



**Training &
Experience Hub
Berlin**

Quantum Materials: from ARPES to trARPES



16/09/2026
online



PHOTONICS PUBLIC PRIVATE PARTNERSHIP

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Michele Puppin



**Time-resolved
ARPES
360 CARLA**



Time- angle-resolved photoelectron spectroscopy on quantum material

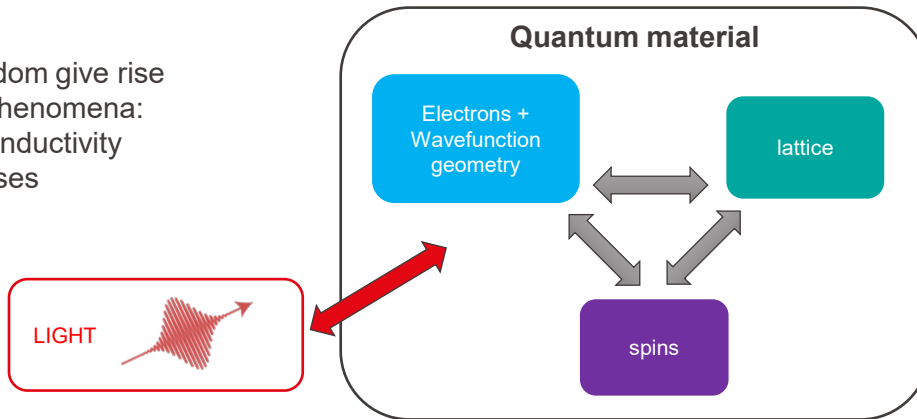
Quantum materials:

Entangled degrees of freedom give rise to macroscopic quantum phenomena:

- High T_c superconductivity
- Topological phases
- ..

ARPES

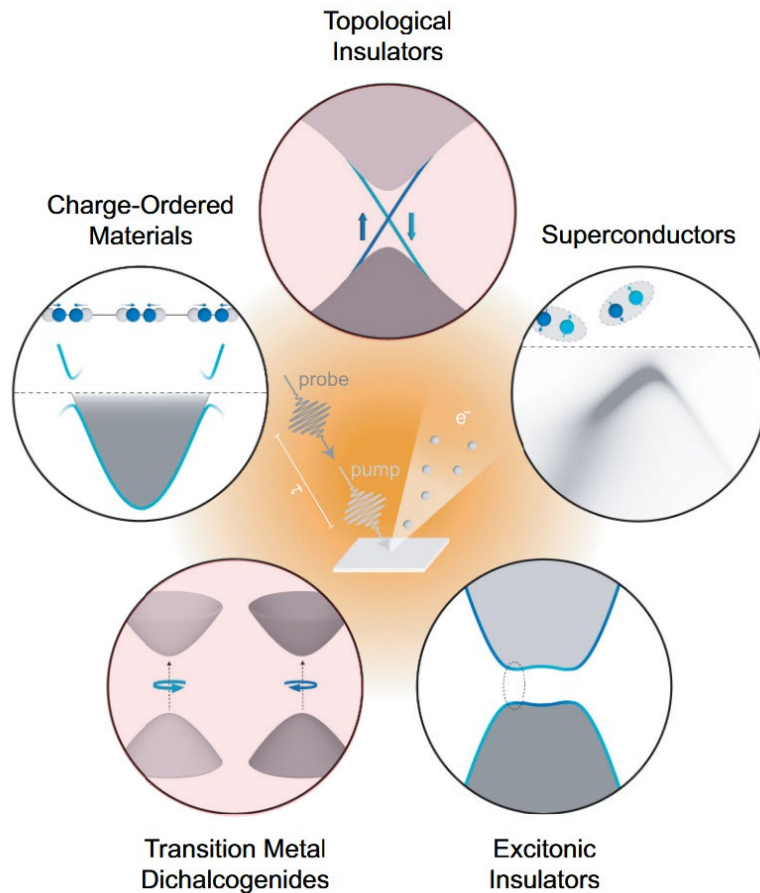
- Direct access to electronic structure
- Spectral function: Microscopic couplings
- Momentum resolution



How does light interact with quantum materials?
Can we use light to control quantum materials?

Time-resolved ARPES

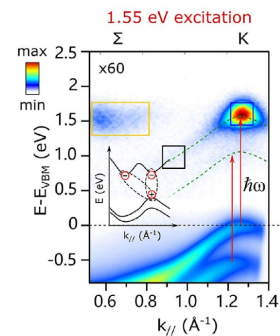
- Study light-matter interaction at its relevant microscopic time scales (femtosecond 10^{-15} fs)
- Use light to control quantum materials/generate novel phases



OUTLINE:

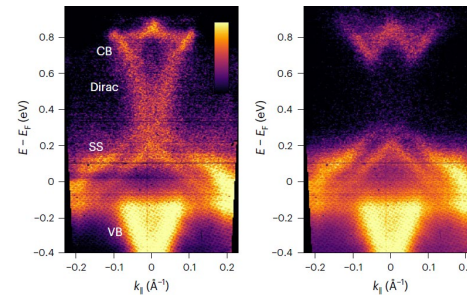
- trARPES basics
- Unoccupied band structure mapping
- Excitonic effects
- Light-driven quantum materials

Band structure and Excitons in TMDS

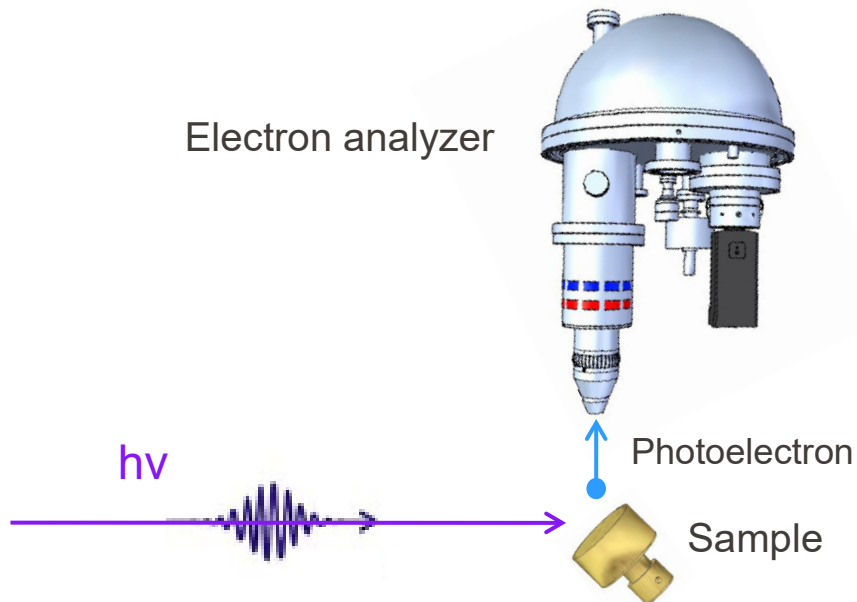


Dong*, Puppin* et al., Nat Sci. 2021;1:e10010

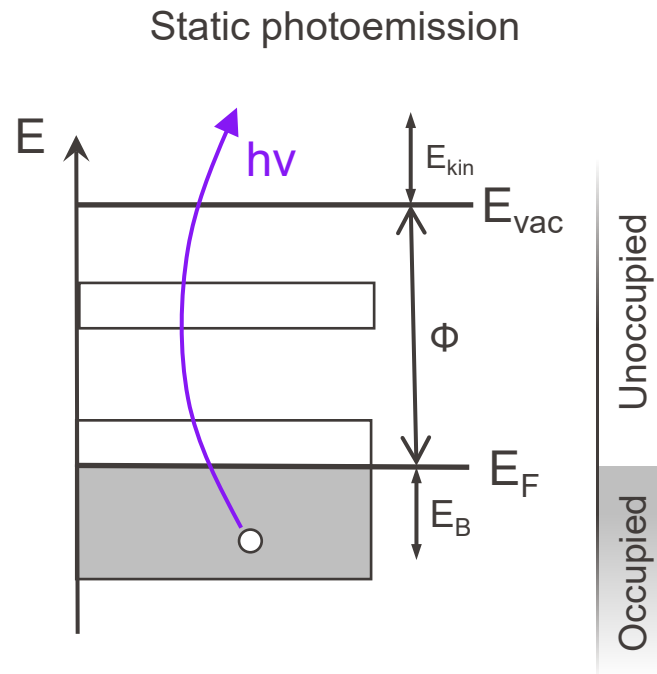
Floquet engineering in SnTe



Chassot, F; Nat. Phys. 2026, 22 (8), 1282–1286.

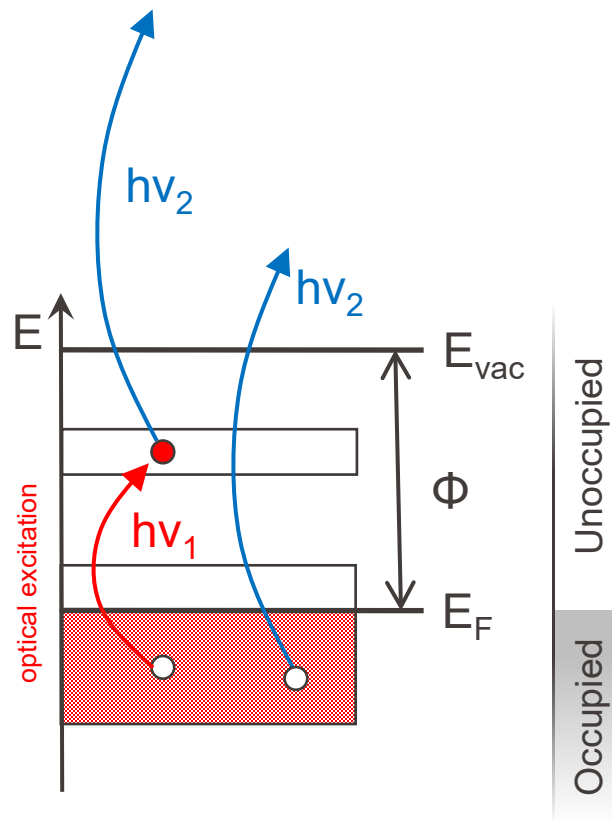
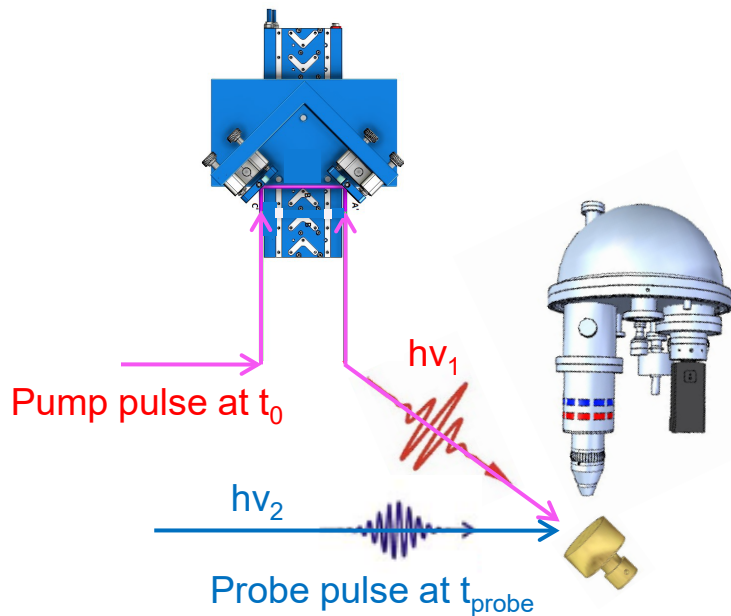


- Probe pulse: $h\nu > \phi$
- VUV photons $> 6 \text{ eV}$



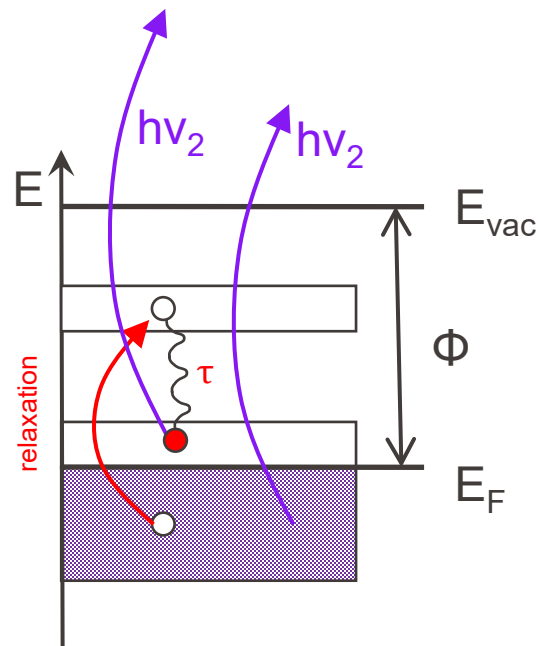
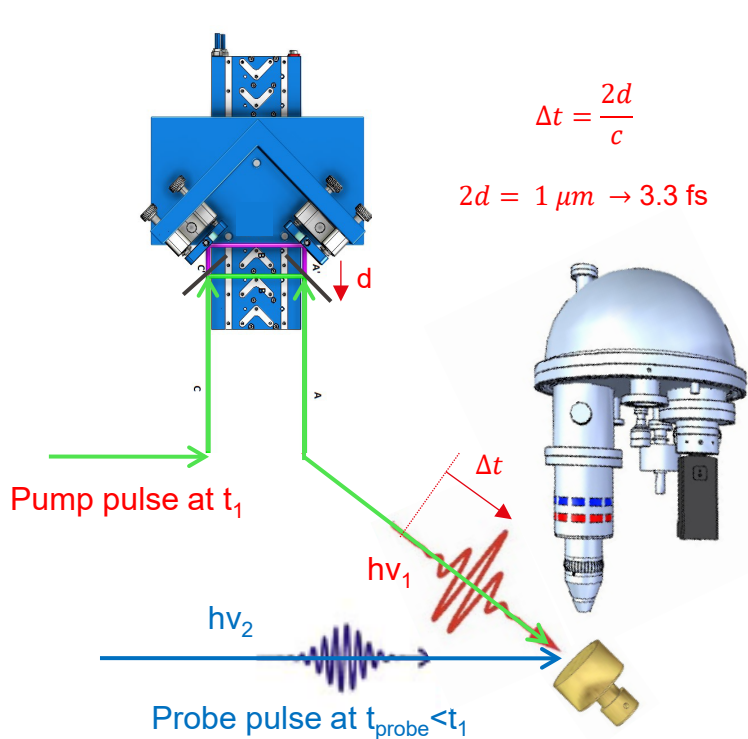
$$E_{\text{kin}} = h\nu - \phi - E_B,$$

- 2 x synchronized pulses: optical pump pulse excites the system at time t_0

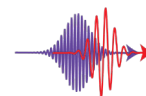


- Populates the unoccupied state that become observable

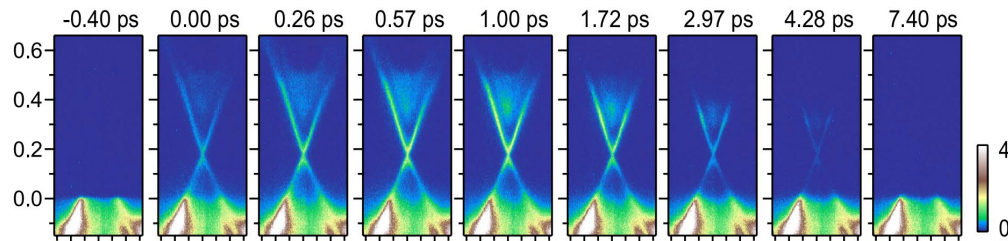
- Excited state lifetime τ

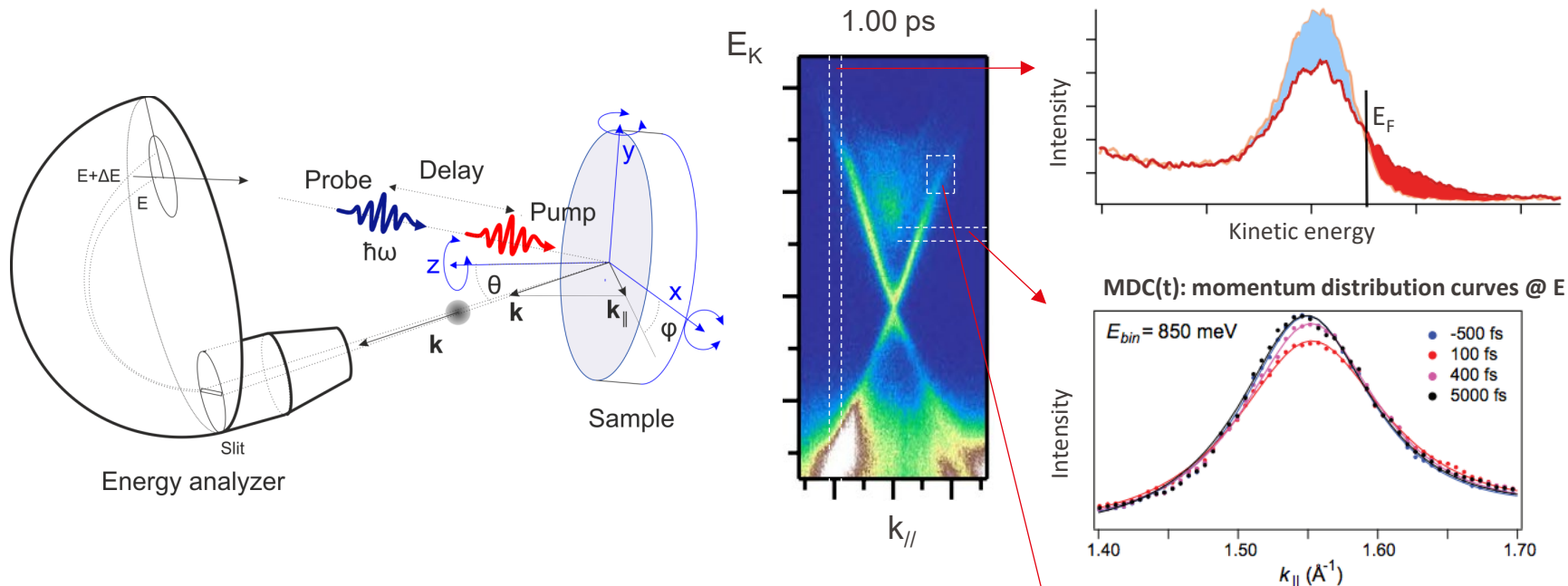


- Several „movie frames“ collected by changing Δt
- Time resolution pump-probe time convolution



- Energy resolution linked to probe duration (1825 meV fs)

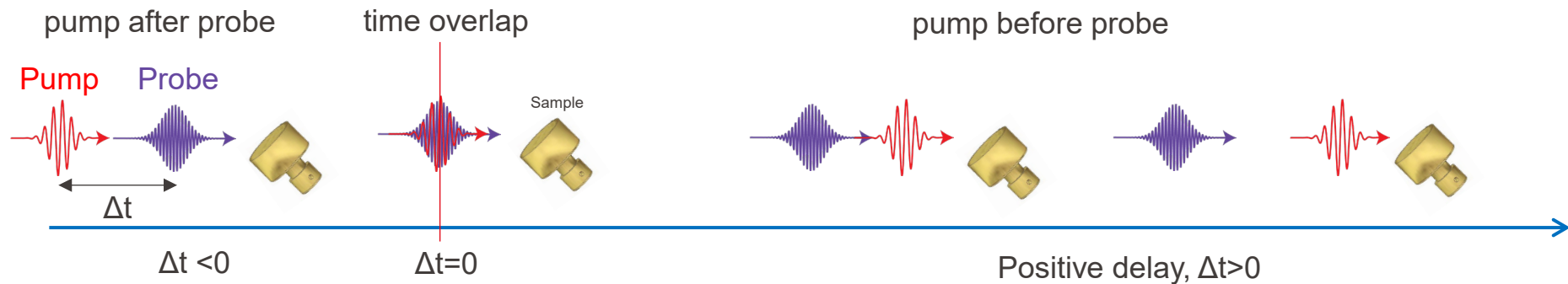




The electron spectrum is measured for a specific angle of emission θ

$$\hbar k_{||} = \sqrt{2mE_{\text{kin}}} \sin(\theta),$$

- Energetic photons ($\approx 10 \text{ eV}$) are needed to access the whole BZ
- Momentum specific dynamics



Equilibrium

Coherent phase

Non-thermal distribution

Hot electron relaxation

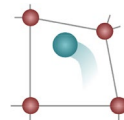
Microscopic scattering processes

fs

ps



Electron-electron scattering



Electron-phonon scattering

$\Delta t = 0$ Δt

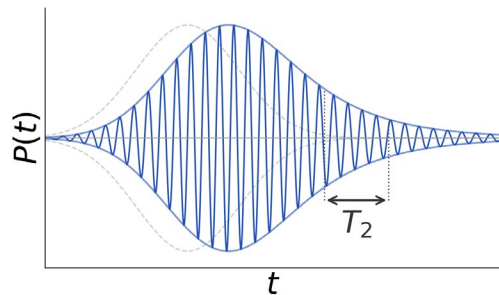
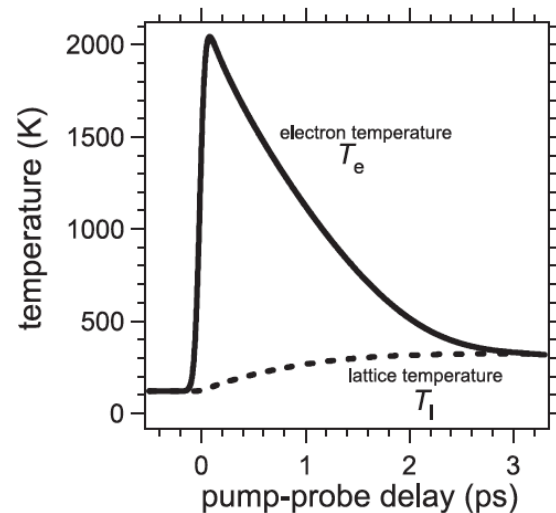
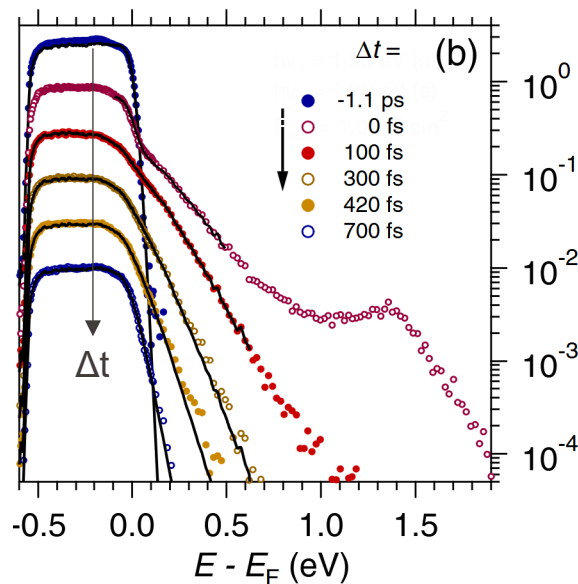
Equilibrium

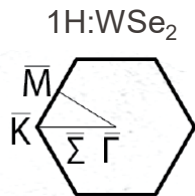
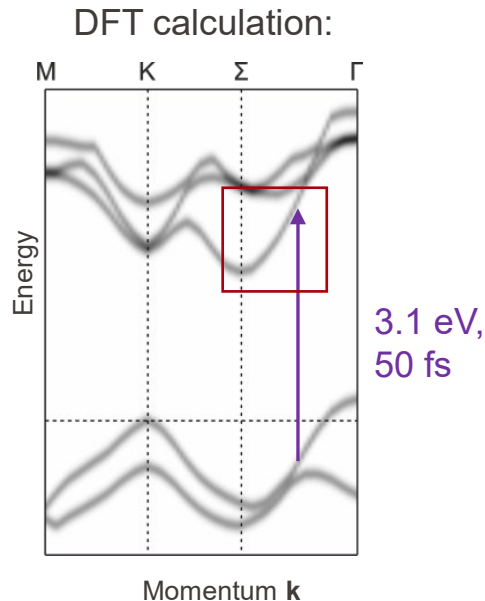
Coherent phase

Non-thermal distribution

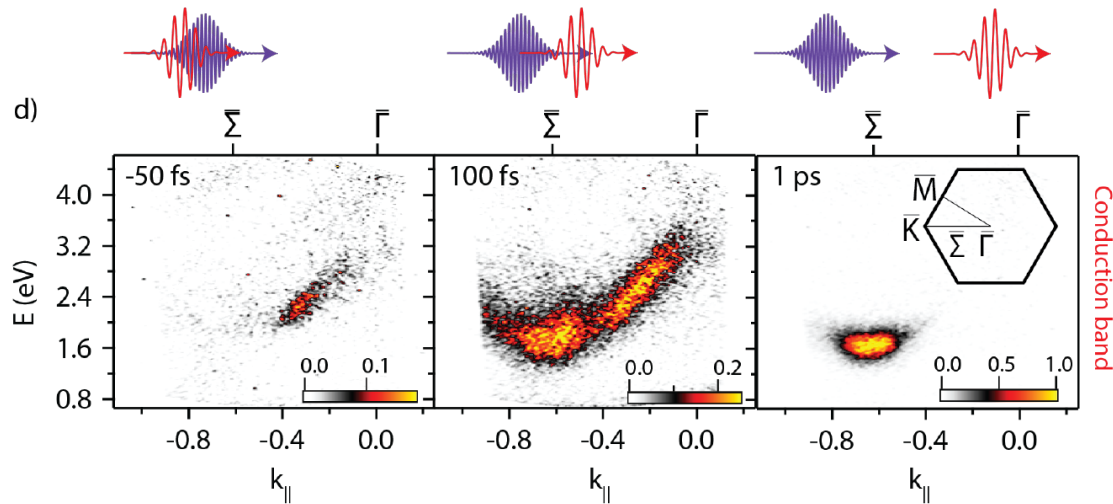
Hot electron relaxation

Dephasing (10s fs)

Thermalization
(10s-100 fs)Inelastic scattering with
phonons (100 fs – 10 ps)Polarization, coherent
with the driving E field



Transition metal dichalcogenide

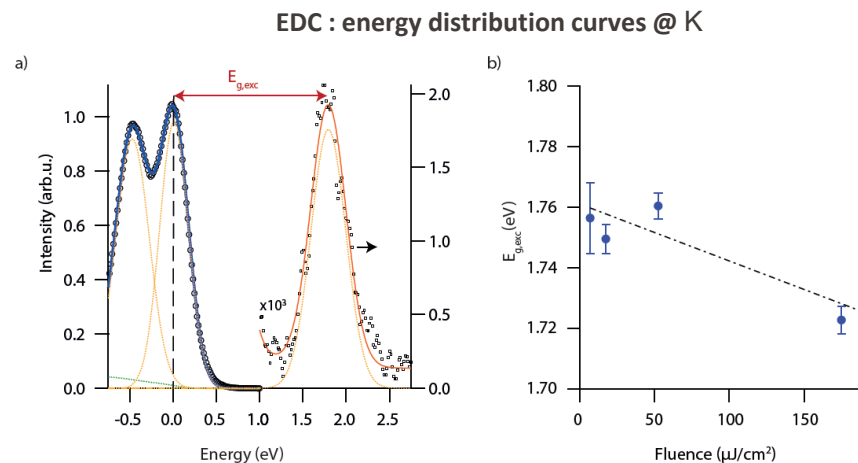
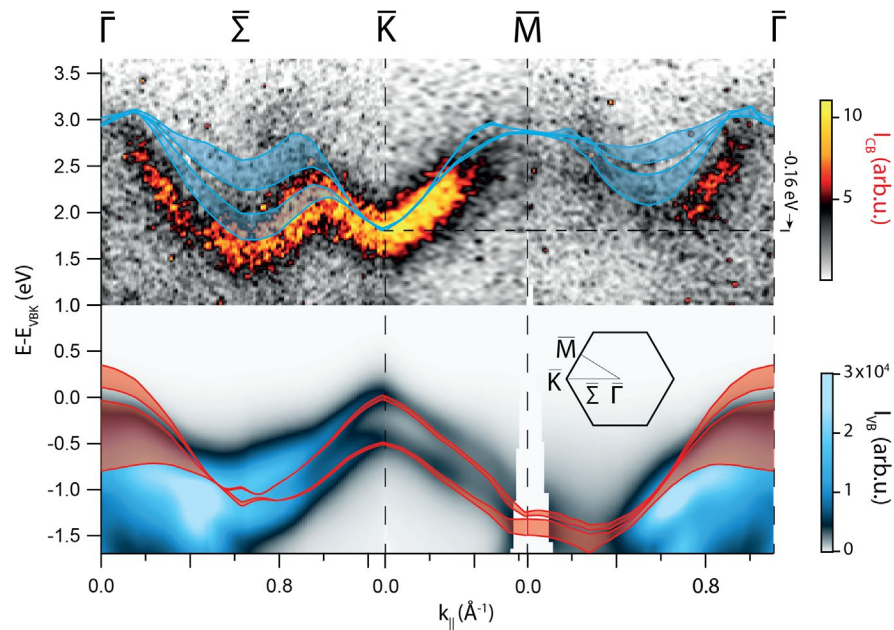


Non-thermal distribution

Hot electron relaxation

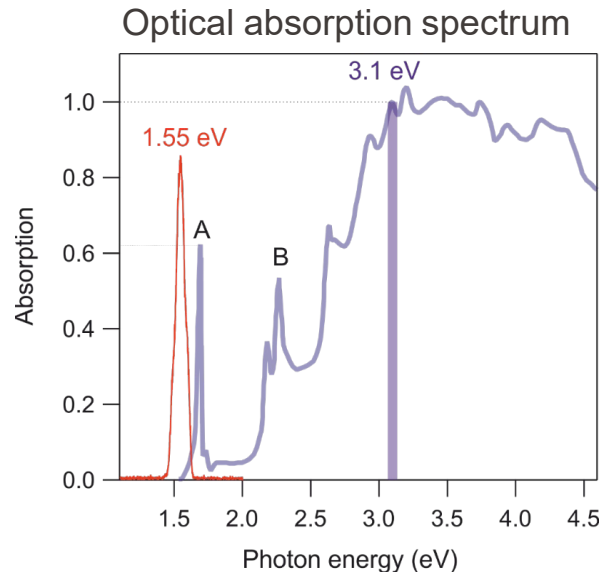
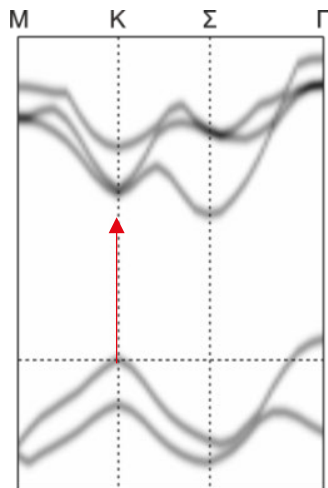
The nonthermal distribution of electrons relaxes (fast!) populating for short time a large portion of the conduction band.

trARPES with fs pulse can map conduction band

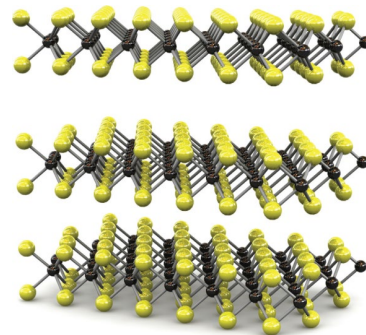


- Full bandgap visualization in a TMD semiconductor (100 fs)
- Comparison with theory (G_0W_0)

- Bandgap renormalization due to many-body effects: low excitation limit 1.76 eV

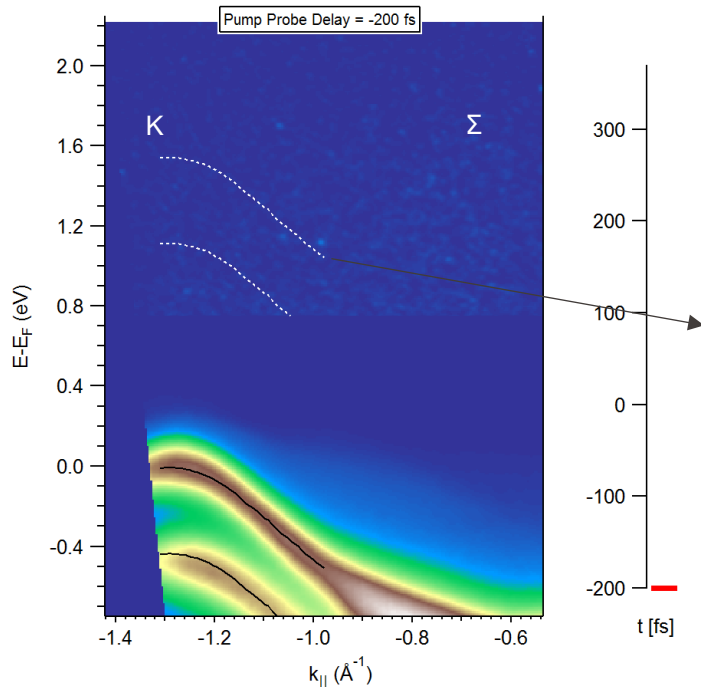
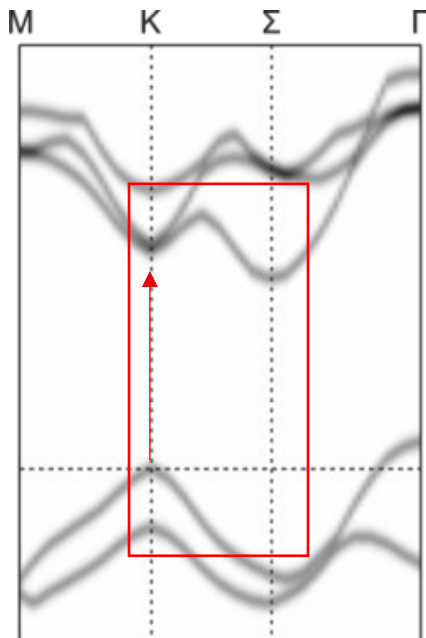


2d layered materials



TMDs are 2d layered semiconductors:

- Weakly screened electron-hole Coulomb interaction
- Strong excitonic resonances (even in bulk 1H:WSe₂)



Unoccupied states dynamics:

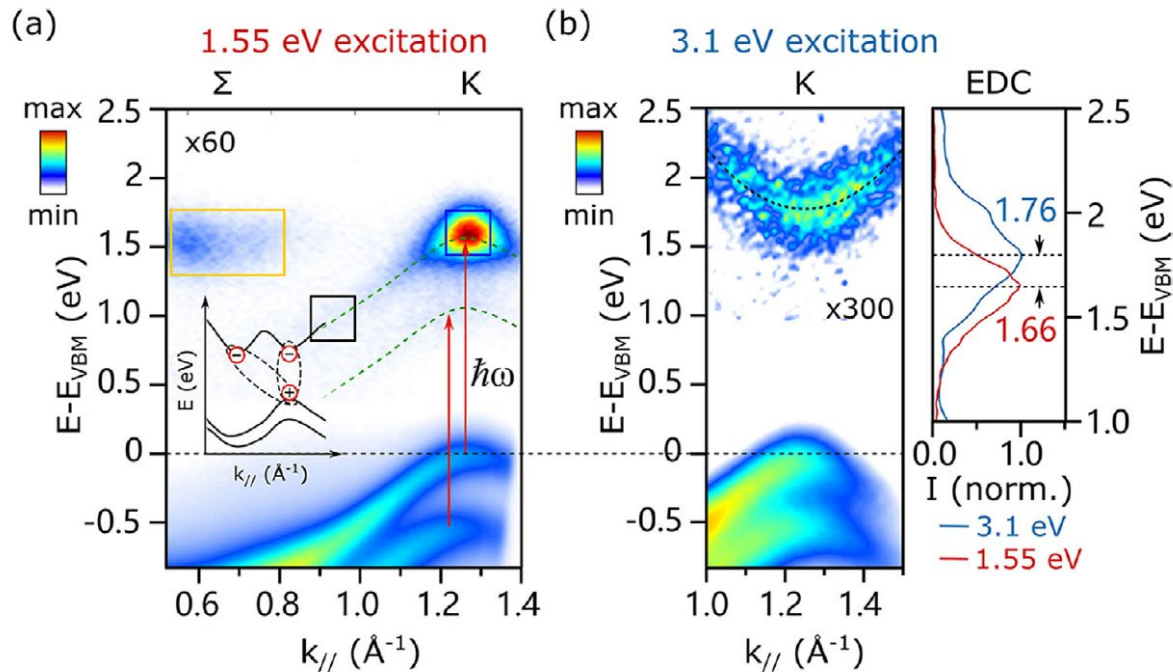
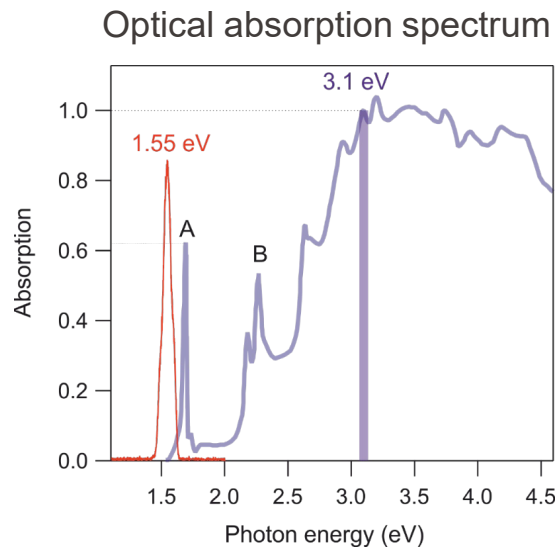
- Initial signal at K
- Ultrafast scattering at sigma

In-gap states:

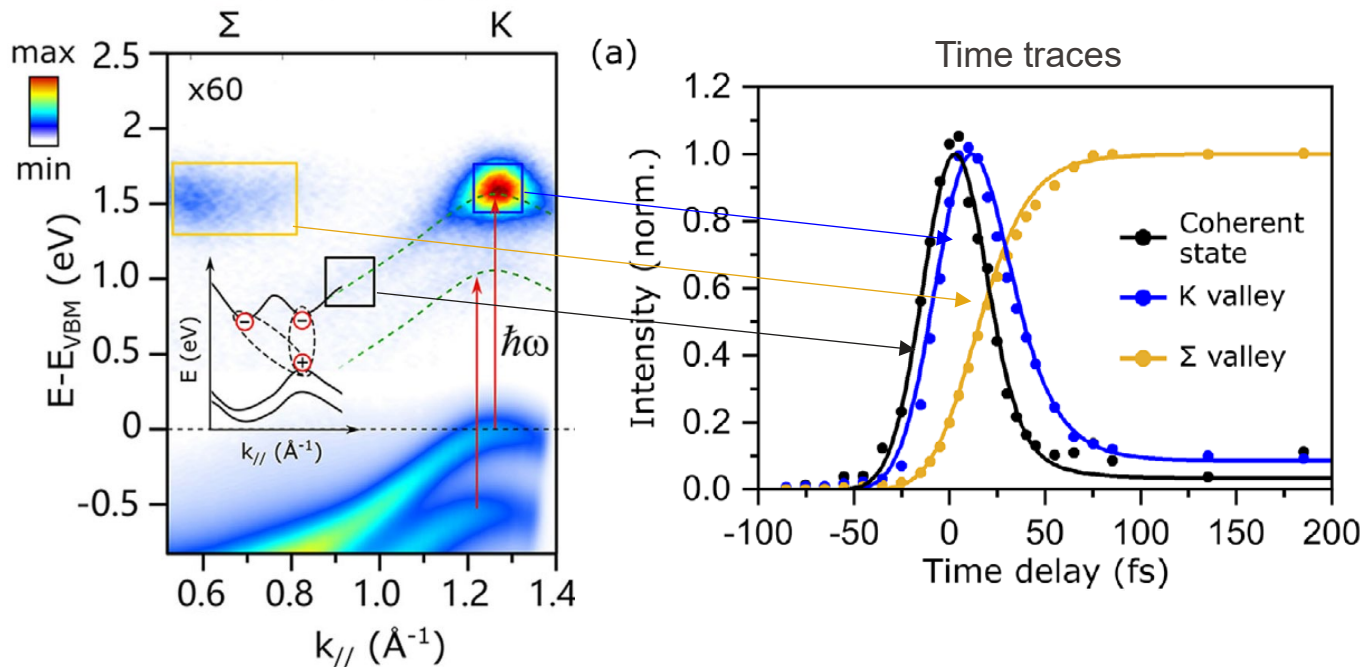
- Replicas of the VB: appear only during temporal overlap:

Coherent phase

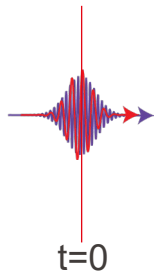
- Pump: 800 nm (1.55 eV), 30 fs
- Excitation (nearly) resonant with the exciton peak



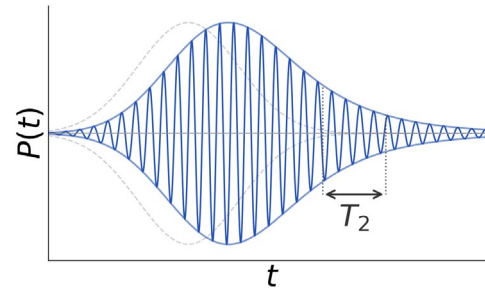
- Energy difference in the excited state spectrum
- Excitonic resonance at K: k distribution and dispersion



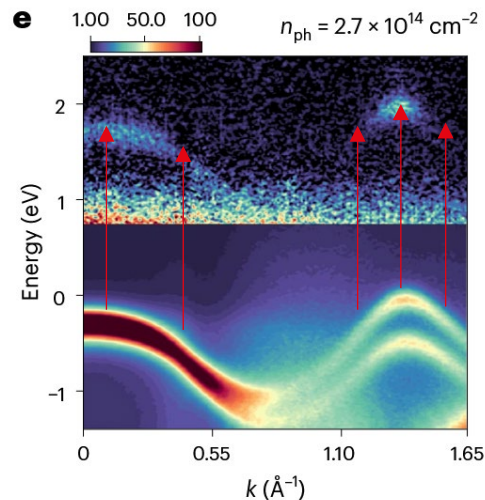
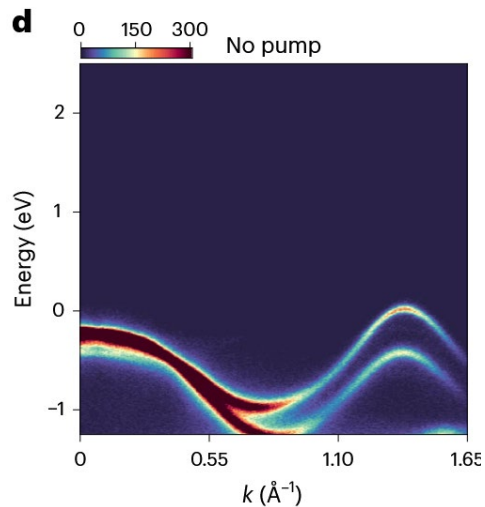
- Multilevel fit of coupled optical-Bloch equations:
 - “Bright” K exciton population decay time to “dark” states $T_1 = 18 \pm 4$ fs (electron-phonon scattering)
 - Band replicas signature of coherent light matter interaction
 - Dephasing time from model fitting $T_2 = 17 \pm 9$ fs



Coherent phase



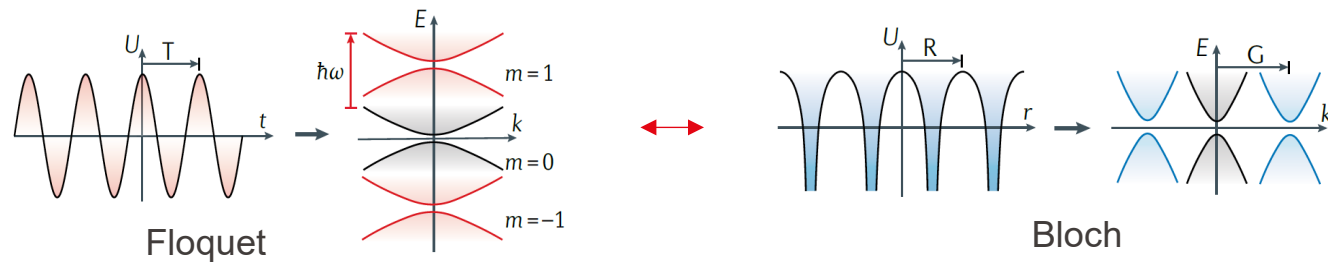
- Replica states signal the presence of coherent light-matter interaction, particularly evident under strong fields
- Minimize dephasing (due to scattering), excitation close or below the band gap.



Pareek, V. *Nat. Phys.*
2026, 22 (2), 209–217

Coherent replicas in
 WS_2 monolayer

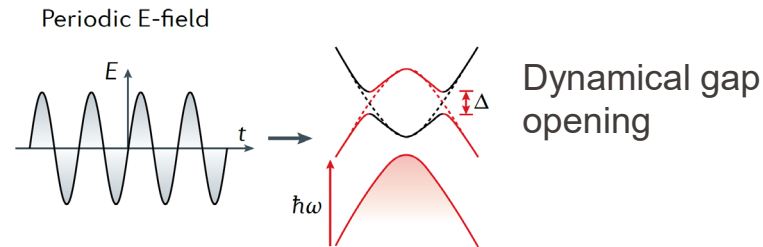
NEW PHENOMENA CAN BE OBSERVED IN QUANTUM MATERIAL IN THE COHERENT REGIME!

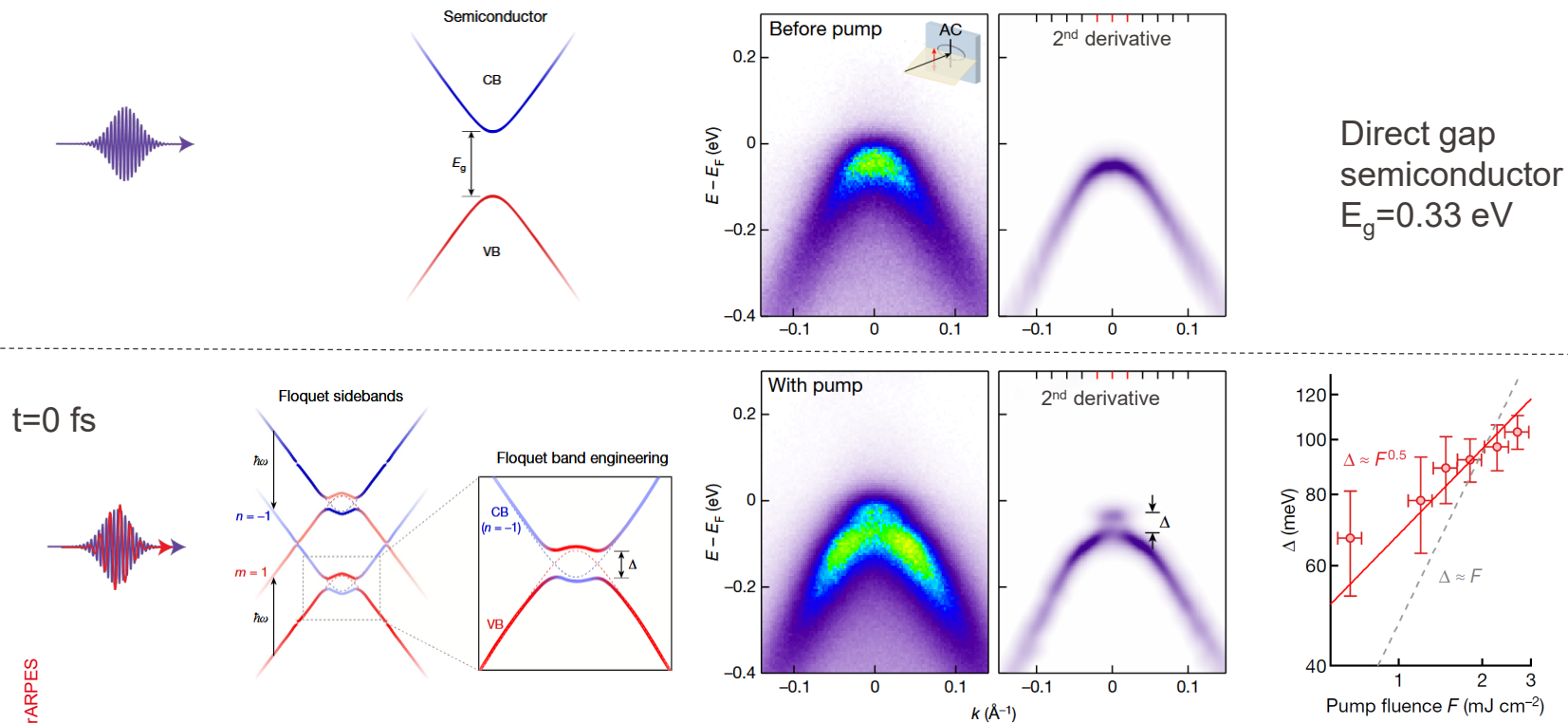


- Treat pump electric field as a time periodic term
- Floquet theorem for time-periodic Hamiltonian: time-periodic function that can be expressed on a basis of sidebands at energy $m\hbar\omega$ (photon-dressed states)

FLOQUET ENGINEERING:

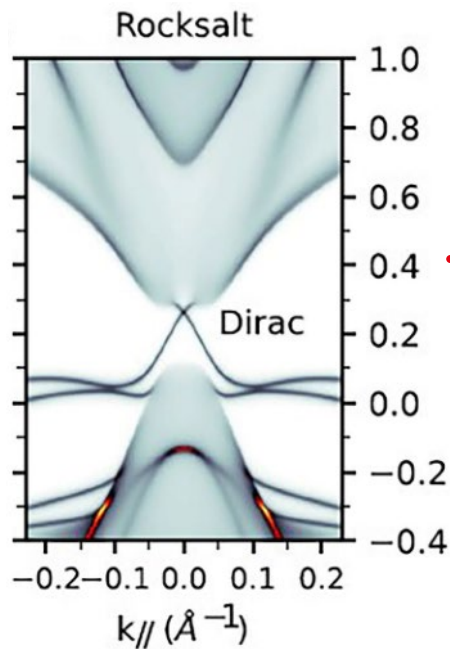
Replicas of the bands (Floquet-Bloch) may overlap and interact with the original band structure: hybridization, gap opening, new topological phases



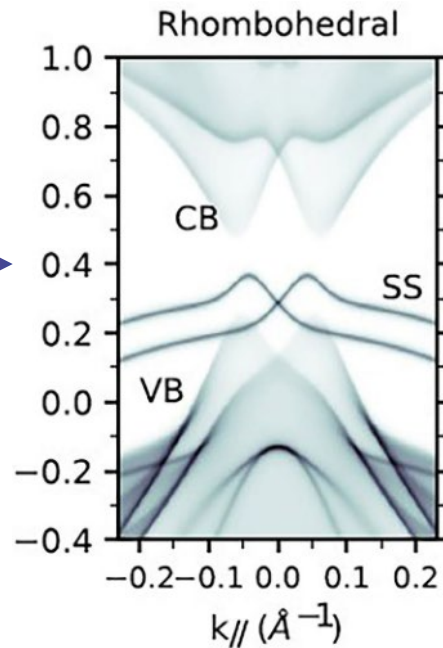


Band structure and electronic properties transiently and reversibly modified on fs time scale
 Challenge: requires **intense** (10^6 - 10^7 V/m) **MIR sources**, combined with **good energy resolution**

High T

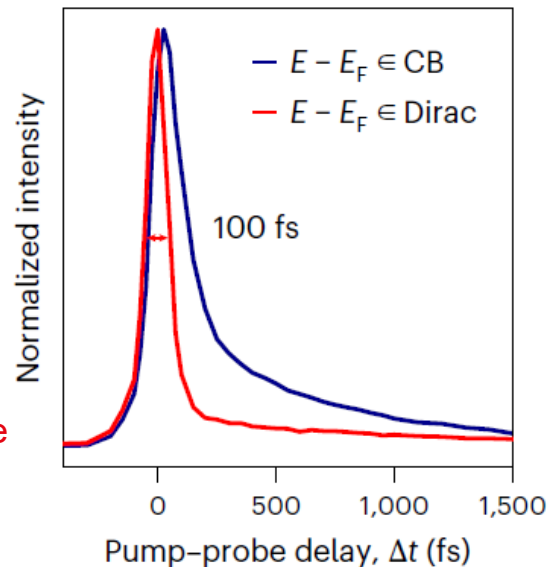
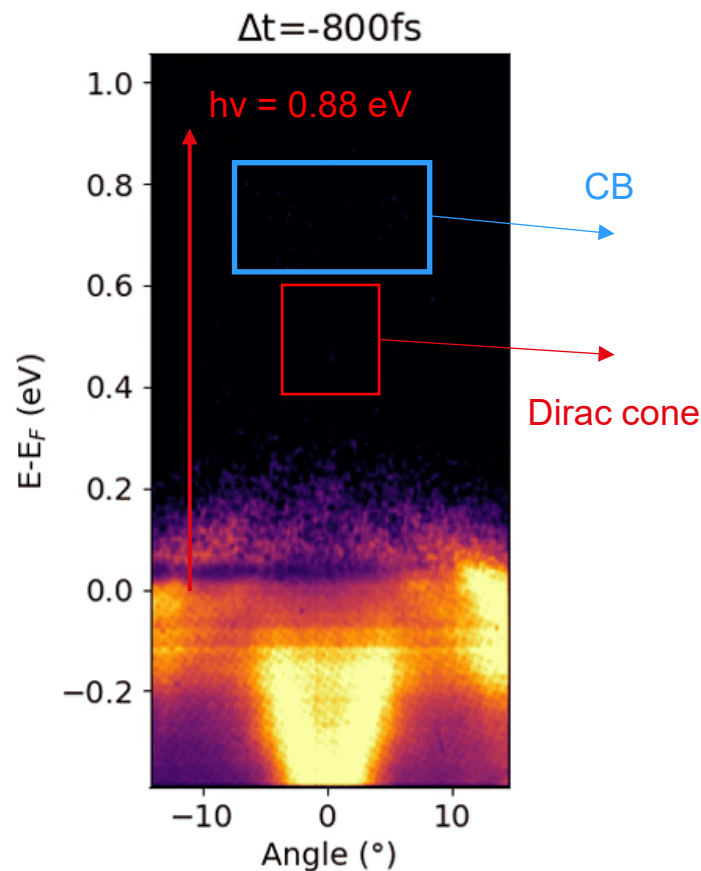


Low T

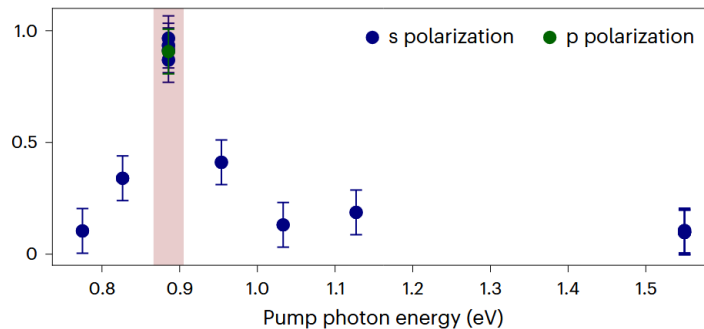


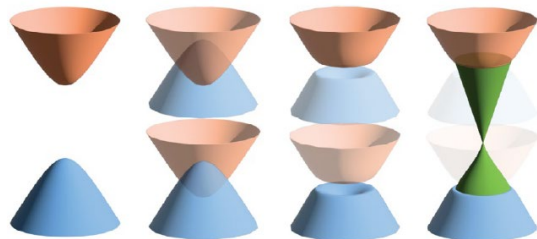
- Topological crystalline insulator
- Ferroelectric polar structure
- Topologically trivial surface states

trARPES experiment in the low T phase (no Dirac cone) at gap resonance.

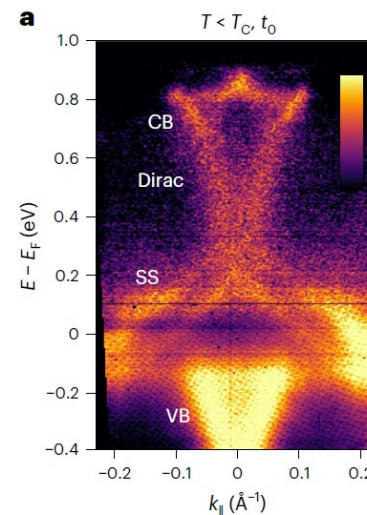
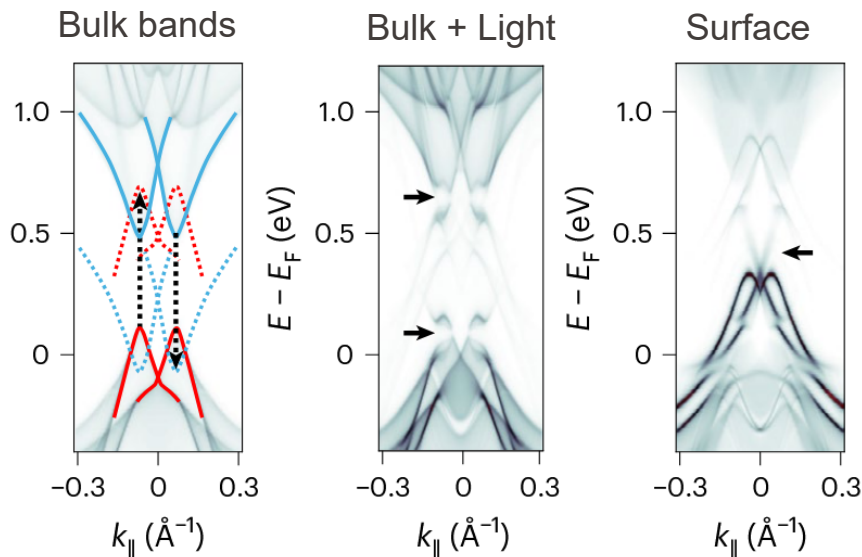


- Resonant at 0.88 eV + polarization independent
- Cannot be explained as replica band of the underlying states





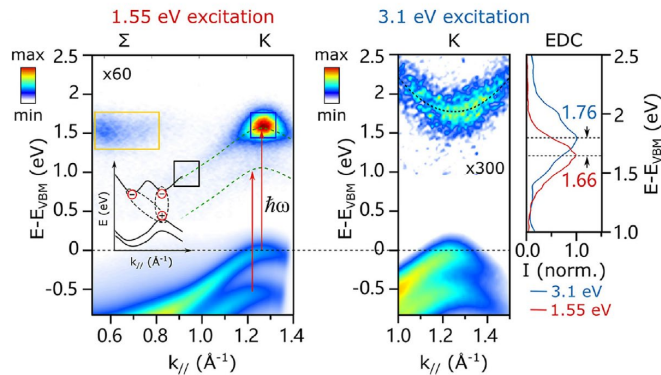
- Floquet driving induces VB-CB state mixing
- Band inversion drives a topological phase transition
- A dirac cone appears connecting driven VB and CB while the field is applied



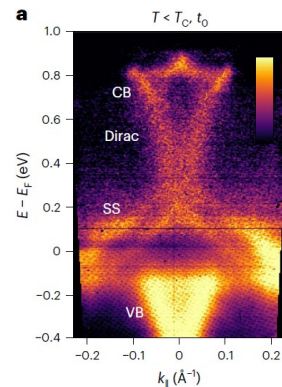
Light driven quantum material

SUMMARY:

- trARPES on TMD semiconductors:
 - Unoccupied band structure mapping
 - Excitonic effects and electron dynamics
- Light-driven quantum materials
 - Floquet sidebands
 - Light induced topological phase



Dong, S. *Natural Sciences* **2021**, 1 (1), e10010



Chassot, F.; *Nat. Phys.* **2026**, 22 (8), 1282–1286.



THANKS FOR YOUR ATTENTION !

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- (10) Dong, S.; Puppin, M.; Pincelli, T.; Beaulieu, S.; Christiansen, D.; Hübener, H.; Nicholson, C. W.; Xian, R. P.; Dendzik, M.; Deng, Y.; Windsor, Y. W.; Selig, M.; Malic, E.; Rubio, A.; Knorr, A.; Wolf, M.; Rettig, L.; Ernstorfer, R. Direct Measurement of Key Exciton Properties: Energy, Dynamics, and Spatial Distribution of the Wave Function. *Natural Sciences* **2021**, 1 (1), e10010. <https://doi.org/10.1002/ntls.10010>
- (11) Mahmood, F.; Chan, C.-K.; Alpichshev, Z.; Gardner, D.; Lee, Y.; Lee, P. A.; Gedik, N. Selective Scattering between Floquet–Bloch and Volkov States in a Topological Insulator. *Nature Phys* **2016**, 12 (4), 306–310. <https://doi.org/10.1038/nphys3609>
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