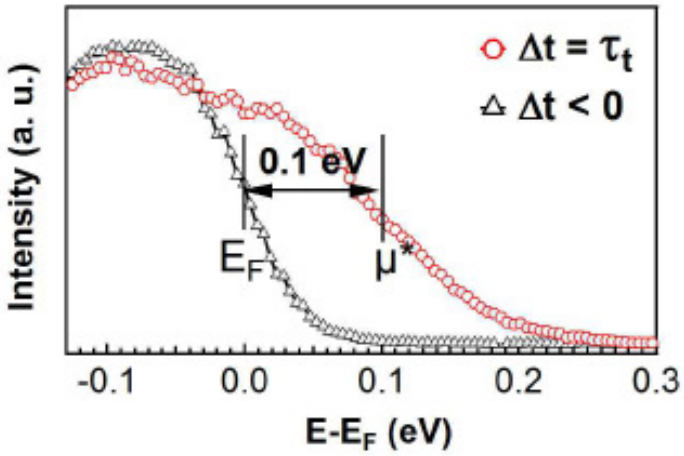


Transient temperature of Dirac fermions & chemical potential

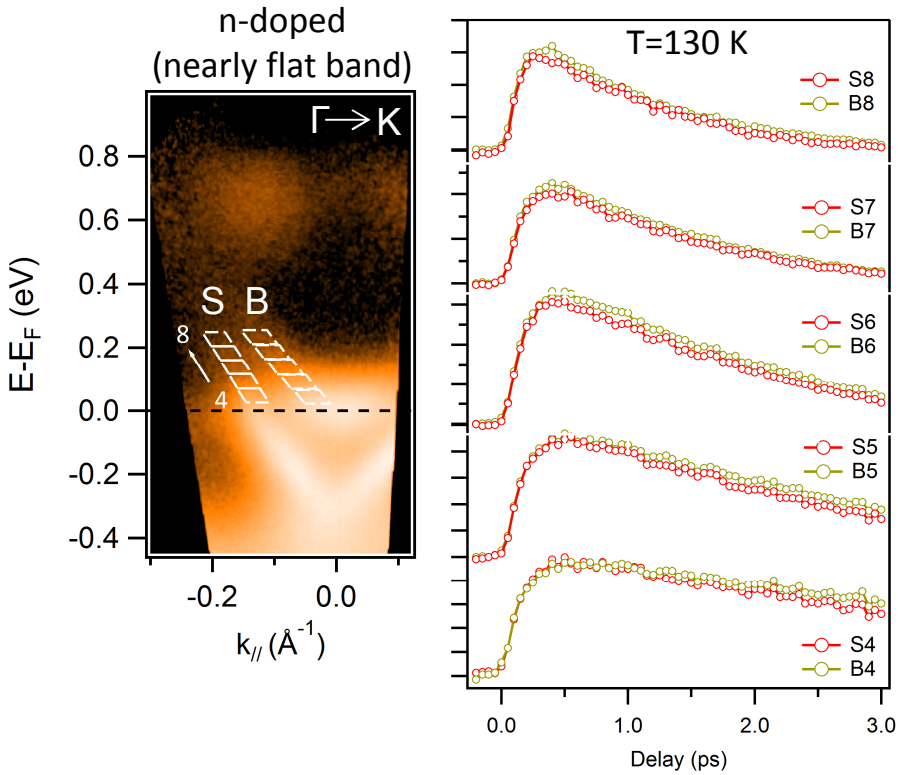


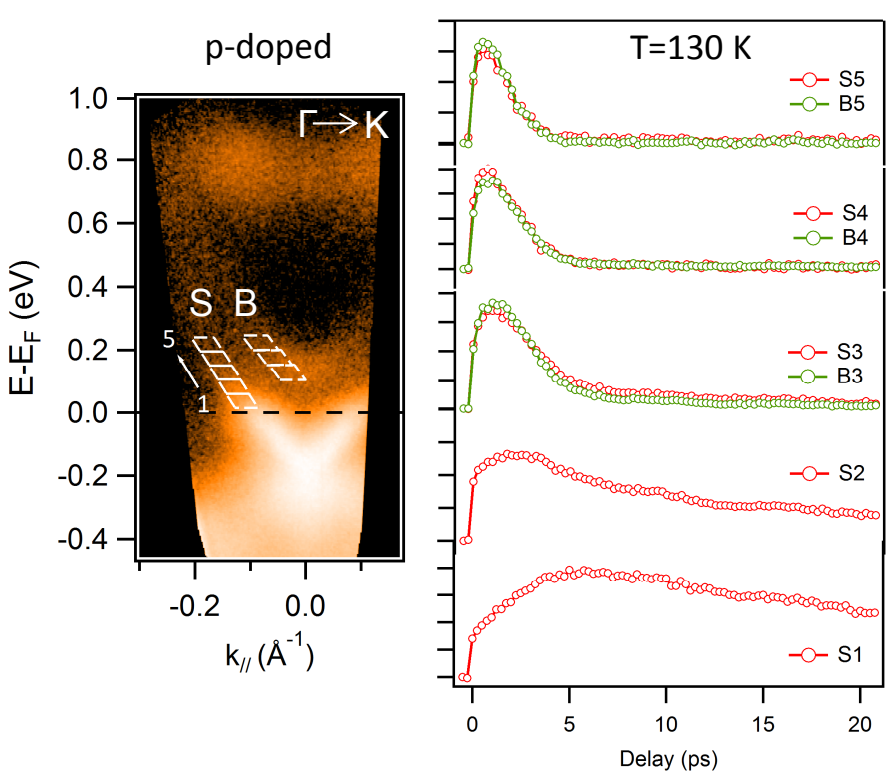






Surface charge carrier relaxation rate at different *k*-*E* windows

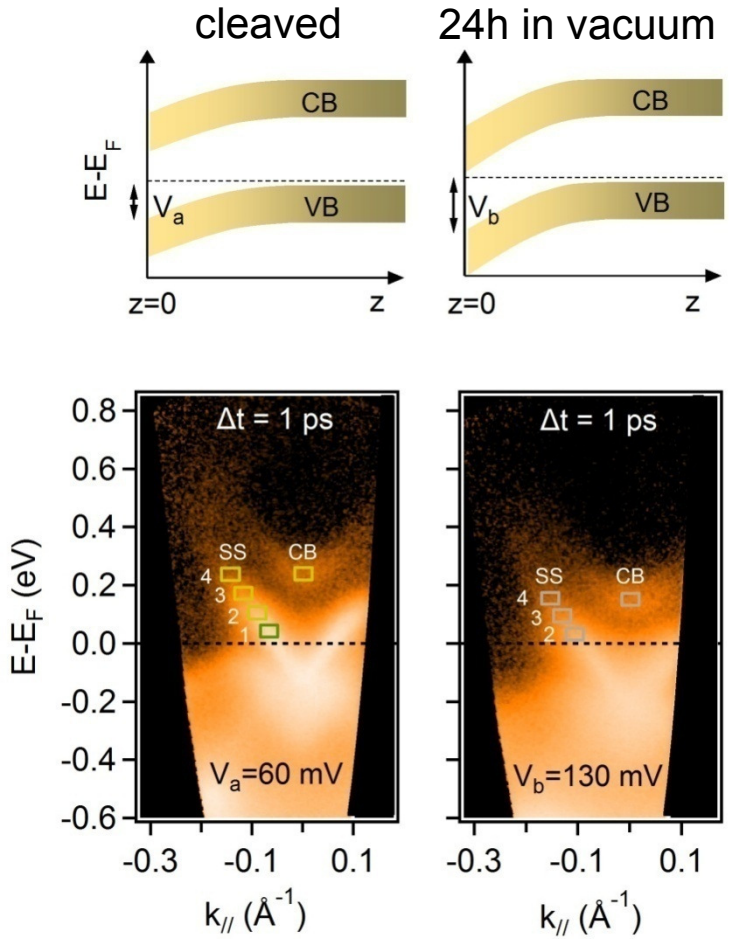


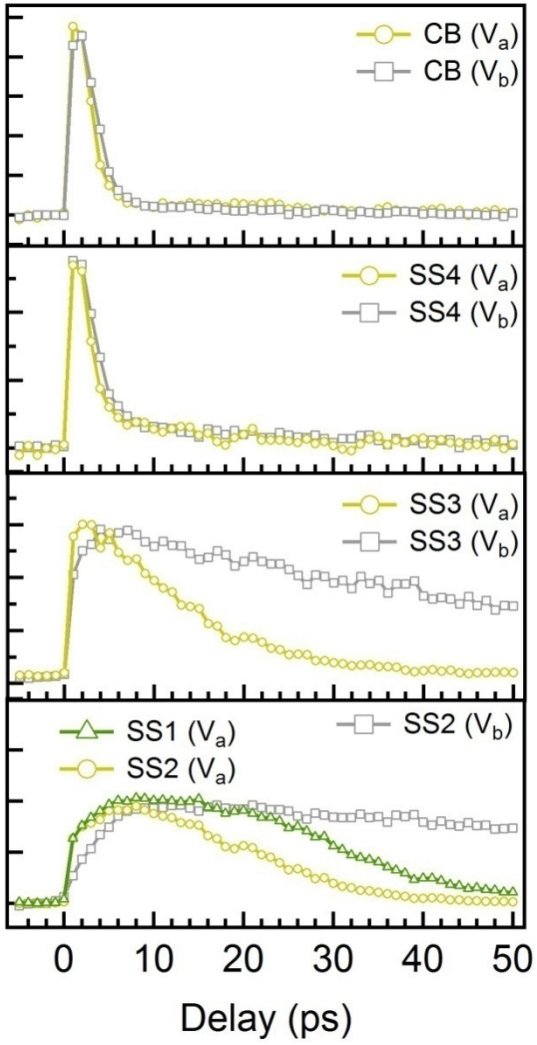


- Dirac and bulk conduction electrons appear to have similar decay rates
  - ⇒ Strong carrier exchange
  - ⇒ Fast decay channel for Dirac electrons through interband scattering with bulk conduction states

- Decay rates defer substantially for states below the conduction band edge
  - ⇒ Slow decay channels for electrons in Dirac states with binding energies below the conduction band edge (in the gap)
  - ⇒ Smaller available phase-space

The effect of band bending on decay rate in p-type $Bi_{2.2}Te_3$





- Strong band bending ⇒ substantial increase of Dirac electron's relaxation rate (>100 ps)
  - ⇒ Confinement of electrons at the surface
  - ⇒ Strongly out-of-equilibrium quantum states (unusual for a metallic state)

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Summary

- Using time-resolved ARPES, we provided a direct visualization of the excess carrier population and of its evolution.
- We found that the ultrafast dynamics of the carriers in the surface Dirac states is up to 0.5 ps; the bulk acts as a reservoir that keeps providing a relevant charge flow
- The subsequent relaxation phase (more than 10 ps), governed by charge diffusion and weak electron−phonon coupling, is less efficient than the typical bulk recombination.
- We show that one can effectively photo-induce a strongly out-of-equilibrium quantum state by acting on the charge balance between bulk and Dirac states

Thank you!