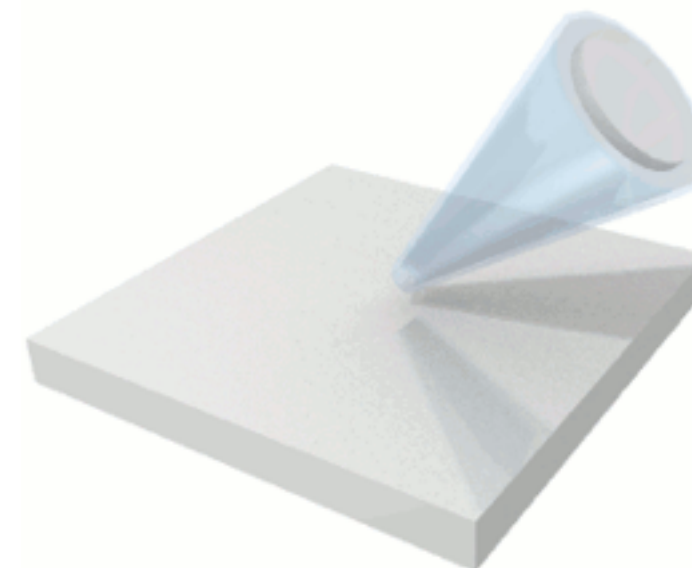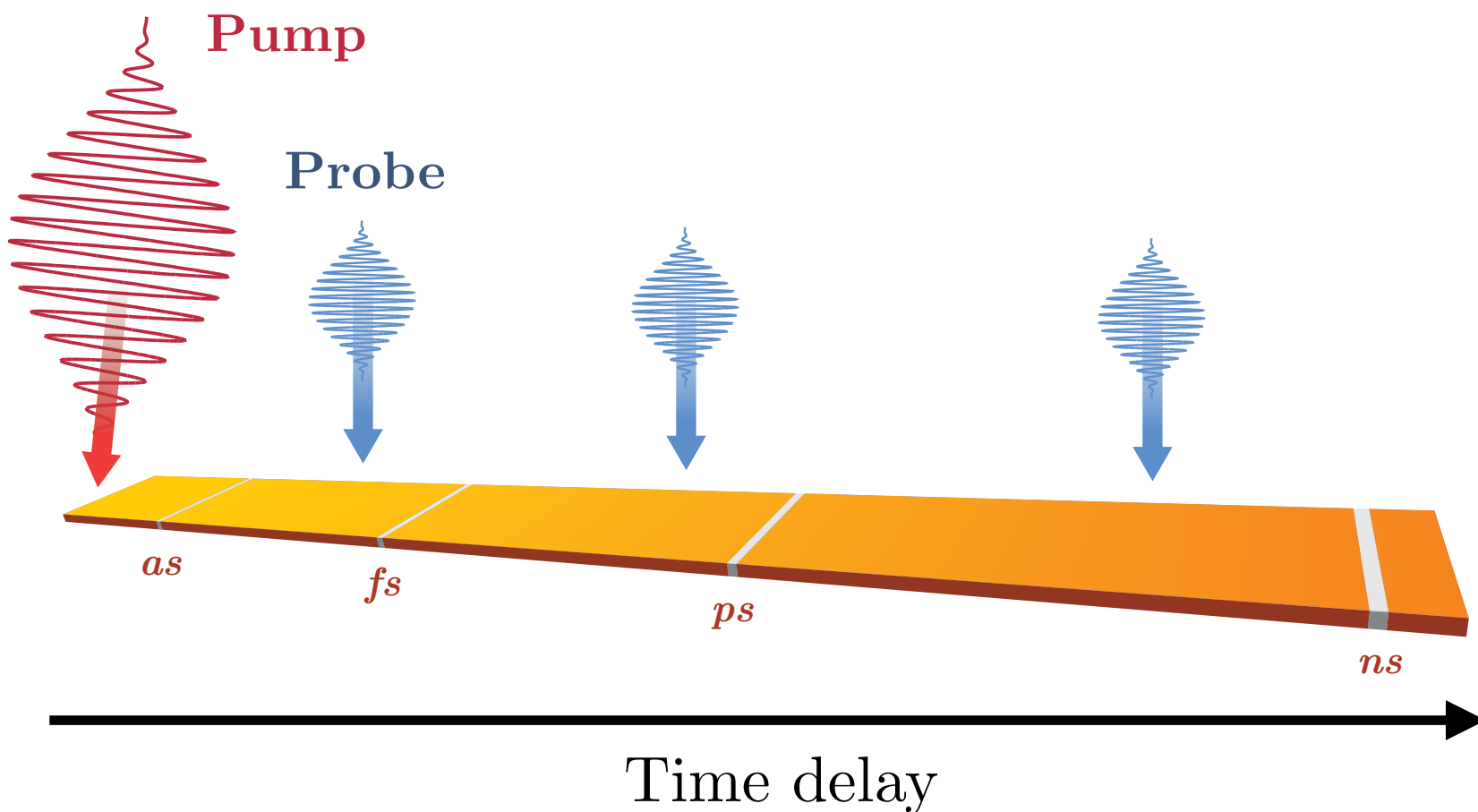# Ab-initio spin and time-resolved ARPES in real materials with TDDFT:
driving TMDs out of equilibrium

Umberto De Giovannini

University of the Basque Country,
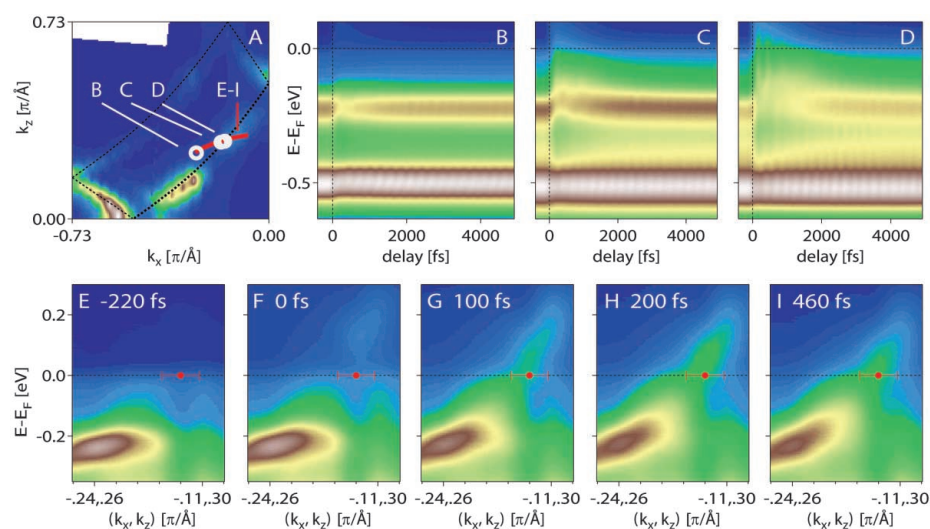San Sebastian, Spain

umberto.degiovannini@ehu.es

Motivations: trARPES I

Pump

Probe

as   fs   ps   ns

Time delay

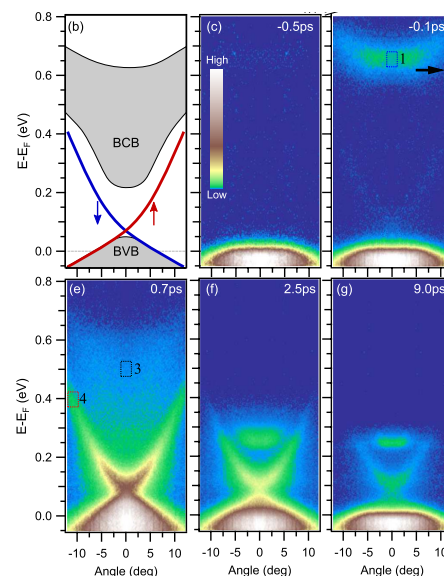Credits: Prof. Zhi-Xun Shen group webpage
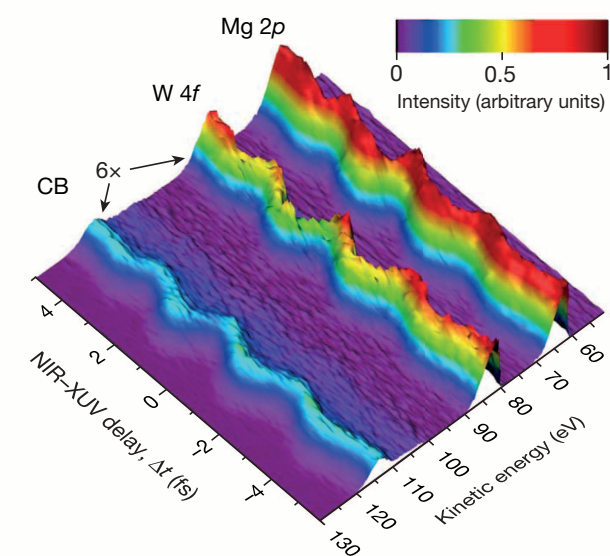
Examples:

Charge density wave melting

F. Schmitt, et al. Science 321, 1649 (2008).

Optical excitation in time

J. A. Sobota, et at. Phys. Rev. Lett. **108**, 117403 (2012).
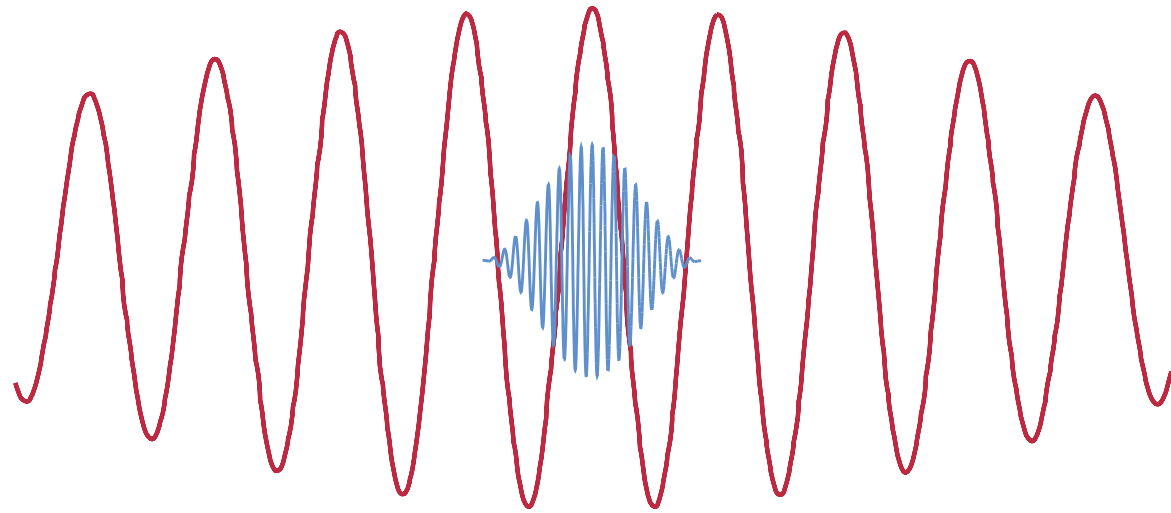
Dielectric screening in time

S. Neppl, et al. Nature **517**, 342 (2015).

Motivations: trARPES II

Pump   **Probe**
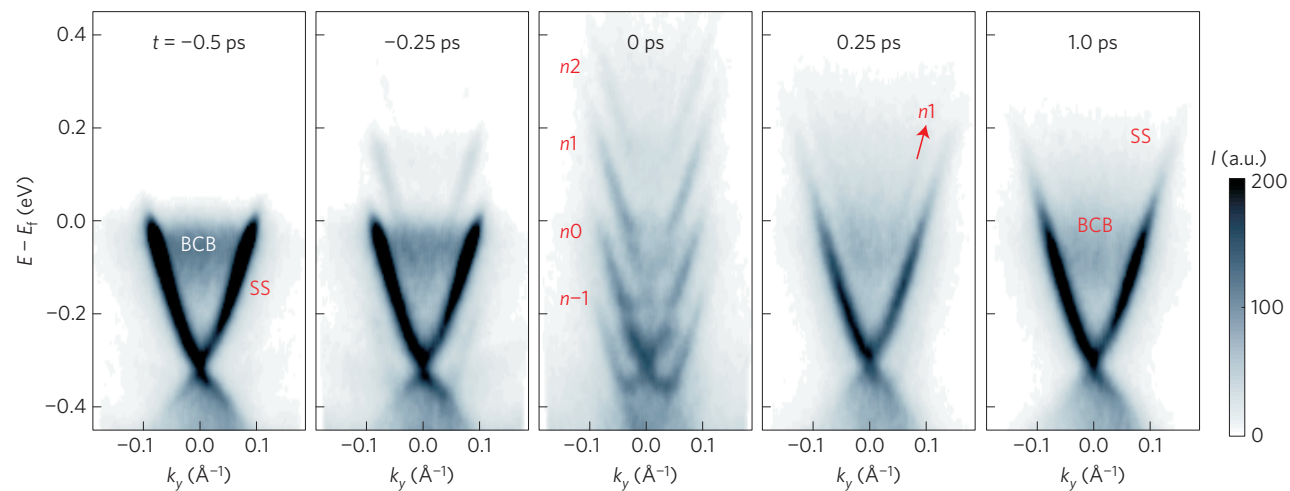
We are probing stationary states in a periodically driven system

New quasiparticle states: photon-dressed electronic states. Can be described with Floquet theory

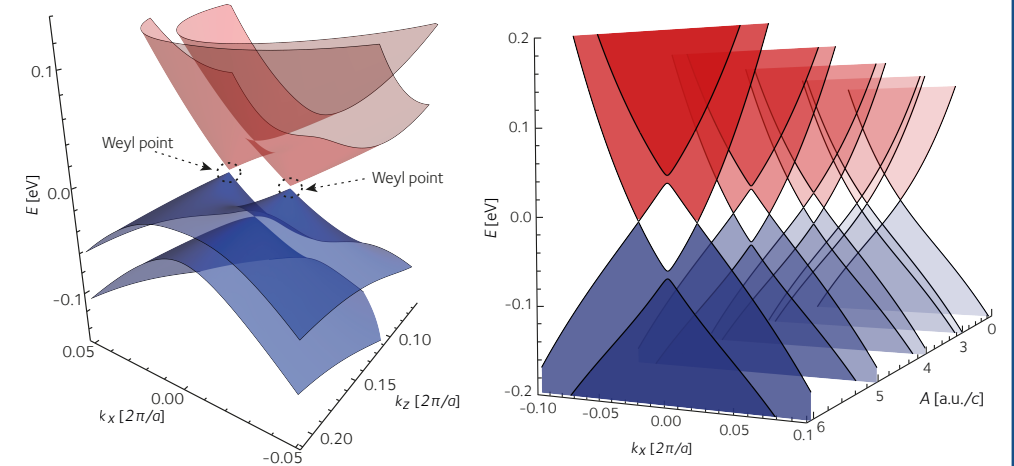## Examples:

Floquet sidebands

F. Mahmood, et al. Nat Phys **12**, 306 (2016)

Floquet Weyl semimetal

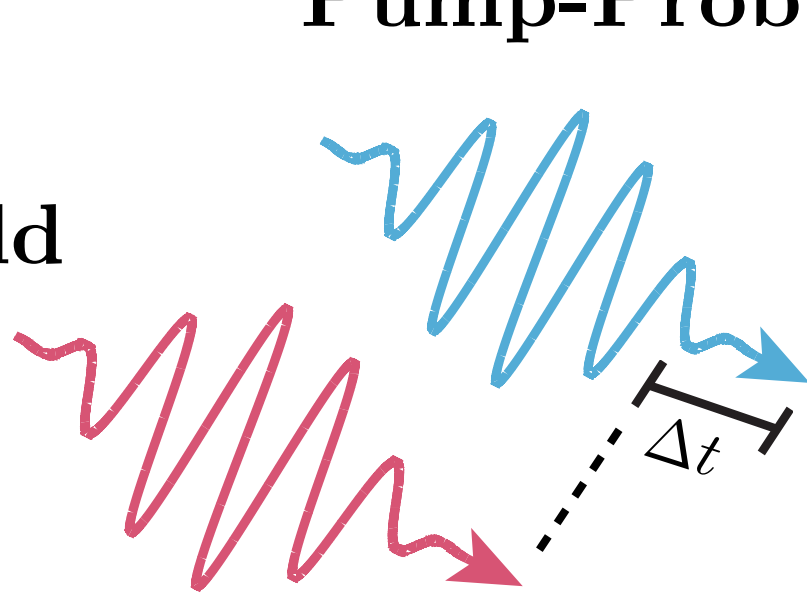H. Hübener, et al. arXiv:1604.03399 (2016)

Outline

- How to simulate trARPES with TDDFT and tSURFFP

- Application of trARPES on WSe2:

  ★ ARPES and spin-ARPES

  ★ Dichroic spin-valley resonant excitation

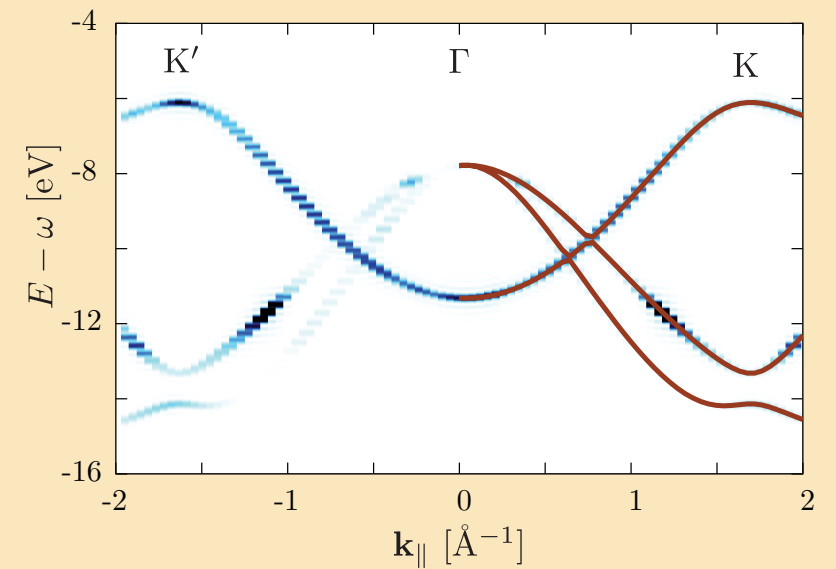 ★ Photon dressing and dressed-band hybridization

ARPES in a nutshell

Laser field

Pump-Probe

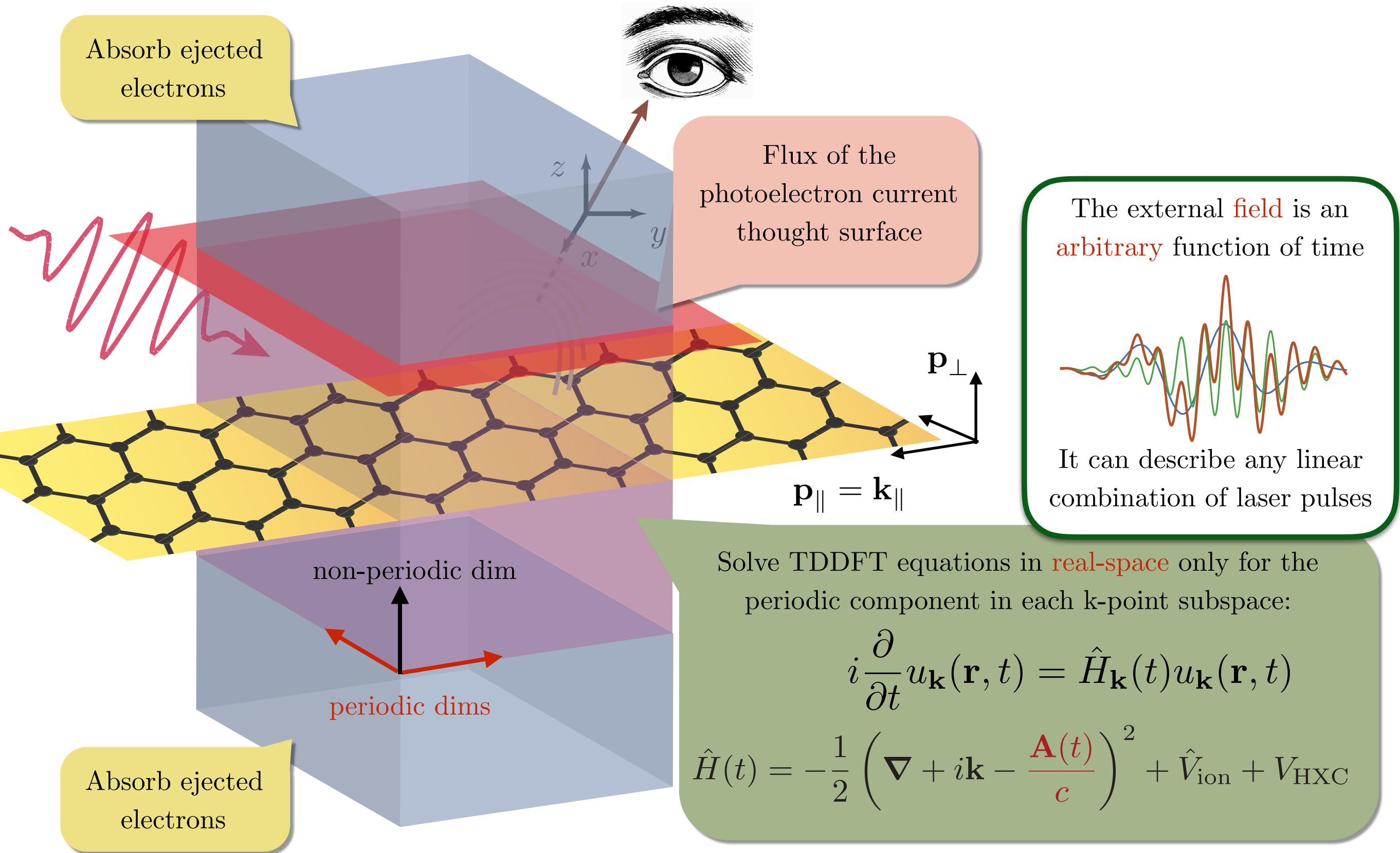$$P(E = \frac{(\mathbf{p}_\parallel + \mathbf{p}_\perp)^2}{2}, \mathbf{p}_\parallel)$$

Δt

z

x

$\mathbf{p}_\perp$

$\mathbf{p}_\parallel = \mathbf{k}_\parallel$

$E_{kin} = \hbar\omega - E_b - \Phi$

E_{kin}

Φ

E_F

E_b

$\hbar\omega$
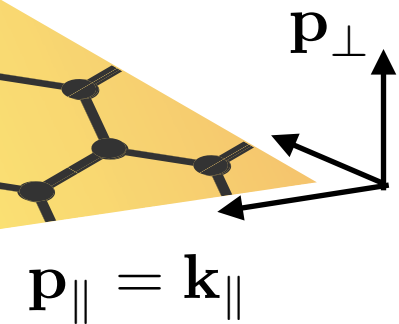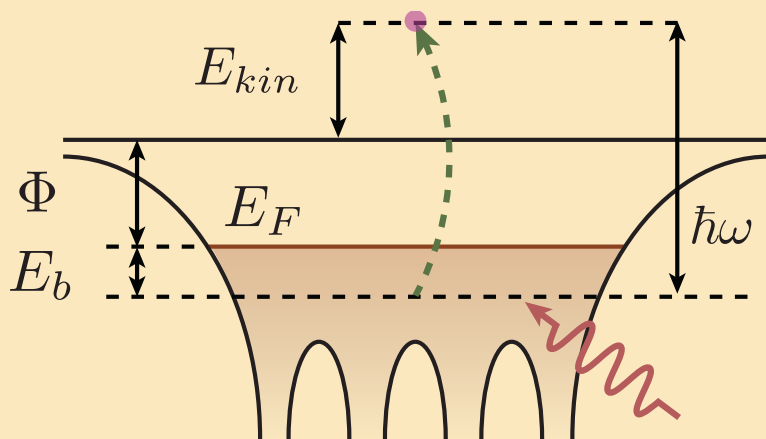
Modeling ARPES in finite volumes

Absorb ejected electrons

Flux of the photoelectron current thought surface

The external field is an arbitrary function of time

It can describe any linear combination of laser pulses

$\mathbf{p}_\perp$

$\mathbf{p}_\parallel = \mathbf{k}_\parallel$

non-periodic dim

periodic dims

Absorb ejected electrons

Solve TDDFT equations in real-space only for the periodic component in each k-point subspace:

$$i\frac{\partial}{\partial t}u_{\mathbf{k}}(\mathbf{r},t) = \hat{H}_{\mathbf{k}}(t)u_{\mathbf{k}}(\mathbf{r},t)$$

$$\hat{H}(t) = -\frac{1}{2}\left(\boldsymbol{\nabla} + i\mathbf{k} - \frac{\mathbf{A}(t)}{c}\right)^2 + \hat{V}_{\text{ion}} + V_{\text{HXC}}$$

UDG, H. Hübener, A. Rubio, arXiv:1609.03092 (2016)

Modeling ARPES with tSURFFP

$B \quad \hat{H}_{\mathrm{V}}(t), \chi_{\mathbf{p}}$

S

$A \quad \hat{H}_{\mathbf{k}}(t), \varphi_{\mathbf{k},j}$

z

x,y
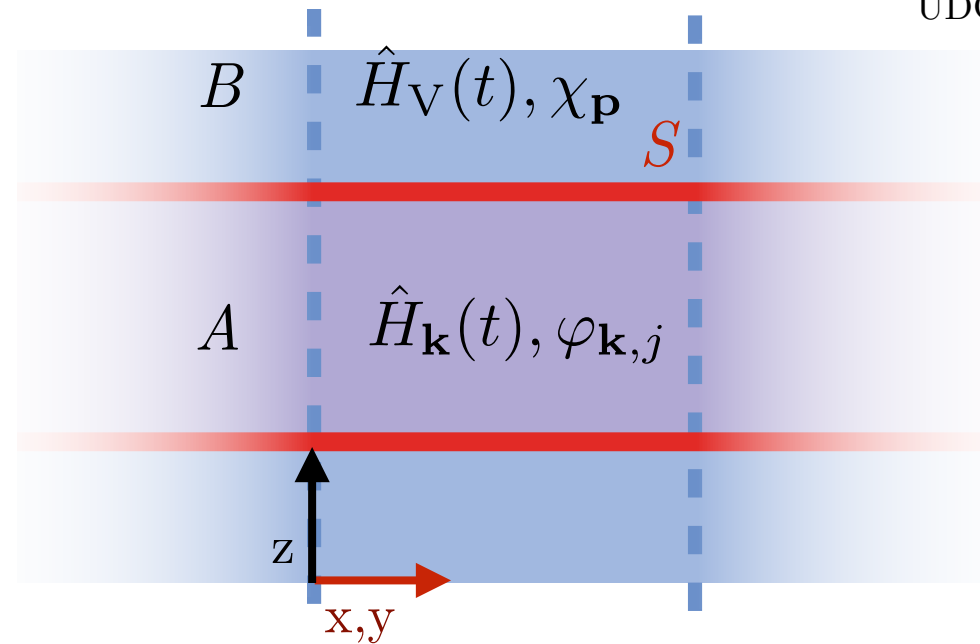
TDKS Hamiltonian

$$\hat{H}_{\mathbf{k}}(t)$$

$$\varphi_{\mathbf{k},j}(\mathbf{r},t) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{\mathbf{k},j}(\mathbf{r},t)$$

Volkov Hamiltonian

$$\hat{H}_{\mathrm{V}}(t) = \frac{1}{2}\left[-i\nabla - \frac{\mathbf{A}}{c}\right]$$

$$\chi_{\mathbf{p}}(\mathbf{r},t) = \sqrt{\frac{2\pi}{a_x a_y}} e^{i\mathbf{p}\cdot\mathbf{r}} e^{-i\phi(\mathbf{p},t)}$$

$$\phi(\mathbf{p},t) = \frac{1}{2}\int_0^t \mathrm{d}\tau \left[\mathbf{p} - \frac{\mathbf{A}(\tau)}{c}\right]^2$$

Working assumptions

$$\hat{H}(t) \approx \underset{z < z_S}{\hat{H}_{\mathbf{k}}(t)} + \underset{z \geq z_S}{\hat{H}_{\mathrm{V}}(t)}$$

$$\varphi_{\mathbf{k},j} = \varphi_{\mathbf{k},j,A} + \varphi_{\mathbf{k},j,B}$$

Use continuity equation

$$N_{\mathrm{esc}}(t) = -\sum_{i=1}^N \int \mathrm{d}\mathbf{p} \int_{BZ} \mathrm{d}\mathbf{k} \left| \int_0^t \mathrm{d}\tau \int_S \mathrm{d}\mathbf{s} \cdot \langle \chi_{\mathbf{p}}(\tau)|\hat{\mathbf{j}}|\varphi_{i,\mathbf{k}}(\tau)\rangle \right|^2$$

Momentum-resolved emission probability

$$\mathcal{P}(\mathbf{p} = \mathbf{g} + \mathbf{k}) = \frac{\partial N_{\mathrm{esc}}}{\partial \mathbf{p}} = \lim_{t\to\infty} \sum_{i=1}^N \int_{BZ} \mathrm{d}\mathbf{k} \left| \int_0^t \mathrm{d}\tau \int_S \mathrm{d}\mathbf{s} \cdot \langle \mathbf{g}|\hat{\mathbf{j}}|u_{i,\mathbf{k}}(\tau)\rangle e^{i\phi(\mathbf{p},t)} \right|^2$$
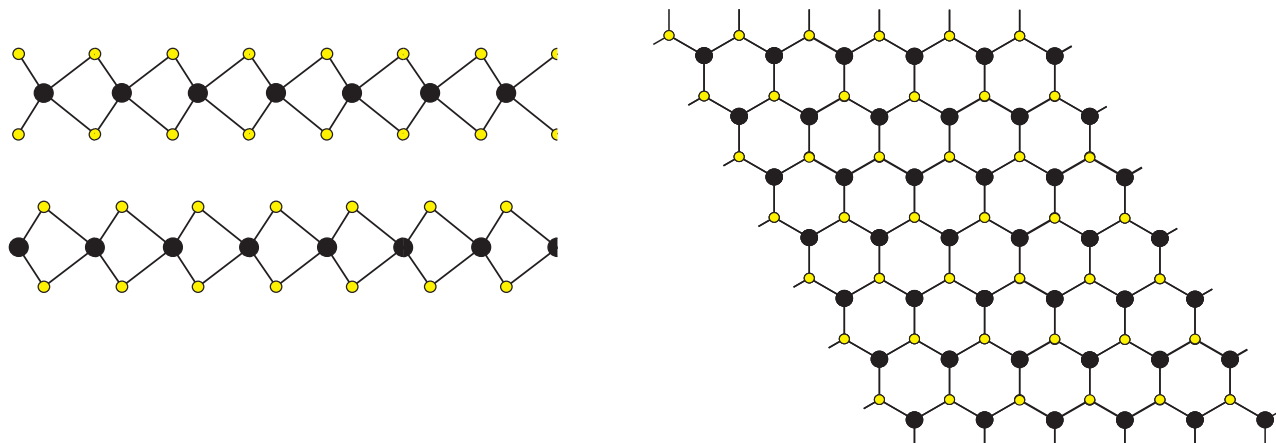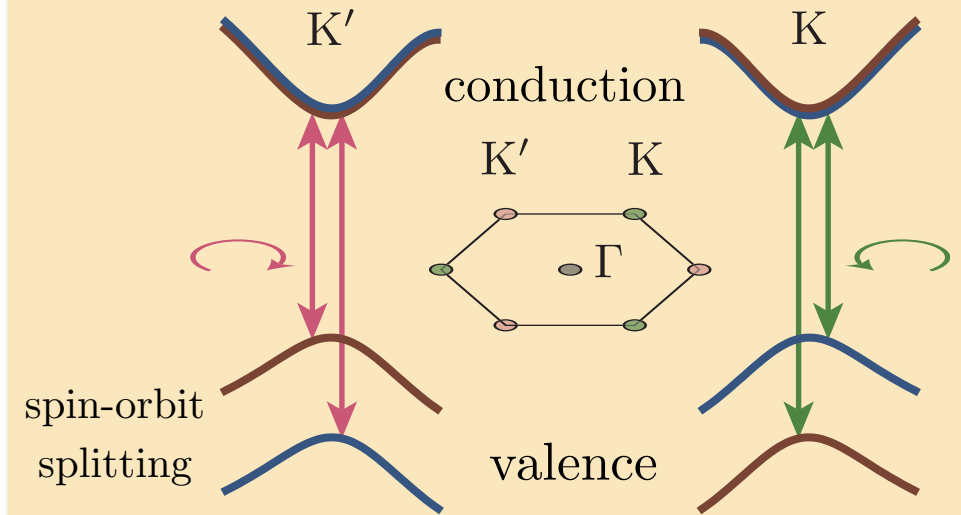
ARPES on WSe2

Monolayer
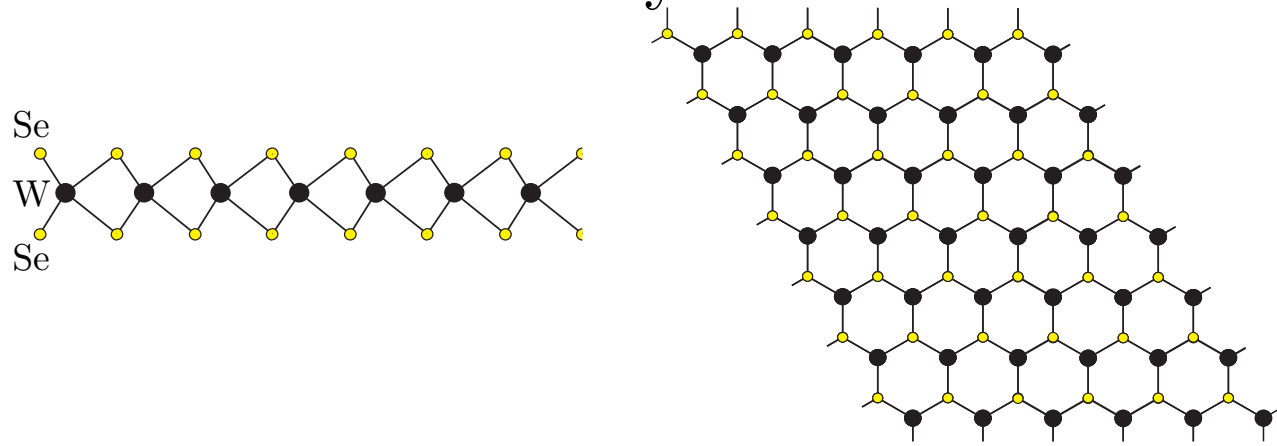
broken inversion symmetry for
odd layer stacking
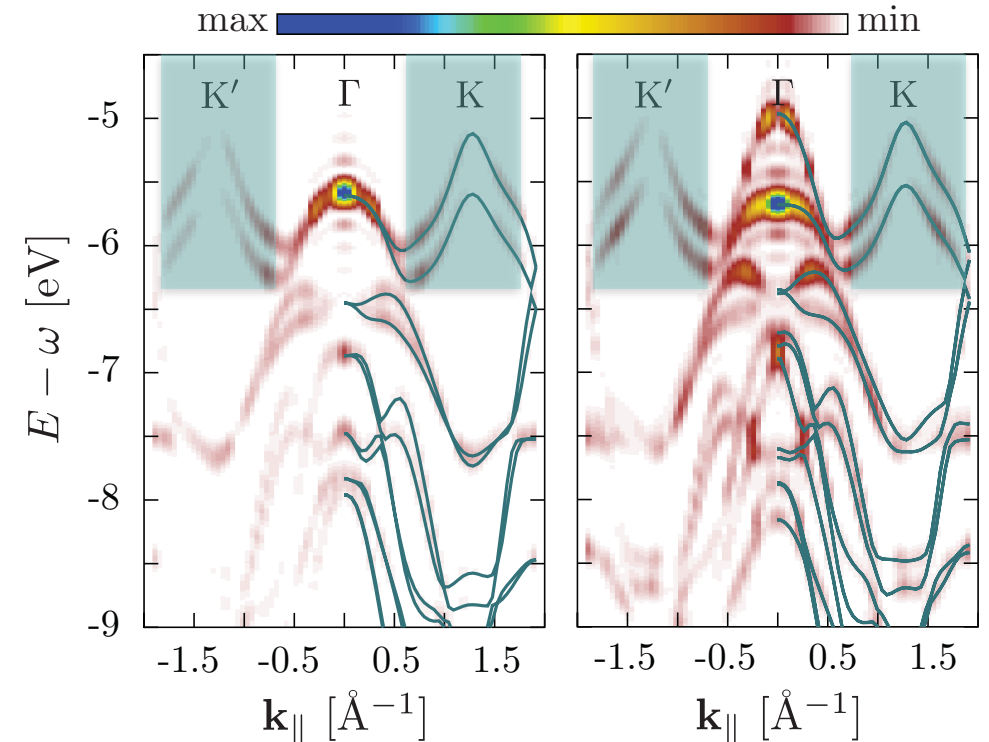
Bilayer

inversion symmetry for even
layer stacking
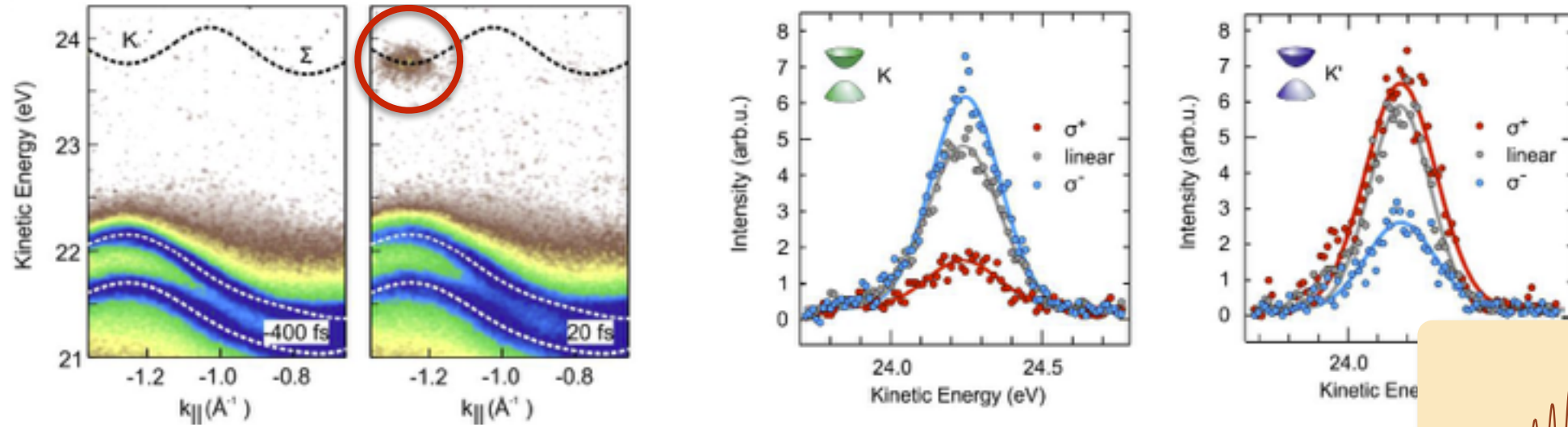
Spin/Valley selective excitation
(valleytronics)

K'   conduction   K

K'   K

Γ

spin-orbit
splitting   valence

TDDFT ARPES
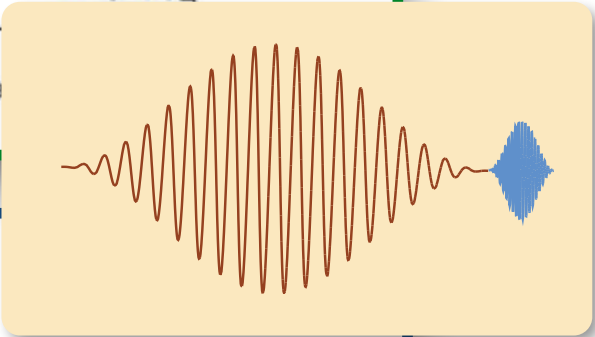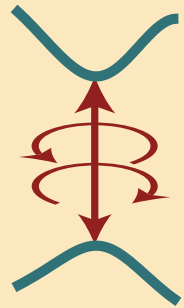
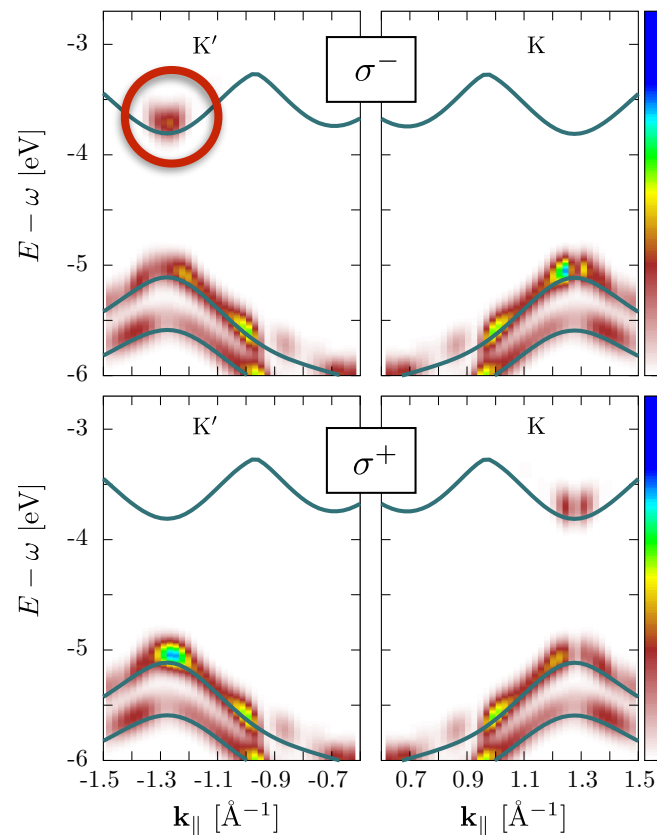Monolayer   Bilayer

max ▬▬▬▬▬▬ min

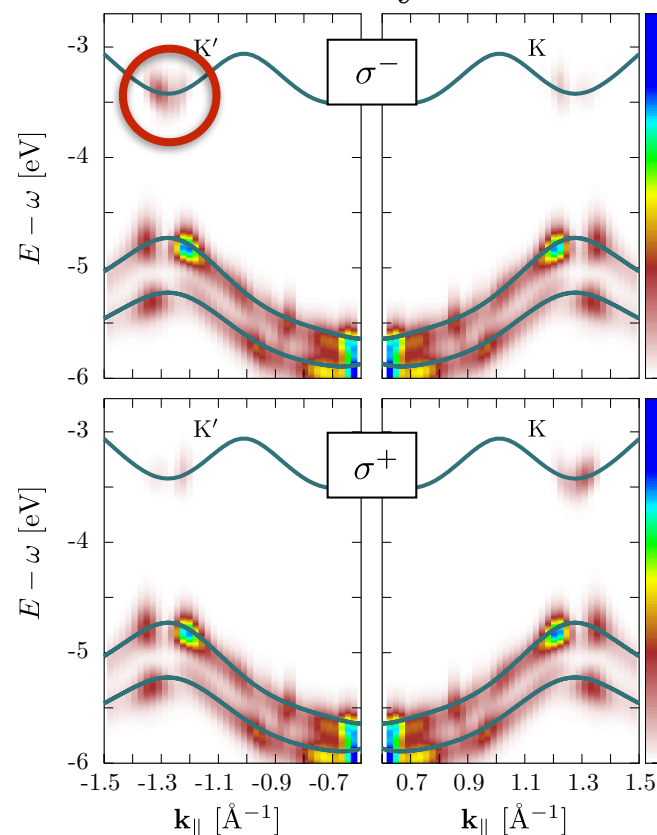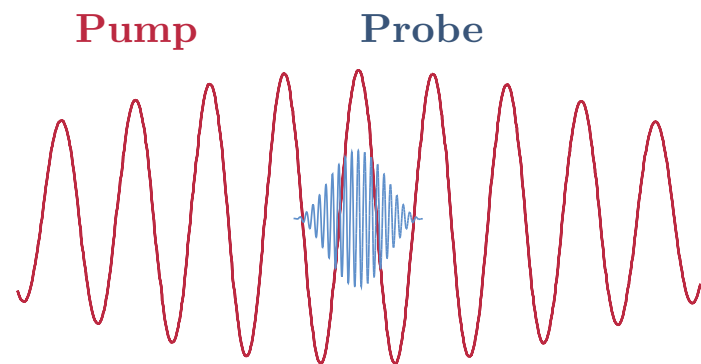# Pumped WSe2 trARPES

Experiment (2h bulk)

dichroic excitation

Theory

Monolayer

Bilayer

Bertoni R. et al. arXiv:1606.03218 (2016)

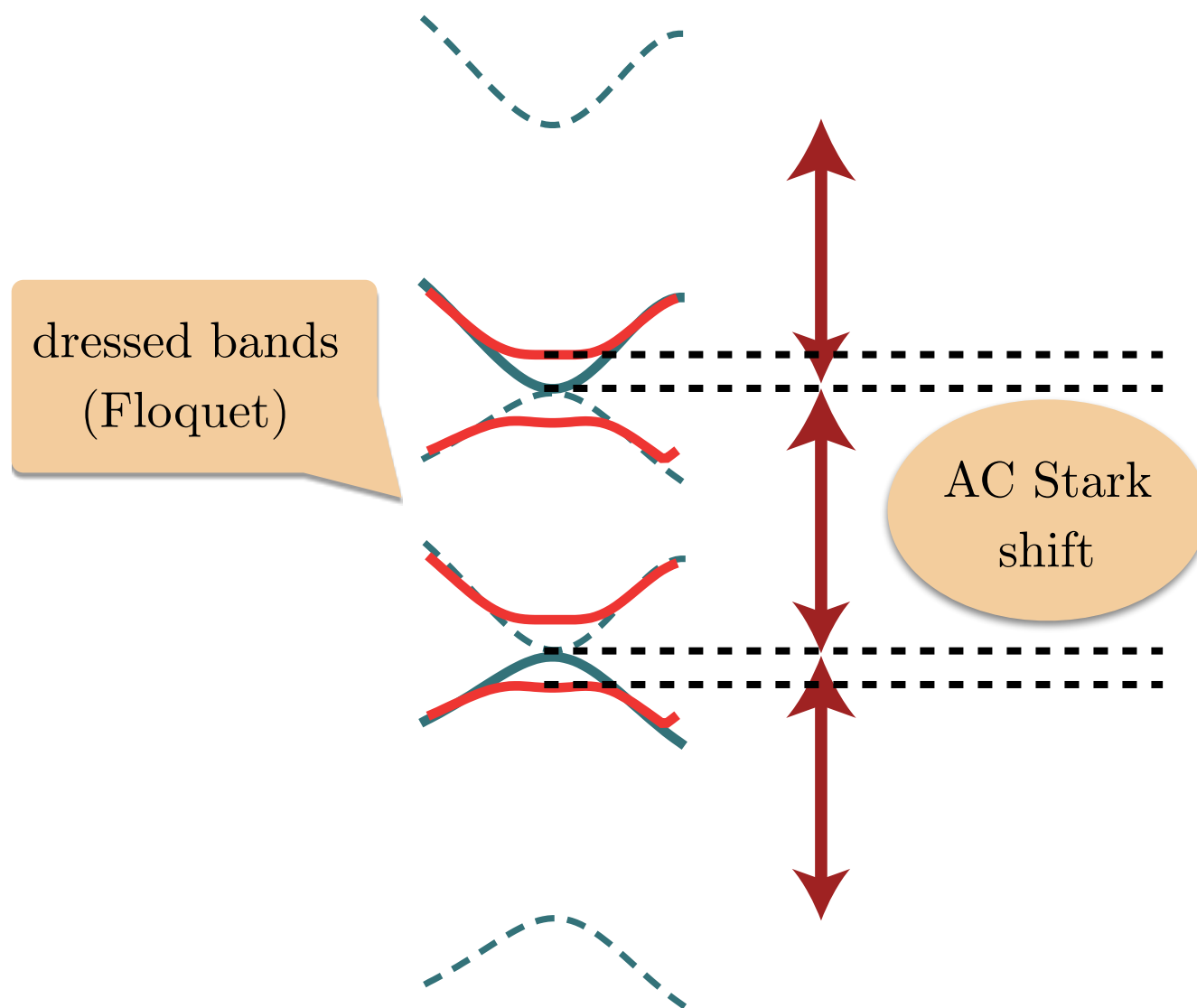Umberto De Giovannini, EHU/UPV   **Applications**
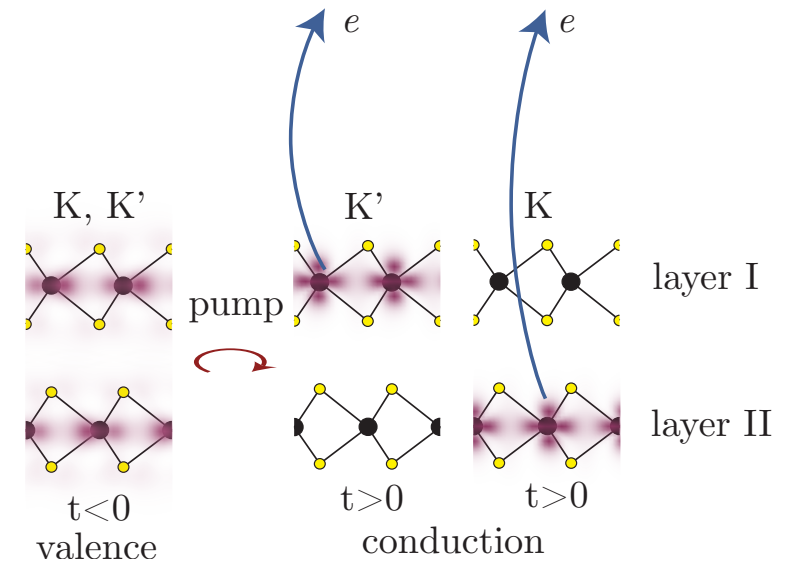
Probing while the pump is switched on?

Pump Probe

A periodically driven system is different!!

conduction

Ω

valence

dressed bands
(Floquet)

AC Stark
shift

Photon dressing and undressing

e^- $\uparrow\downarrow$ $\boldsymbol{k}$

Δt

Two independent approaches

trARPES	Floquet

K

Ω

Time delay

K

$E - \omega_p$ [eV]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

t

t

t

t

Photon dressing and undressing

K

Ω

Field amplitude A

$1 \ [\text{a.u.}] = 0.02 \ \text{V/Å}$

A=1

A=2

A=5

$\Omega - \delta$

$\Omega + \delta$

$E - \omega_p$ [eV]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

$\mathbf{k}_{\parallel}$ [Å^{-1}]

$\mathbf{k}_{\parallel}$ [Å^{-1}]
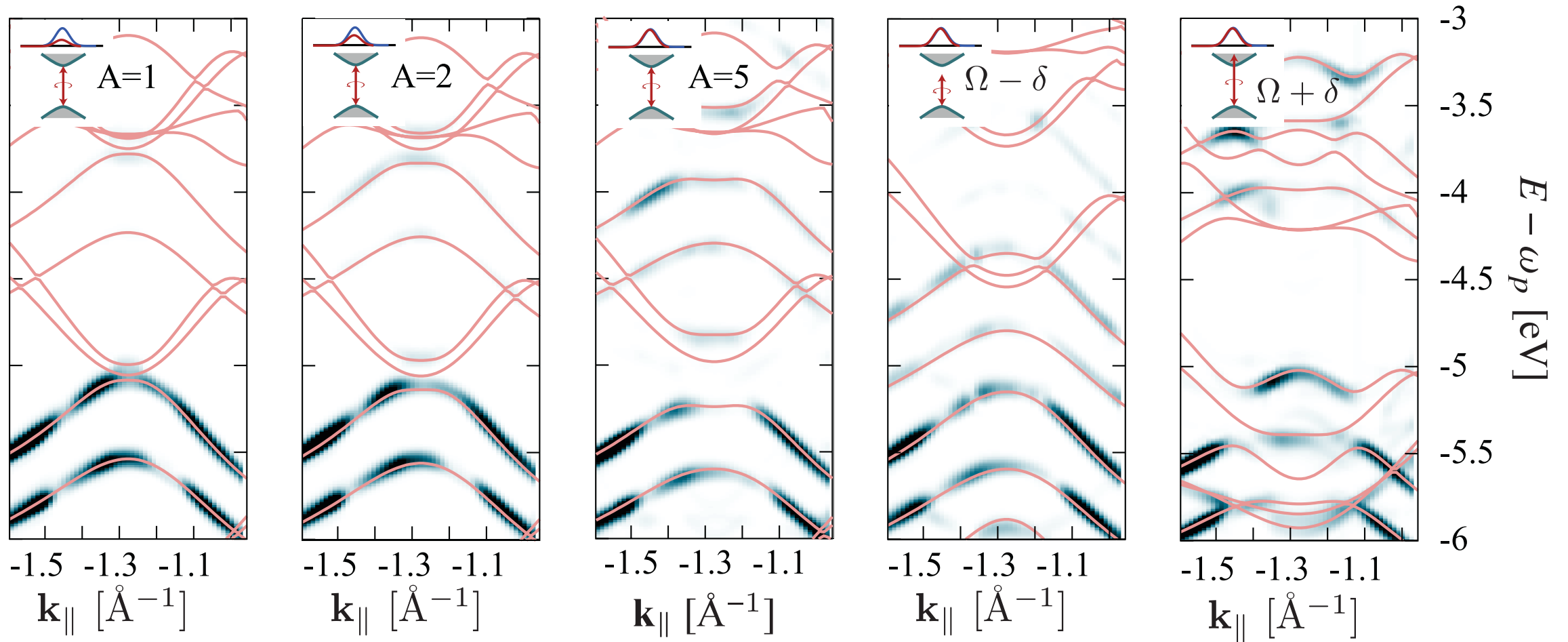
Field amplitude

Detuning

Photon dressing and undressing

Off-resonant

Resonant

AC Stark shift

$\delta = 0.3$ [eV]
$\delta = 0.2$ [eV]
$\delta = 0.1$ [eV]
$\delta = 0$ [eV]

Photon dressing and undressing
Spin texture

Summary

- Time and spin resolved ARPES can be efficiently simulated with TDDFT

- Ab-initio simulations indicate that Floquet sidebands hybridization can be observed in trARPES experiments

References

H. Hübener, et al. arXiv:1604.03399 (2016)

UDG, et al. arXiv:1609.03092 (2016)

Bertoni R. et al. arXiv:1606.03218 (2016)

UDG, et al. arXiv:1609.03218 (2016)

http://www.tddft.org/programs/octopus

Acknowledgements