Ab-initio spin and time-resolved ARPES with QED-TDDFT: driving WSe2 out of equilibrium

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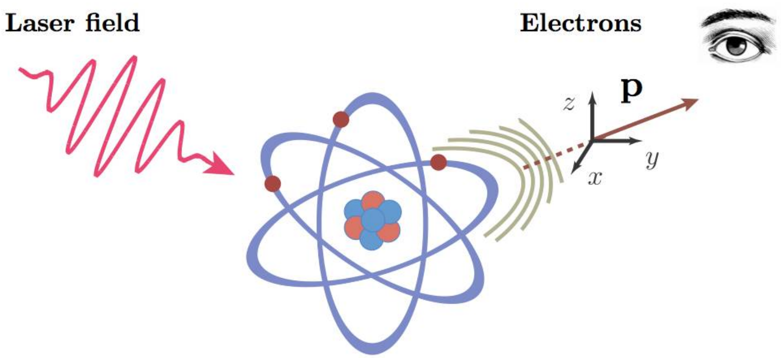
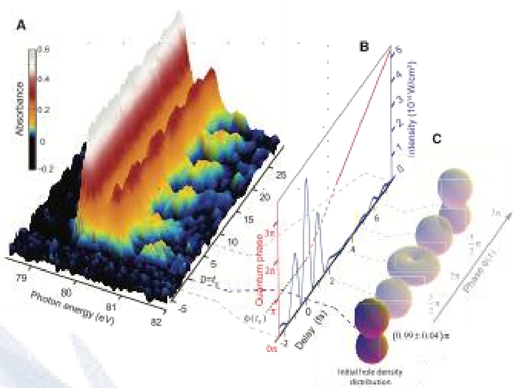
Transient Absorption and Photoelectron Spectroscopies

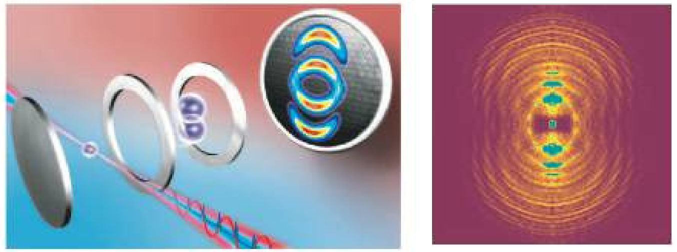


Hole states time evolution



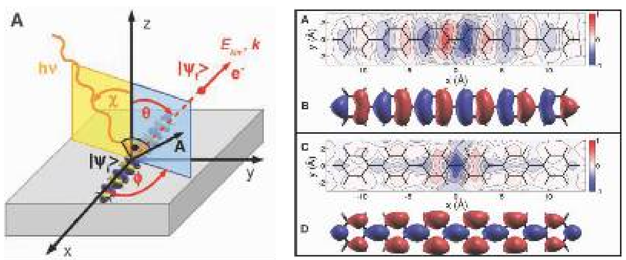
A. Wirth, et al. *Science* 334, 195 (2011).

Shearing interferograms



T. Remetter *et al.*, *Nat Phys* **2**, 323 (2006)

Orbital reconstruction



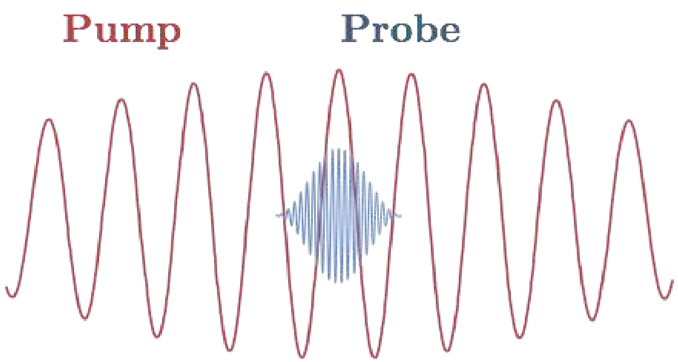
P. Puschnig et al. *Science* **326**, 702 (2009).

Time resolved pump-probe spectroscopy

Image reconstruction, monitoring electron-ion dynamics,

time-dependent band-structure ("light induced topological states")

Transient Absorption and Photoelectron Spectroscopies

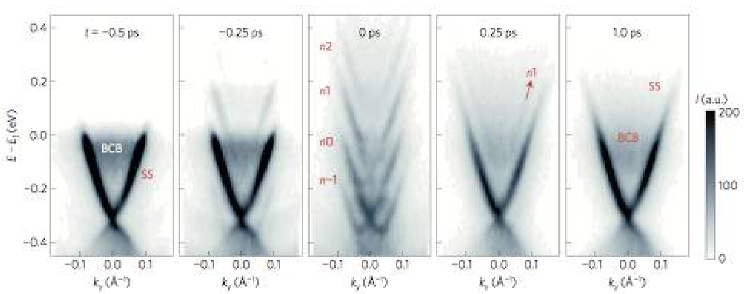


We are probing stationary states in a periodically driven system

Harvard Natural Sciences Lecture Demonstrations

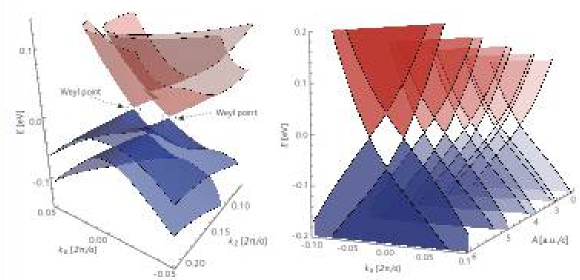
Examples:

Floquet sidebands



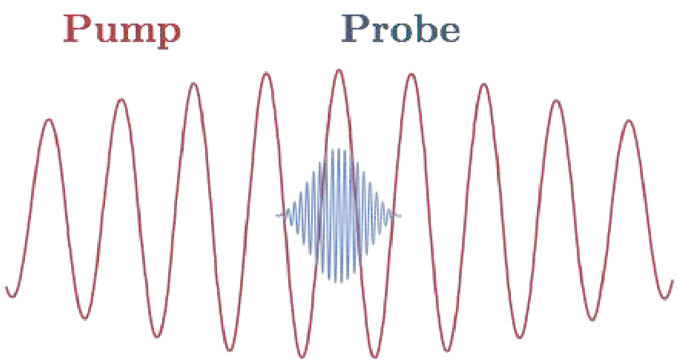
F. Mahmood, et al. Nat Phys **12**, 306 (2016)

Floquet Weyl semimetal



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

Transient Absorption and Photoelectron Spectroscopies



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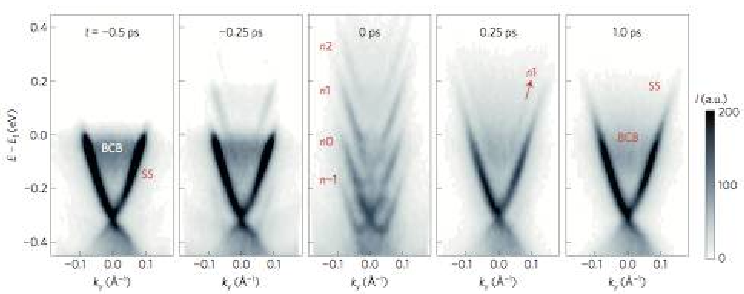

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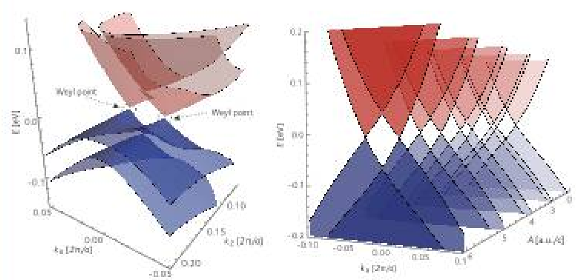
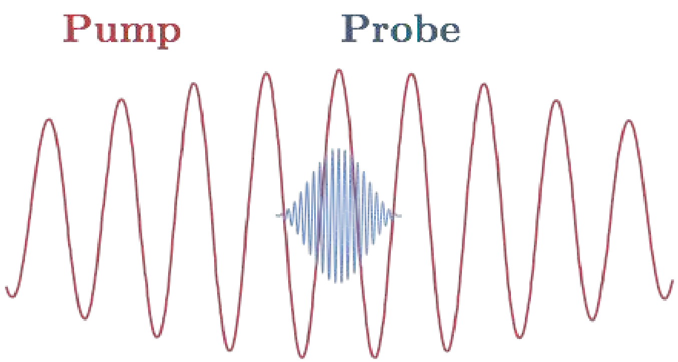


H. Hübener, et al. arXiv:1604.03399 (2016)

Transient Absorption and Photoelectron Spectroscopies

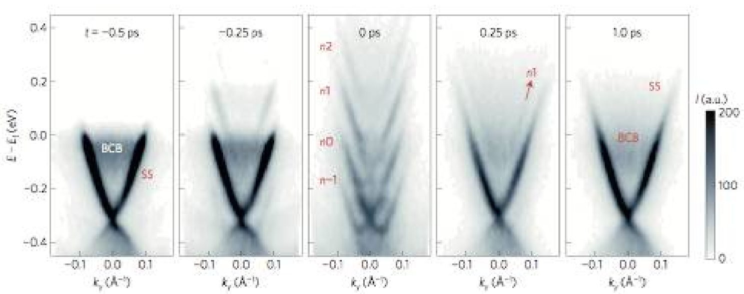


We are probing stationary states in a periodically driven system

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

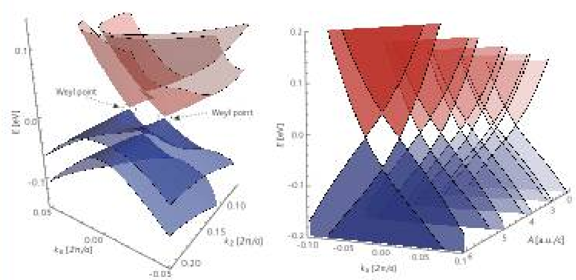
Examples:

Floquet sidebands



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

Floquet Weyl semimetal



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

Outline

Many body problem out of equilibrium:

A Time Dependent DFT approach
Some example(s):

Simulating real-time photon-dressing
of quasiparticles with TD-ARPES in TMDs

Quantum Electrodynamics DFT: QED-TDDFT

bridging quantum optics and materials modeling

QED Materials: *light induced topology*

Theoretical Development: QED-TDDFT
“New States of Matter”

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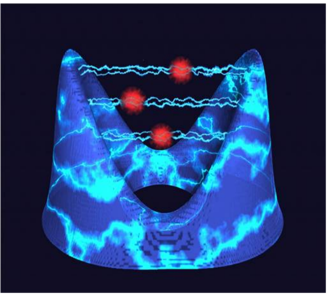


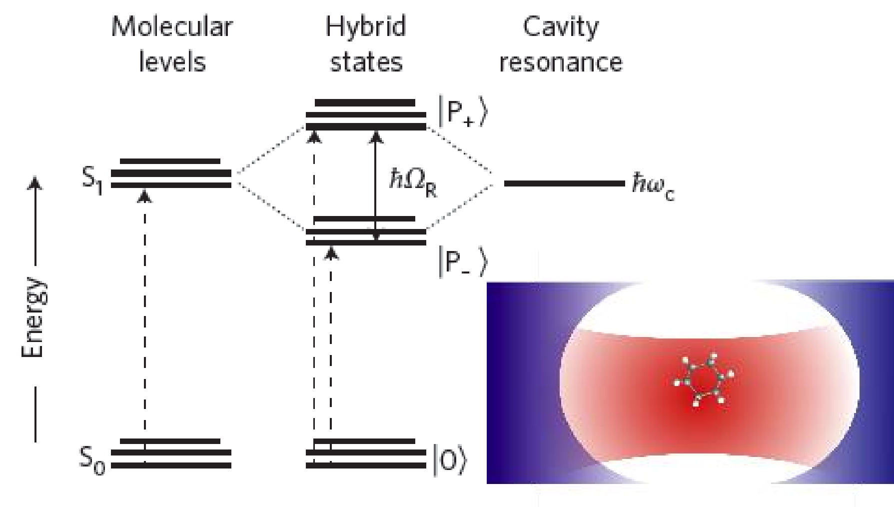
“bridging quantum optics and materials modeling”

Correlated photon-electron wavefunctions in cavity QED

Why?

QED-Chemistry:
Modifying Chemical Landscapes by Coupling to Vacuum Fields

QED-Materials
“New states of matter”



M. Ruggenthaler, J. Flick, C. Pellegrini, H. Appel, I.V. Tokatly, AR PRA (2014), J. Flick, M. Ruggenthaler, H. Appel, AR PNAS (2015) and PNAS (in press 2017)

New Development: QED-DFT

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Electronic system $\mapsto \{\mathbf{r}_i\}_{i=1}^{N}$, photons $\mapsto \{q_\alpha, p_\alpha, \omega_\alpha\}_{\alpha=1}^{M}$

Dipole length-form Hamiltonian

$$\hat{H} = \underbrace{\hat{T} + \hat{V}_{ee} + \hat{V}_{ext}(t)}_{\hat{H}_0} + \underbrace{\frac{1}{2}\sum_\alpha \left[\hat{p}_\alpha^2 + \omega_\alpha^2\left(\hat{q}_\alpha - \frac{\boldsymbol{\lambda}_\alpha}{\omega_\alpha}\hat{\mathbf{R}}\right)^2\right]}_{\frac{1}{8\pi}\int(\hat{B}^2+\hat{E}^2)d\mathbf{r},\ \text{where } \hat{E}=\hat{D}-\hat{P}}$$

$$\hat{V}_{ext} = \sum_i v_{ext}(\mathbf{r}_i t) \text{ classical, } \hat{\mathbf{R}} = \sum_i \mathbf{r}_i,\ \hat{q}_\alpha = -(\hat{a}_\alpha + \hat{a}_\alpha^\dagger)/\sqrt{2\omega_\alpha}$$

M. Ruggenthaler, J. Flick, C. Pellegrini, H. Appel, I.V. Tokatly, AR PRA (2014),
J. Flick, M. Ruggenthaler, H. Appel, AR PNAS (2015) and PNAS (2017)

TDDFT: All observables are functionals of n(r,t) (Runge&Gross 1984)

Linear and non linear phenomena accessible
Octopus Code http://www.tddft.org

Time-Dependent Density Functional Theory, Lecture Notes in Physics, Springer Vols. 837, 706 (2012, 2006)

New Development: QED-TDDFT

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QED-TDDFT Theorem

Basic variables: electron density and mean photon coordinates

$$\Psi(\{\mathbf{r}_j\},\{q_\alpha\},t)[\Psi_0, n, Q_\alpha]$$

where $n(\mathbf{r}t) = \langle \Psi | \hat{n}(\mathbf{r}) | \Psi \rangle$ and $Q_\alpha(t) = \langle \Psi | \hat{q}_\alpha | \Psi \rangle$

$$|\Psi(t)\rangle \overset{1:1}{\leftrightarrow} (n(\mathbf{r}t), q(t)) \overset{1:1}{\leftrightarrow} |\Phi(t)\rangle \otimes |\chi(t)\rangle$$

KS system: Schrödinger-Maxwell dynamics for N noninteracting e

$$i\partial_t \phi_j(\mathbf{r}t) = \left[-\nabla^2/2 + v_s(\mathbf{r}t)\right] \phi_j(\mathbf{r}t)$$

$$\underbrace{\ddot{Q}_\alpha + \omega_\alpha^2 Q_\alpha - \omega_\alpha \boldsymbol{\lambda}_\alpha \mathbf{R} = 0}_{\frac{\partial D}{\partial t} = \nabla \times B}$$

$$n_s(\mathbf{r}t) = n(\mathbf{r}t) \text{ if } v_s = v_{\text{ext}} + v^{\text{el}}_{\text{Hxc}}[n] + v^{\alpha}_{\text{eff}}[n, Q_\alpha],\ v^{\alpha}_{\text{eff}} = v^{\alpha}_{\text{MF}} + v^{\alpha}_{\text{xc}}$$

M. Ruggenthaler, J. Flick, C. Pellegrini, H. Appel, I.V. Tokatly, AR PRA (2014), I.V. Tokaly PRL -82104)
J. Flick, M. Ruggenthaler, H. Appel, AR PNAS (2015) and PNAS (accepted 2017)

"New States of Matter"

- **Quantum electrodynamics QED-TDDFT**

Test exact Cases:

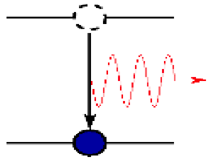
Spontaneous Emission: entangled photons

Strong light-mater coupling: Photon bound states

QED-Chemistry

Bond-length and vibrational control HD
Modifying and controlling conical intersections

bridging quantum optics and materials modeling

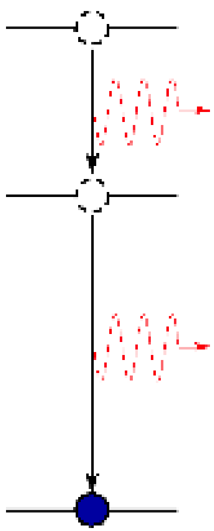


Spontaneous Emission

J. Flick, M. Ruggenthaler, H. Appel, AR PNAS (2015); PNAS(2017)

Spontaneous emission: correlated photon state

Weak Coupling



Strong Coupling

Time-dependent Coupled Maxwell-Kohn-Sham

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Towards large molecular systems and materials

Maxwell's equations in Schrödinger form

Riemann-Silberstein (R-S) vector in vacuum [1]:

$$\vec{F}(\vec{r},t) = \sqrt{\epsilon_0/2}\ \vec{E}(\vec{r},t) + i\sqrt{1/(2\mu_0)}\ \vec{B}(\vec{r},t)$$

Maxwell's equations in vacuum:

$$\vec{\nabla}\cdot\vec{E} = \frac{1}{\epsilon_0}\rho \qquad \vec{\nabla}\times\vec{E} = -\partial_t\vec{B}$$

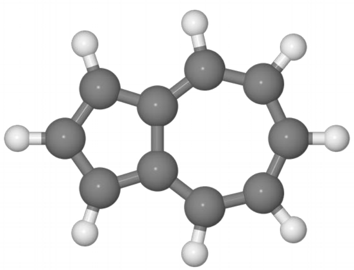
$$\vec{\nabla}\cdot\vec{B} = 0 \qquad \vec{\nabla}\times\vec{B} = \mu_0\epsilon_0\partial_t\vec{E} + \mu_0\vec{j}$$

$$\Downarrow \qquad\qquad \Downarrow$$

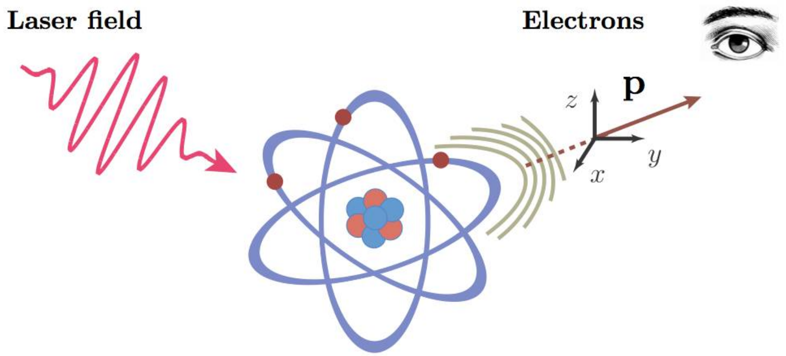
$$\vec{\nabla}\cdot\vec{F} = \frac{1}{\sqrt{2\epsilon_0}}\rho \qquad i\partial_t\vec{F}(\vec{r},t) = \pm c_0\vec{\nabla}\times\vec{F}(\vec{r},t) - \frac{i}{\sqrt{2\epsilon_0}}\vec{j}$$

René Jestädt, Heiko Appel, Angel Rubio (to be published 2017)

Time-dependended Coupled Maxwell-Kohn-Sham eg. Azulene molecule



Transient Absorption and Photoelectron Spectroscopies



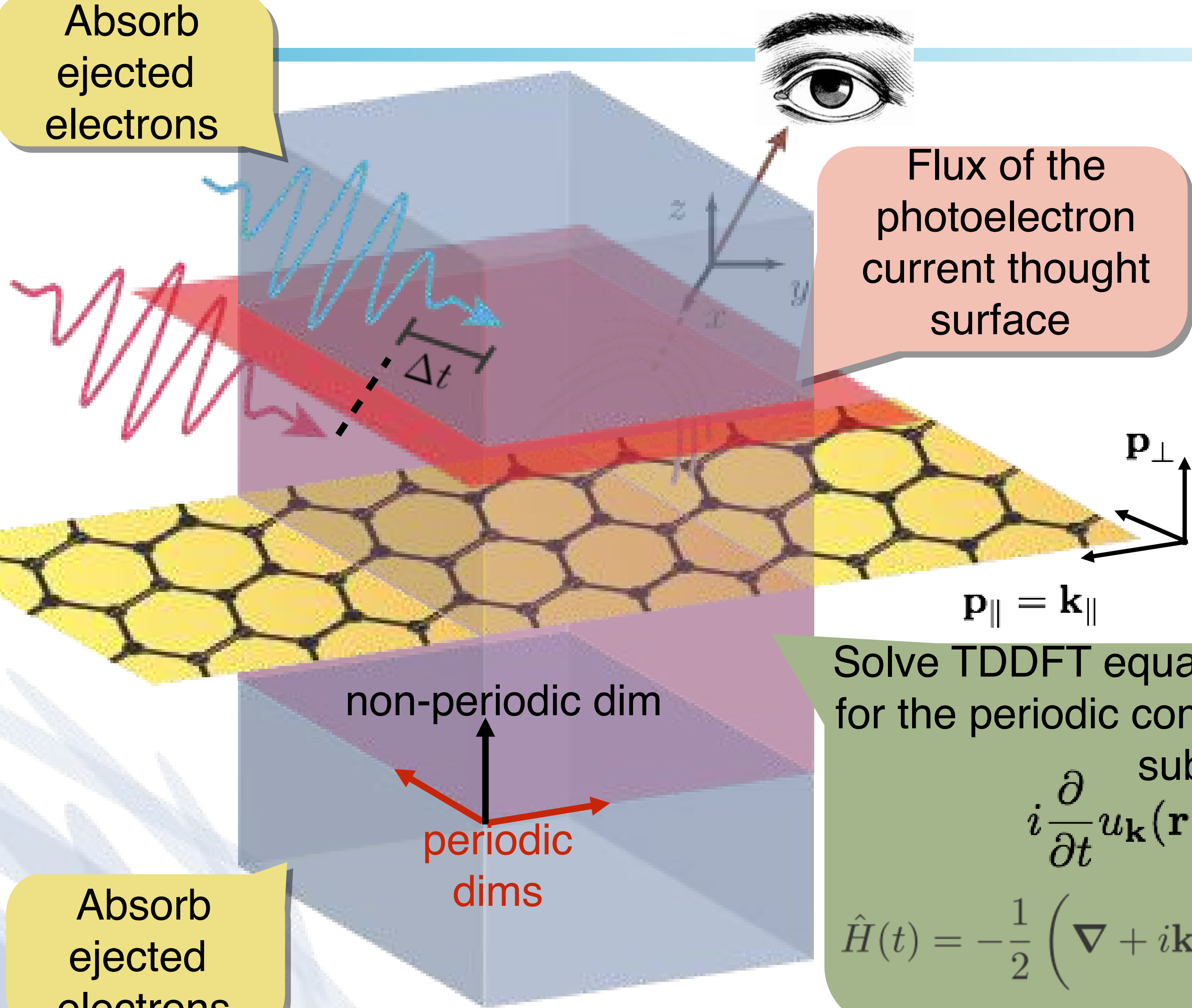
How to simulate trARPES with TDDFT

Application of trARPES on WSe2:

- *ARPES and spin-ARPES*
- *Dichroic spin-valley resonant excitation*
- *Photon dressing and dressed-band hybridization*

Time resolved pump-probe spectroscopy
time-dependent band-structure ("light induced topological states")

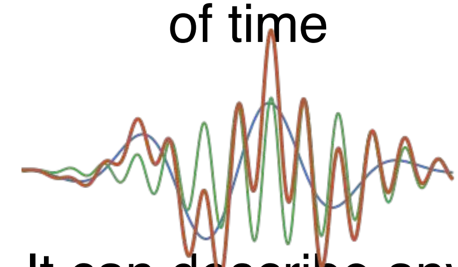
Modeling ARPES in finite volumes



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

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

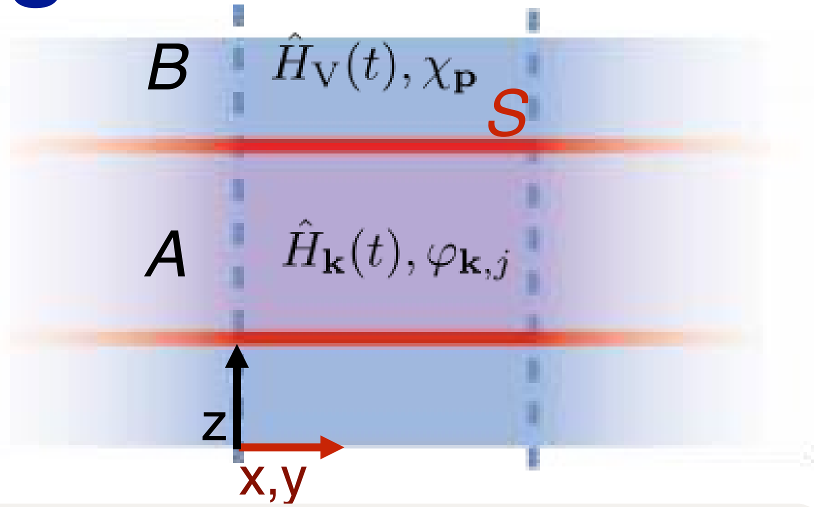
$$i\frac{\partial}{\partial t}u_{\mathbf{k}}(\mathbf{r},t) = \hat{H}_{\mathbf{k}}(t)u_{\mathbf{k}}(\mathbf{r},t)$$

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

Absorb ejected electrons

U. de Giovanninni, H. Hübener, AR (2016)

Modeling ARPES with tSURFFP



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

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

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

Use continuity equation

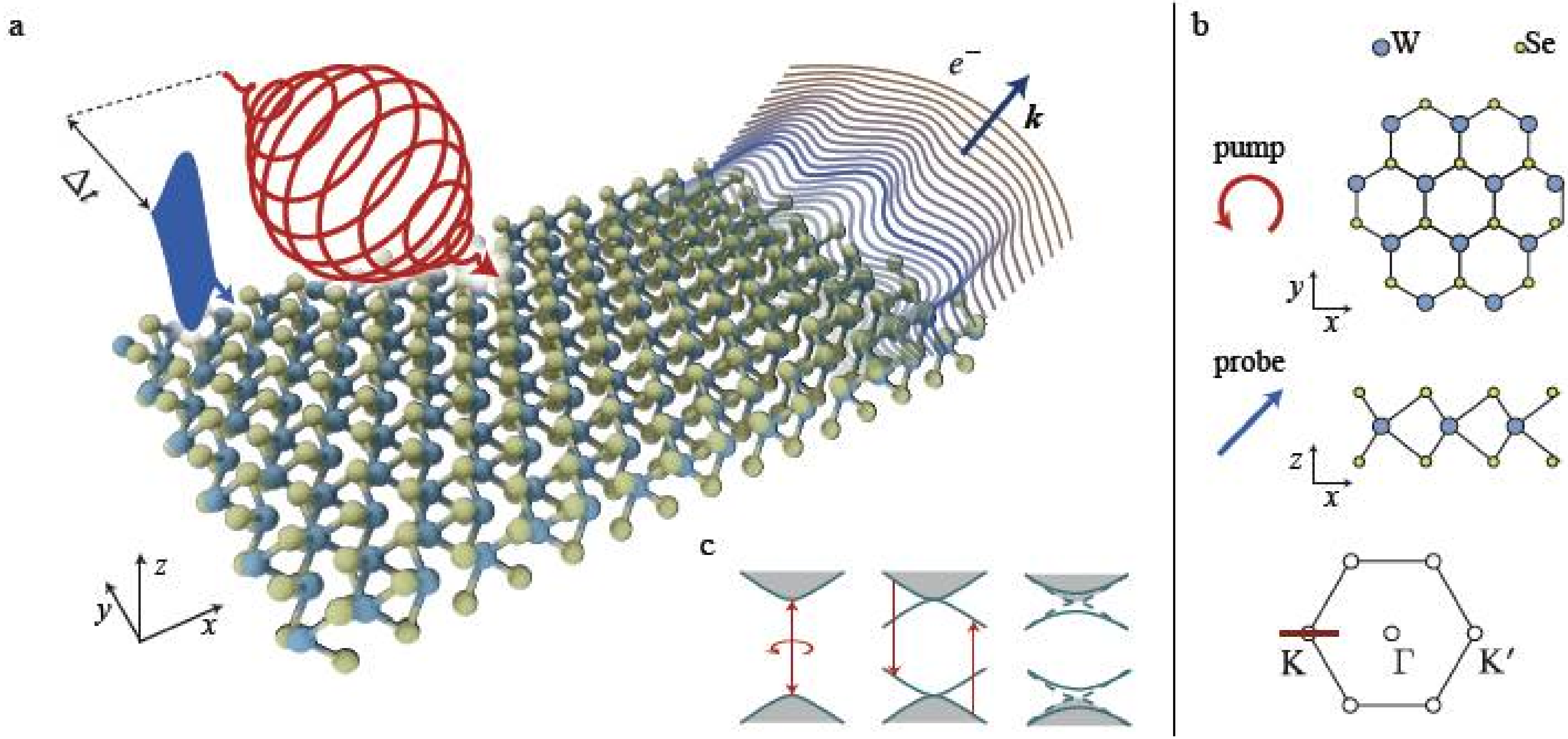
$$N_{\mathrm{esc}}(t) = -\sum_{i=1}^{N} \int \mathrm{d}\mathbf{p} \int_{BZ} \mathrm{d}\mathbf{k} \left| \int_0^t \mathrm{d}\tau \int_S \mathrm{d}\mathbf{s} \cdot \langle \chi_{\mathbf{p}}(\tau)|\hat{\mathbf{j}}|\varphi_{i,\mathbf{k}}(\tau)\rangle \right|^2$$

Momentum-resolved emission probability

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

U. de Giovanninni, H. Hübener, AR (2016)

Simulating real-time photon-dressing of quasiparticles with TD-ARPES in TMDs



U. de Giovanninni, H. Hübener, AR, Nano Letters (2016)

Simulating real-time photon-dressing of quasiparticles with TD-ARPES in TMDs

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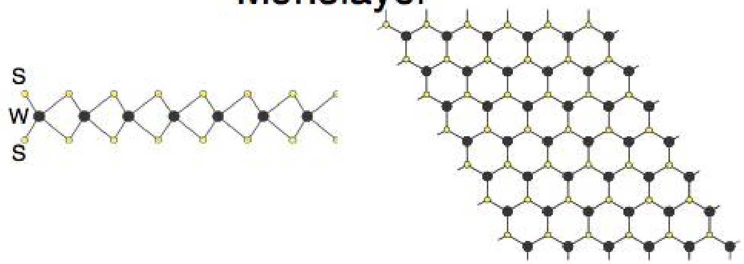
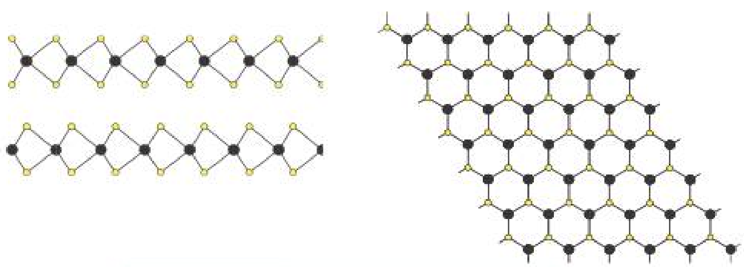
ARPES on WSe2

Monolayer



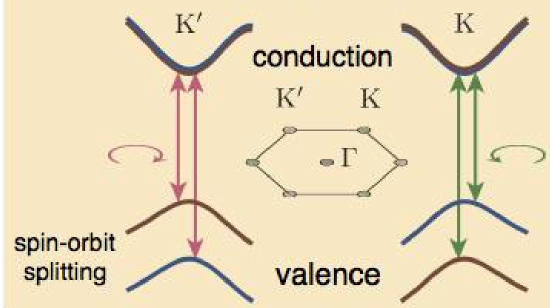
broken inversion symmetry for odd layer stacking

Bilayer

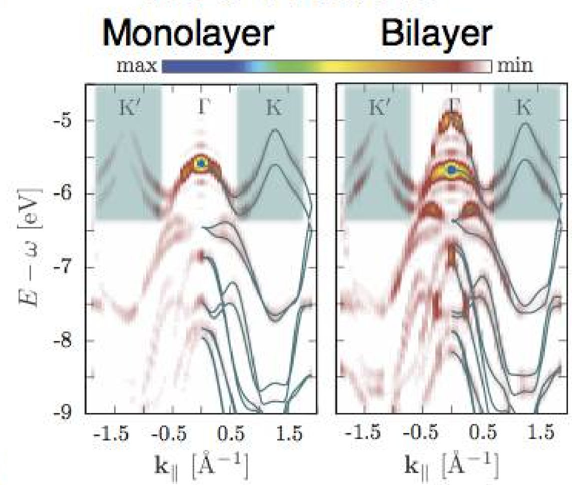


inversion symmetry for even layer stacking

Spin/Valley selective excitation (valleytronics)

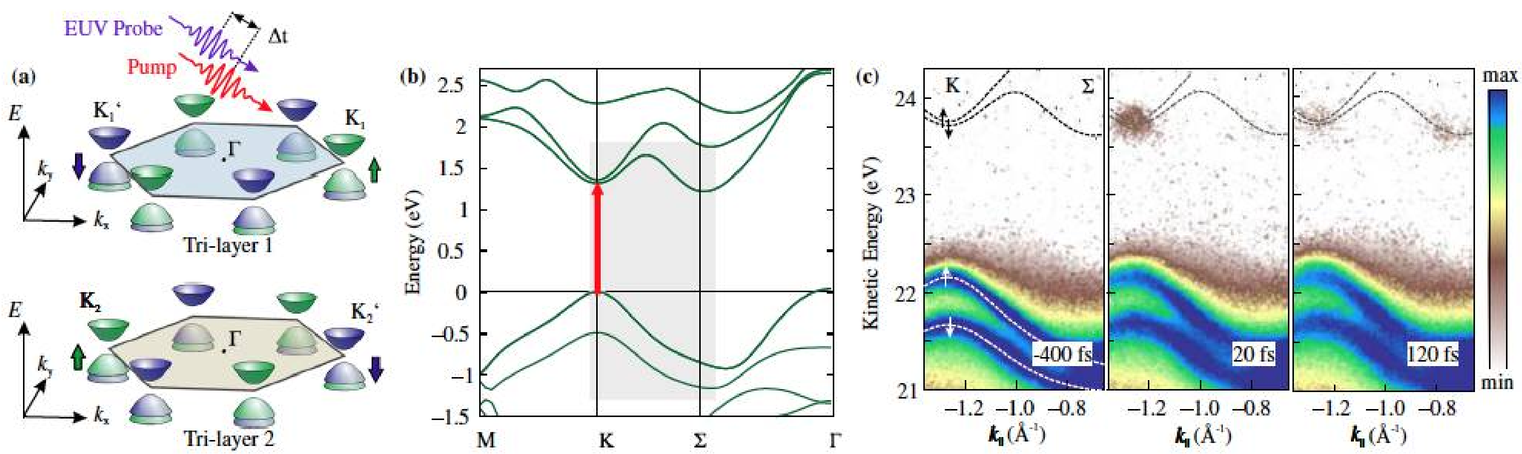


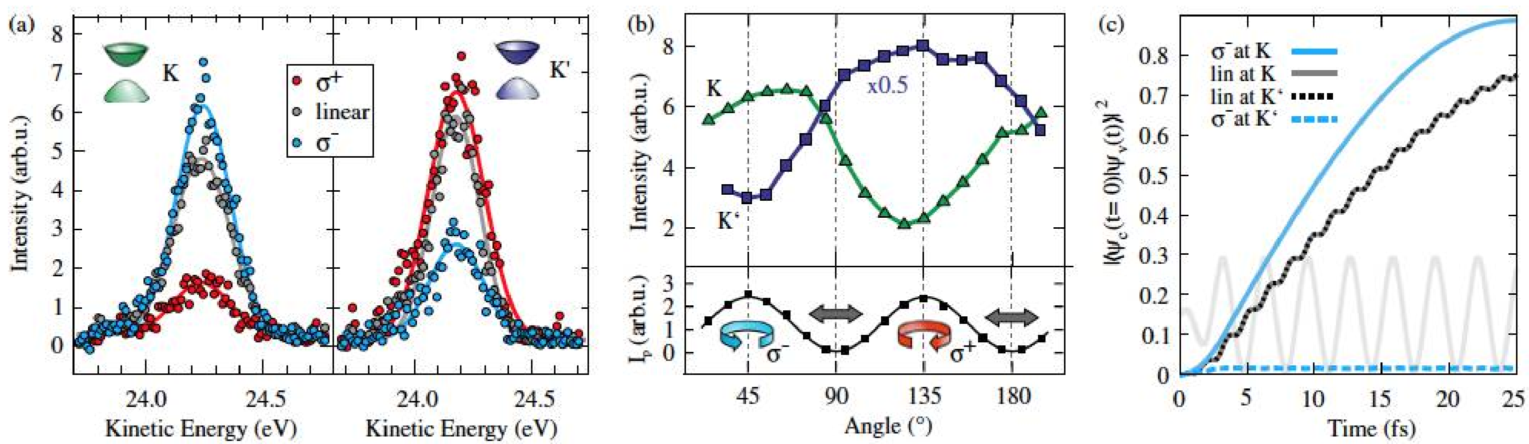
TDDFT ARPES



U. de Giovanninni, H. Hübener, AR (2016)

Simulating real-time photon-dressing of quasiparticles with TD-ARPES in TMDs



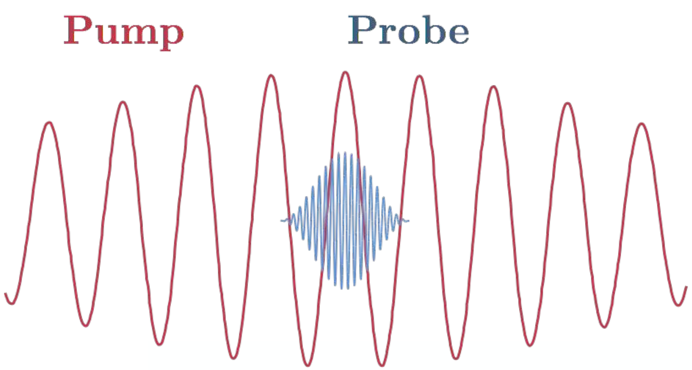


Ralph Ernstorfer

R. Bertoni, C. W. Nicholson, L. Waldecker, H. Hübener, C. Monney, U. De Giovannini, M. Puppin, M. Hoesch, E. Springate, R. T. Chapman, C. Cacho, M. Wolf, A. Rubio , R. Ernstorfer PRL (2016)

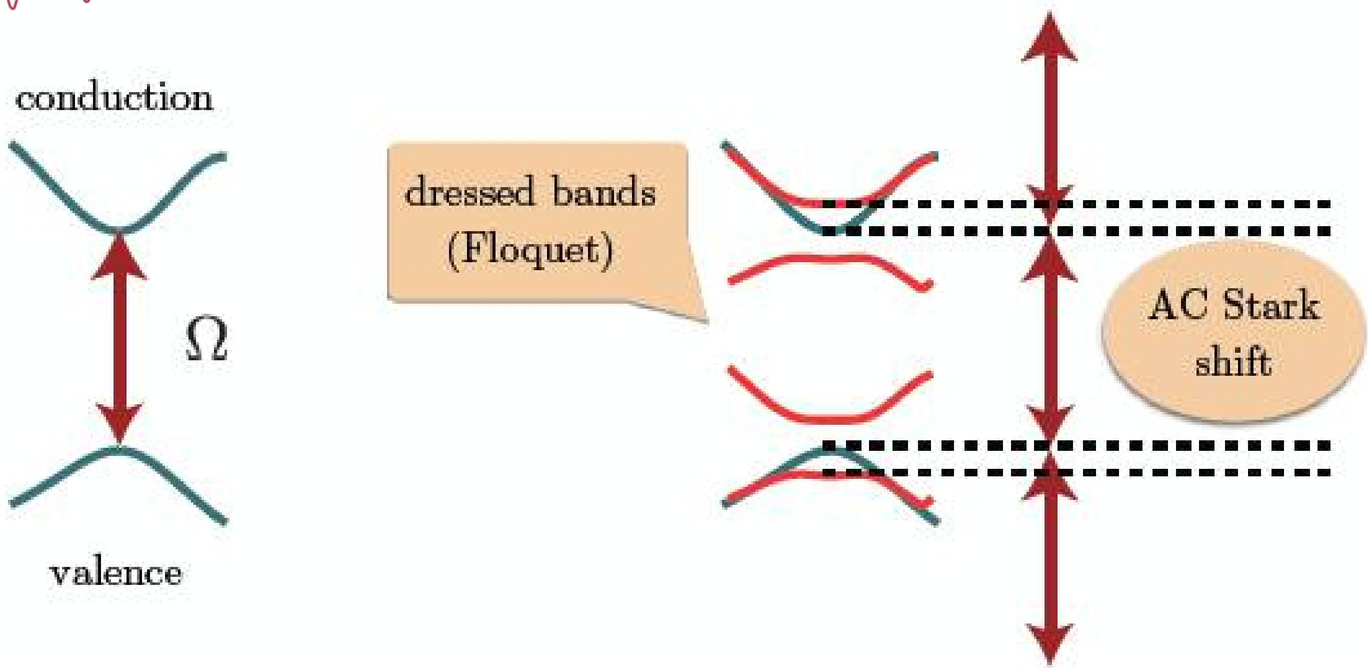
U. de Giovanninni, H. Hübener, AR NanoLetters (2016);

Simulating real-time photon-dressing of quasiparticles with TD-ARPES in TMDs



Probing while the pump is switched on?

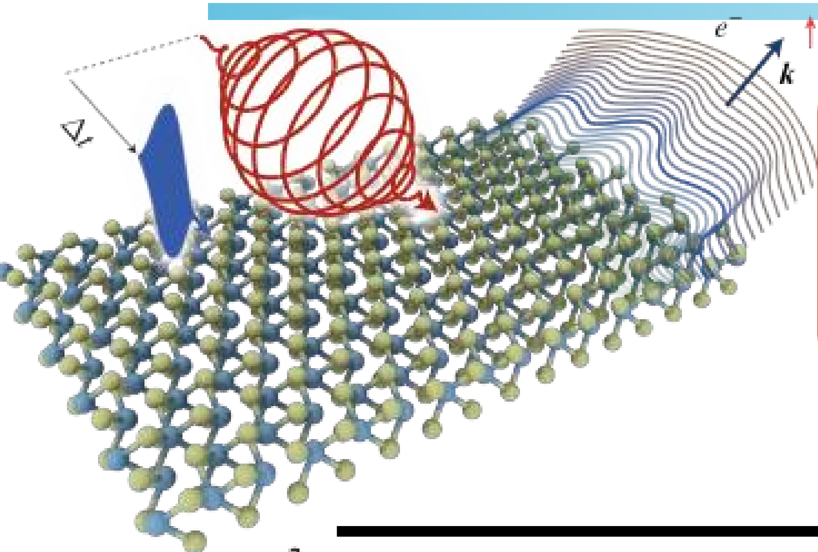
driven is different



U. de Giovanninni, H. Hübener, AR NanoLetters (2016)

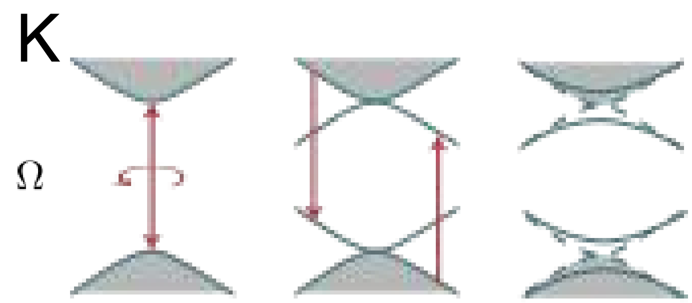
Photon dressing and undressing

U. de Giovanninni, H. Hübener, AR NanoLetters (2016)

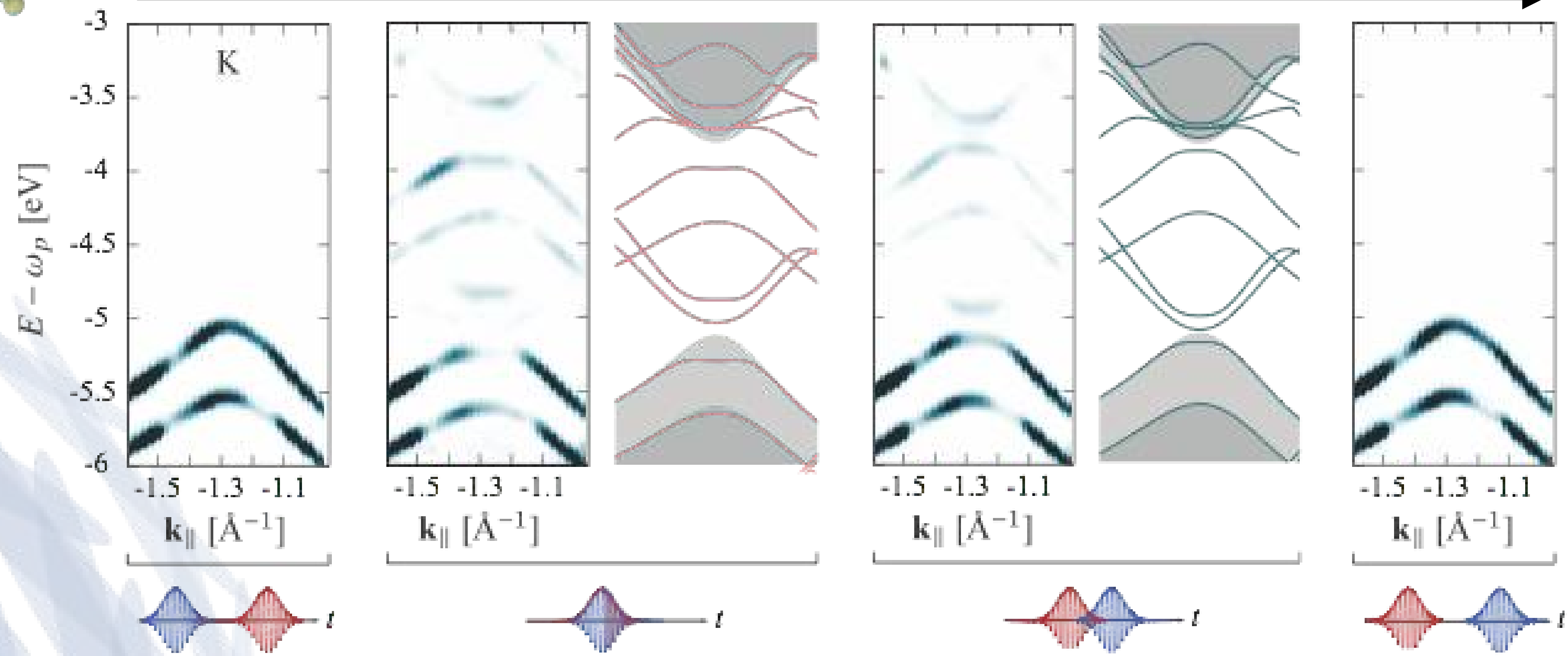


Two independent approaches

trARPES	Floquet



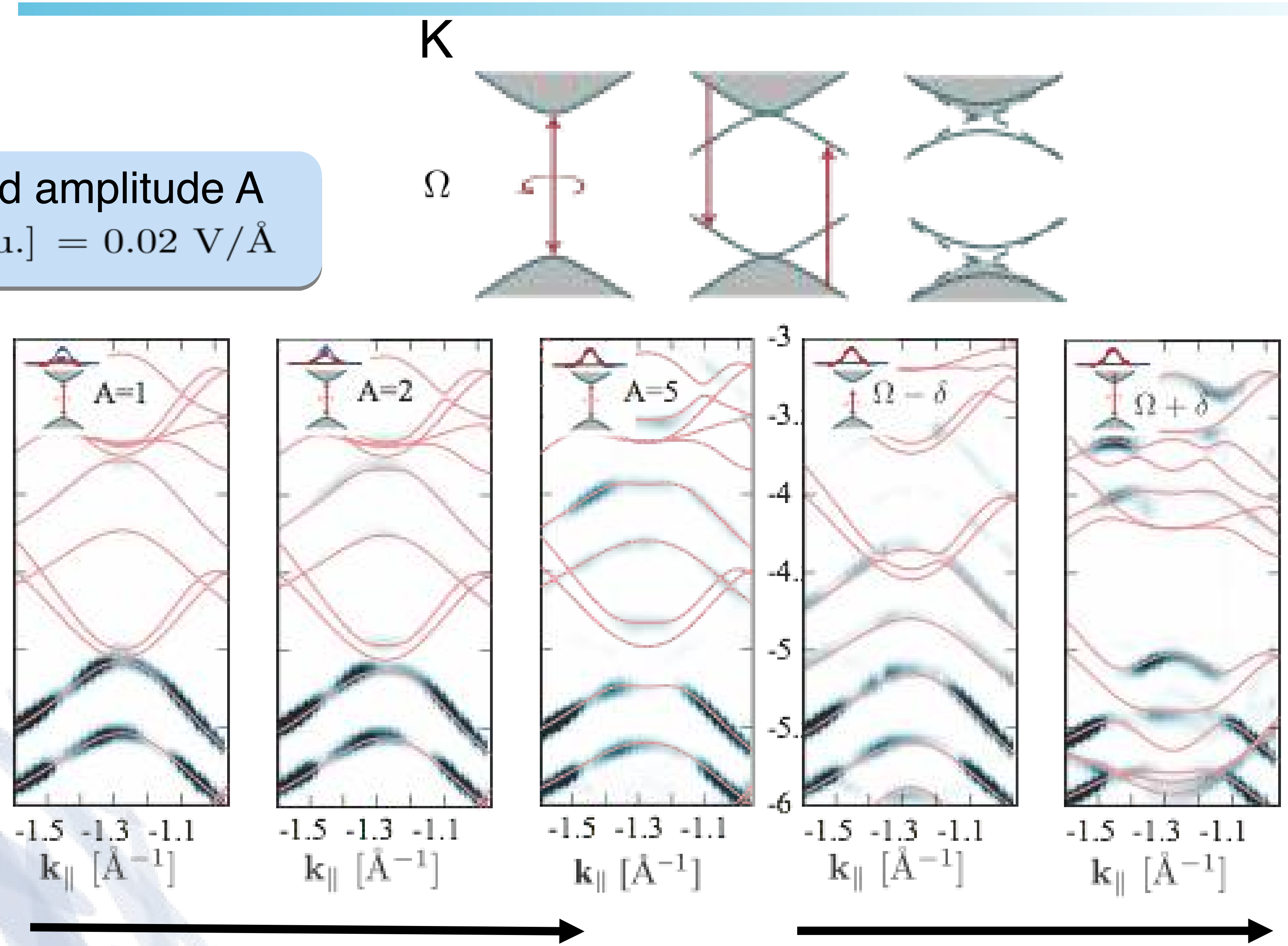
Time delay



Photon dressing and undressing

U. de Giovanninni, H. Hübener, AR NanoLetters (2016)

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

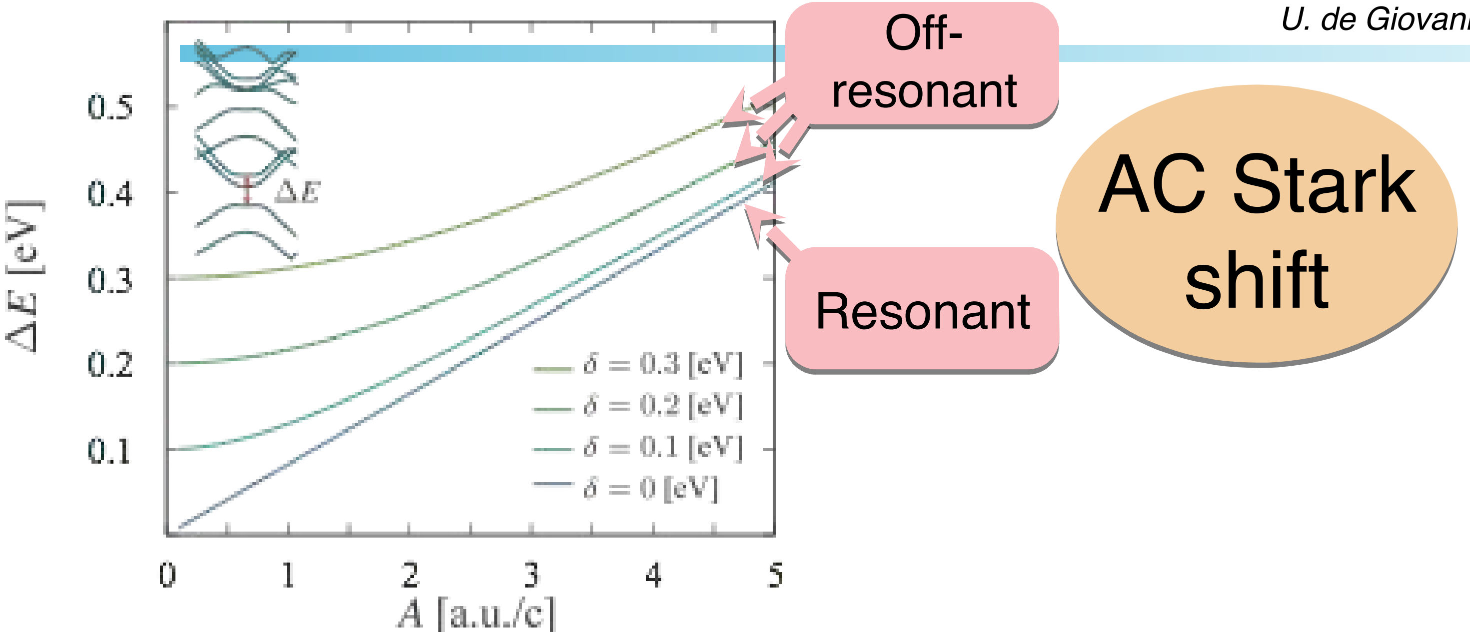


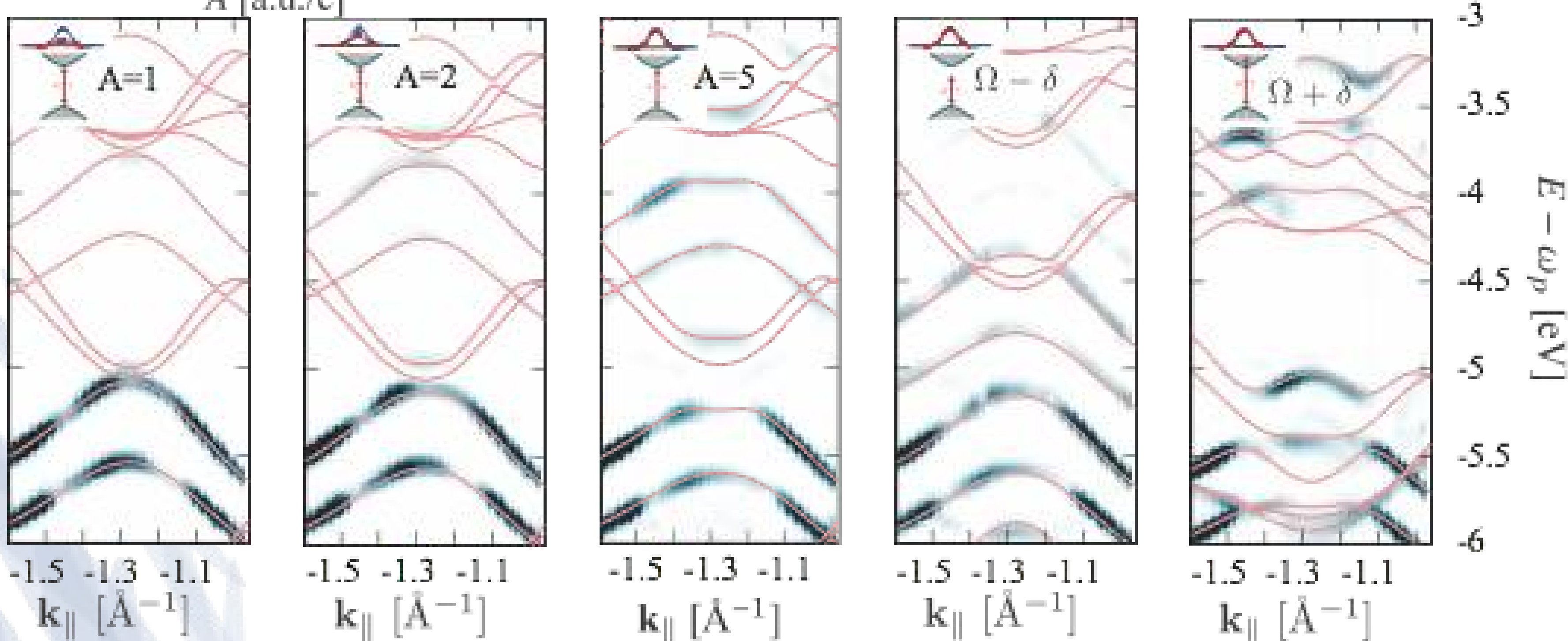
Field amplitude

Detuning

Photon dressing and undressing

U. de Giovanninni, H. Hübener, AR NanoLetters (2016)

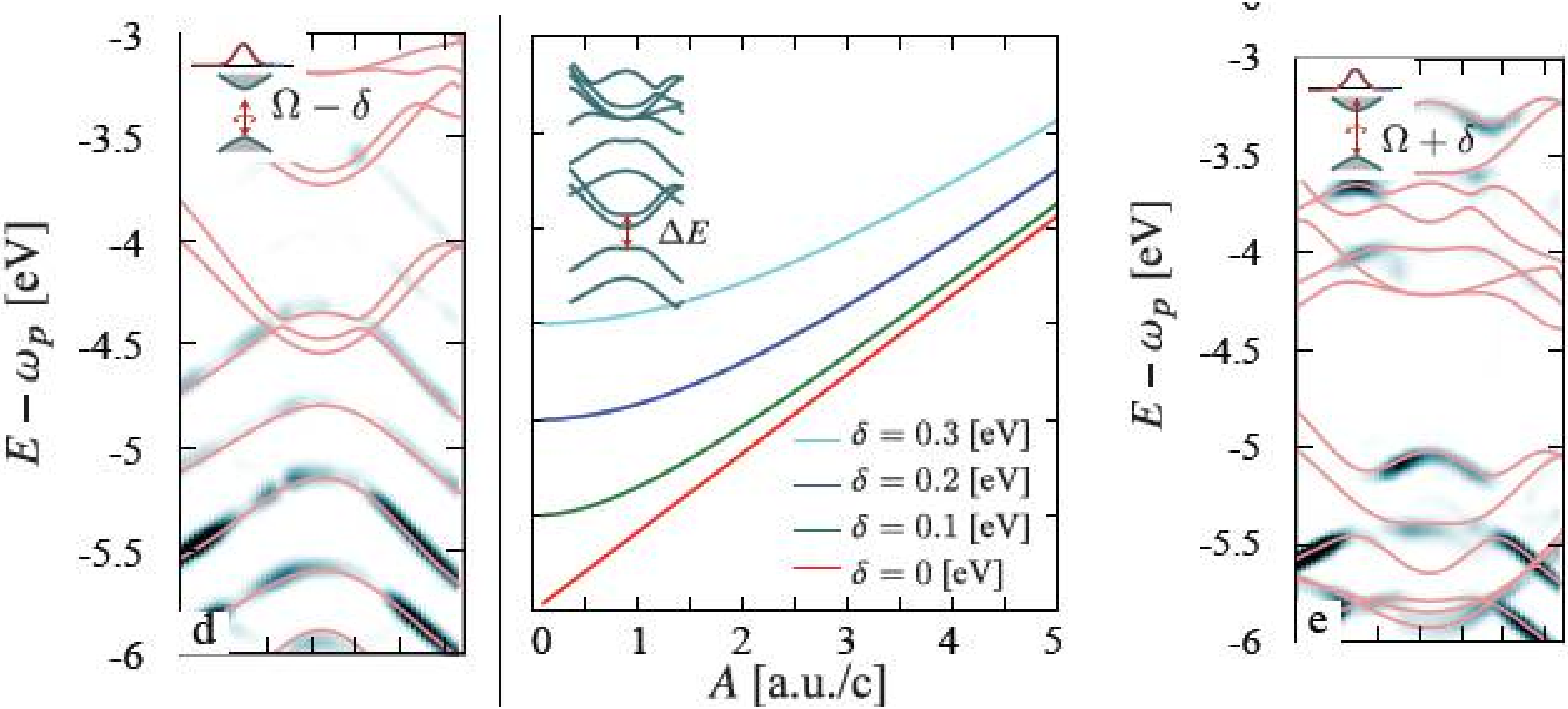




Simulating real-time photon-dressing of quasiparticles with TD-ARPES in TMDs

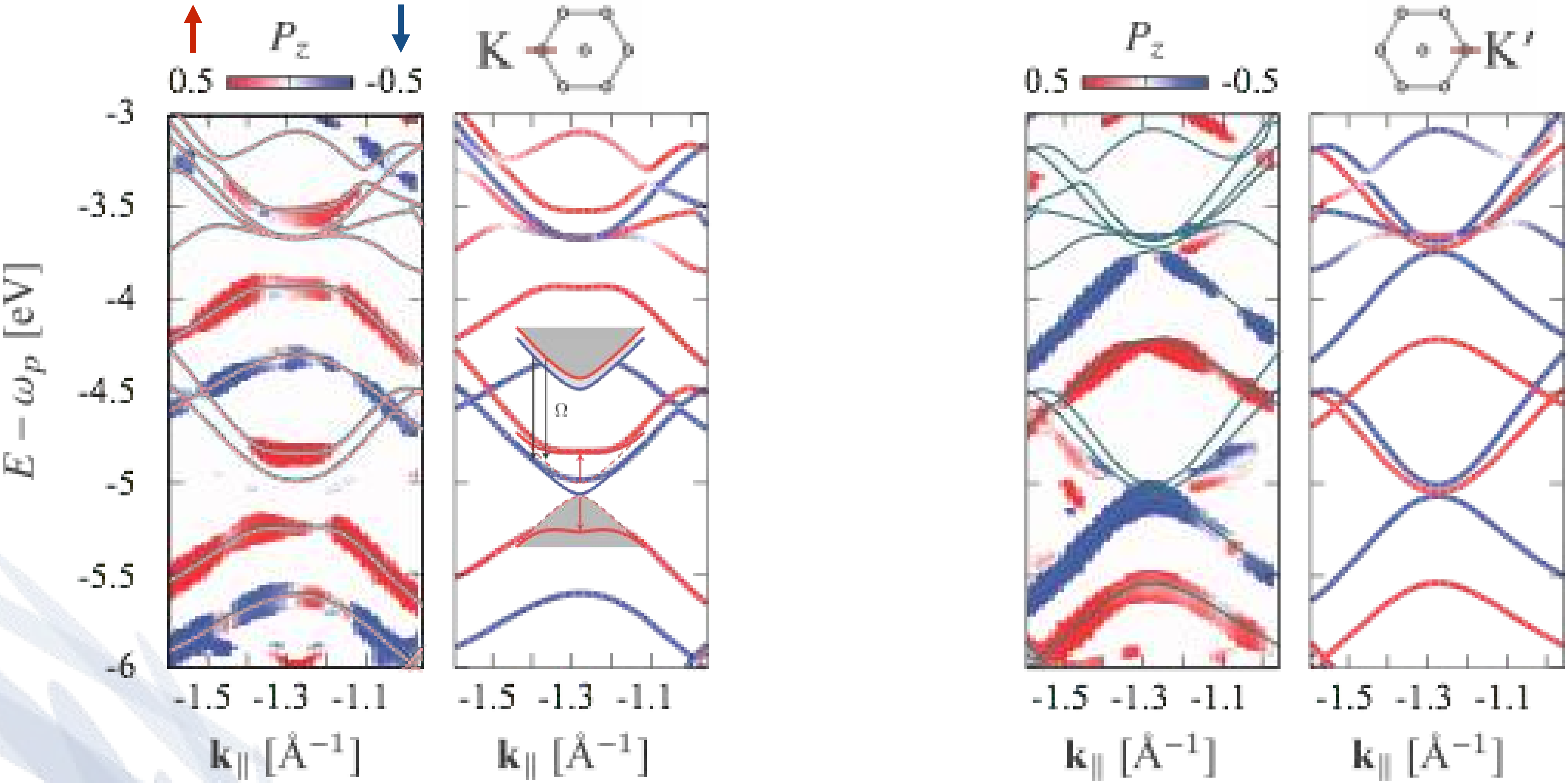
Dependence on the laser frequency and intensity

off-resonant case



U. de Giovanninni, H. Hübener, AR NanoLetters (2016)

Photon dressing and undressing
Spin texture



Dichroic optical Stark effect

U. de Giovanninni, H. Hübener, AR (2016)

How real is all this??

E. J. Sie, N. Gedik et al. (MIT,unpublished)

New Theoretical Developments:
QED-TDDFT

"bridging quantum optics and materials modeling"

QED-Materials

"New states of matter"

QED-Chemistry:

Modifying Chemical Landscapes
by Coupling to Vacuum Fields

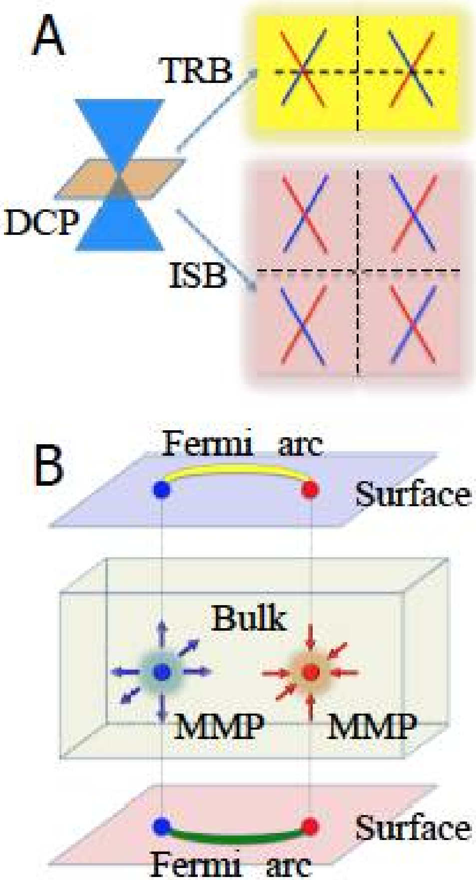
M. Ruggenthaler, J. Flick, C. Pellegrini, H. Appel, I.V. Tokatly, AR PRA (2014),
J. Flick, M. Ruggenthaler, H. Appel, AR PNAS (2015) and PNAS (2017)

Light-induced topological states

Discovery of a Weyl Fermion semimetal

Weyl semimetal in crystalline TaAs, TaP, $SrSi_2$
NbAs, NbP

B.Q. Lv et al, Phys. Rev. X (2015),
S-X, Xu et al, Science (2015)
Z.K. Liu, et al, Nat. Comm. (2015)

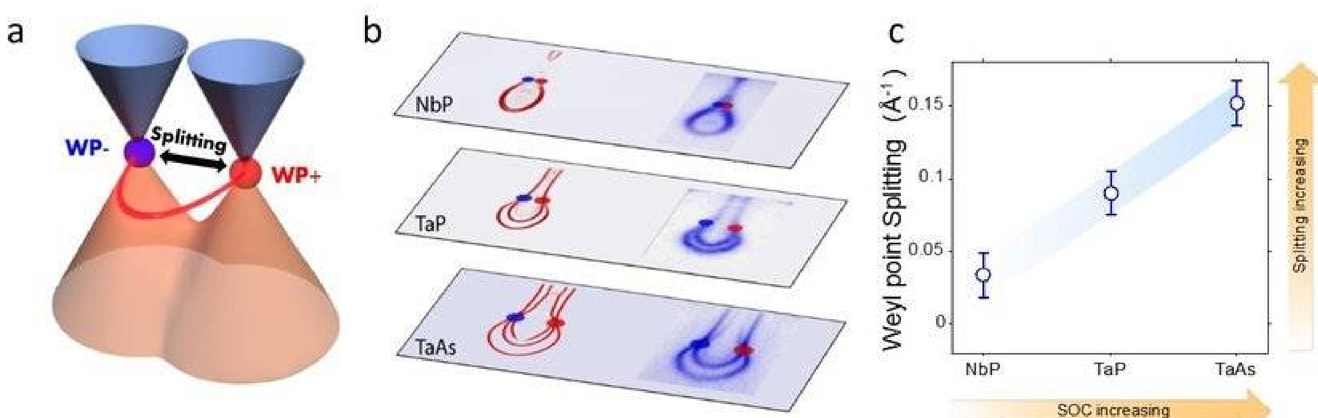


Dirac cones: two overlapping Weyl nodes with opposite chirality

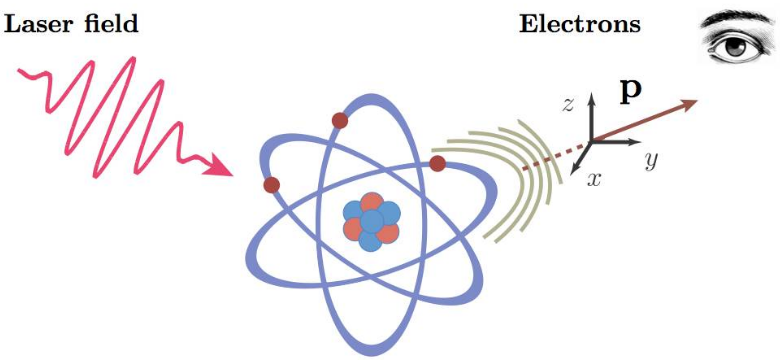
Weyl fermions: exotic properties,
monopoles in crystal momentum space
chiral anomaly: negative magneto-resistance, chiral magnetic effects and anomalous Hall effect

Separated Weyl nodes are topologically stable:
a perturbation respecting the translational symmetry can only shift but not annihilate them

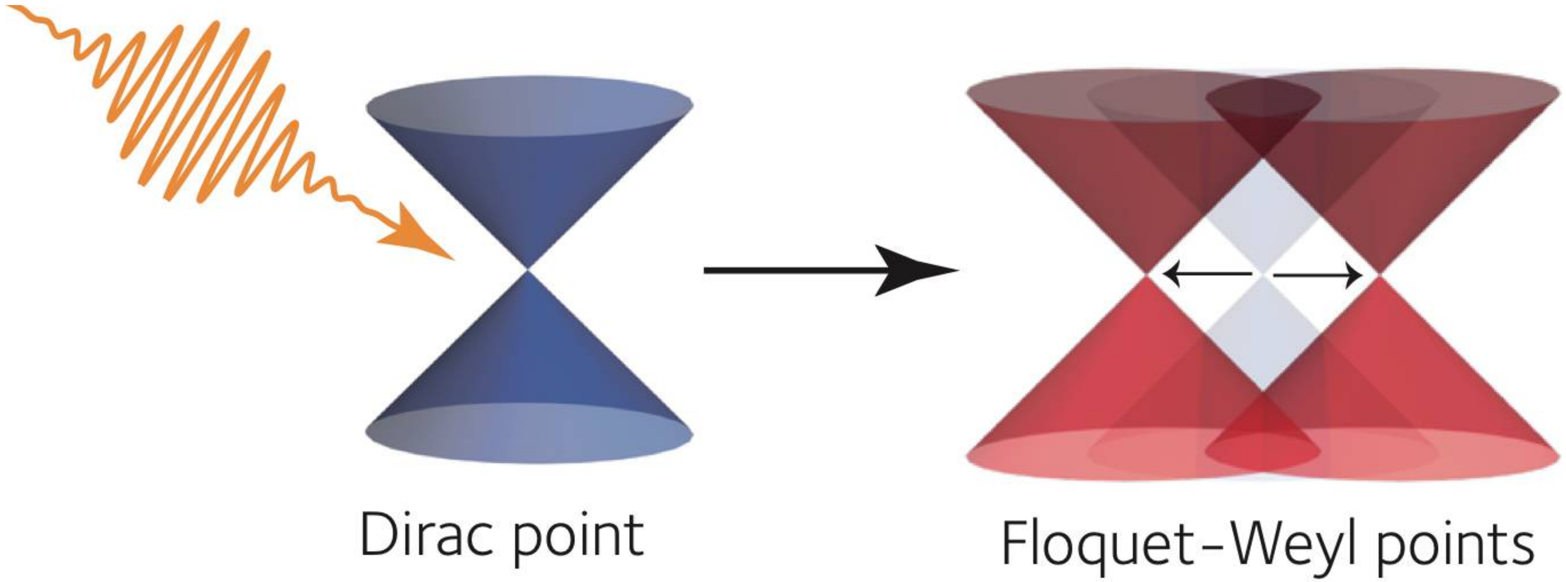
Weyl semimetals show characteristic Fermi arcs on the surface



TD-ARPES and transient absorption spectroscopies
Light induced Floquet Topological states

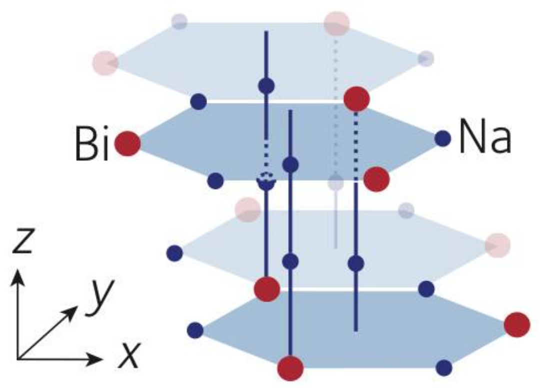


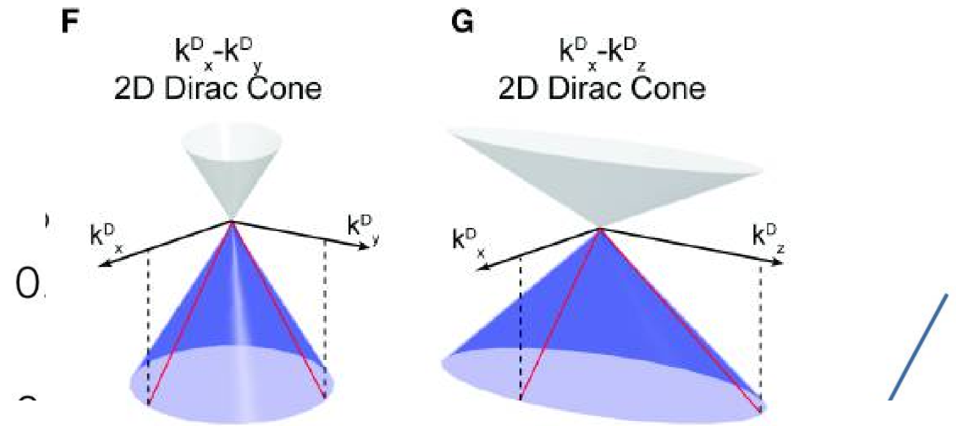
$$\hat{H}_{\mathrm{Dirac}}(\mathbf{k}-\mathbf{A}(t)) = \begin{pmatrix} \hat{H}_{\mathrm{Weyl}}(\mathbf{k}-\mathbf{A}(t)) & 0 \\ 0 & \hat{H}_{\mathrm{Weyl}}(\mathbf{k}-\mathbf{A}(t))^* \end{pmatrix} \quad \hat{H}_{\mathrm{Weyl}}(\mathbf{k}-\mathbf{A}(t)) = \begin{pmatrix} k_z - A_0\sin(\Omega t) & k_x - i(k_y - A_0\cos(\Omega t)) \\ k_x + i(k_y - A_0\cos(\Omega t)) & -k_z + A_0\sin(\Omega t) \end{pmatrix}$$

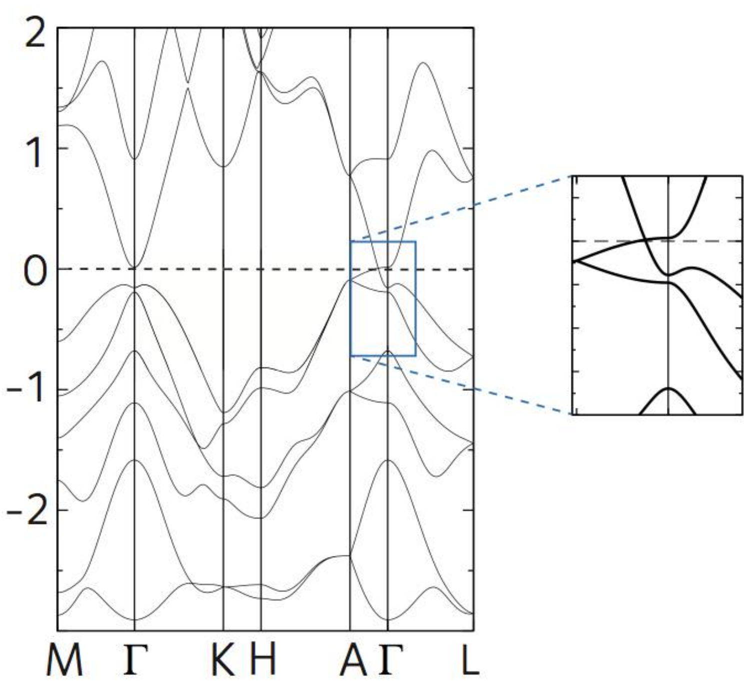


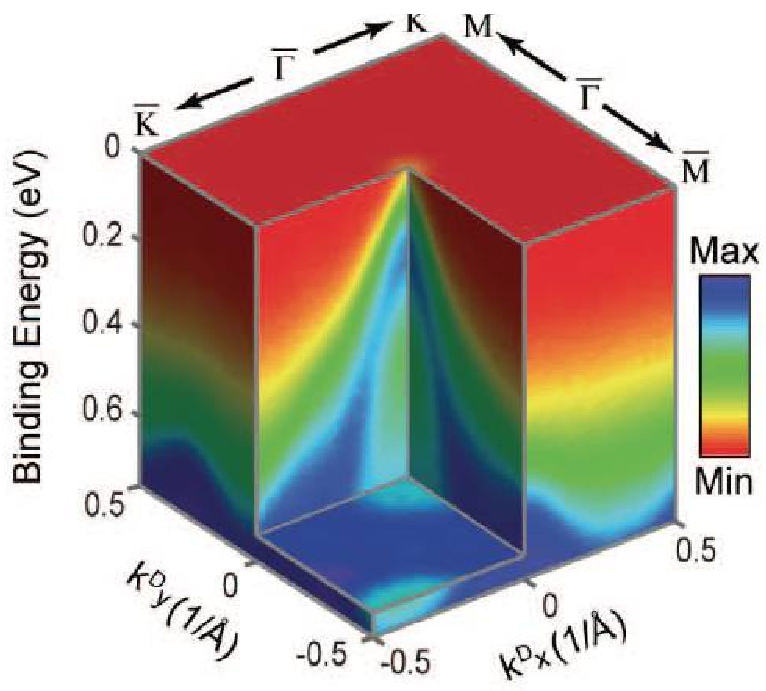
H. Hübener, M. Sentef, A. F. Kemper, U. de Giovannini, AR Nature Comm. (2017)
U. de Giovannini, H. Hübener, AR NanoLetters (2016)

Light-induced Floquet topological states
3D real Dirac material Na_3Bi







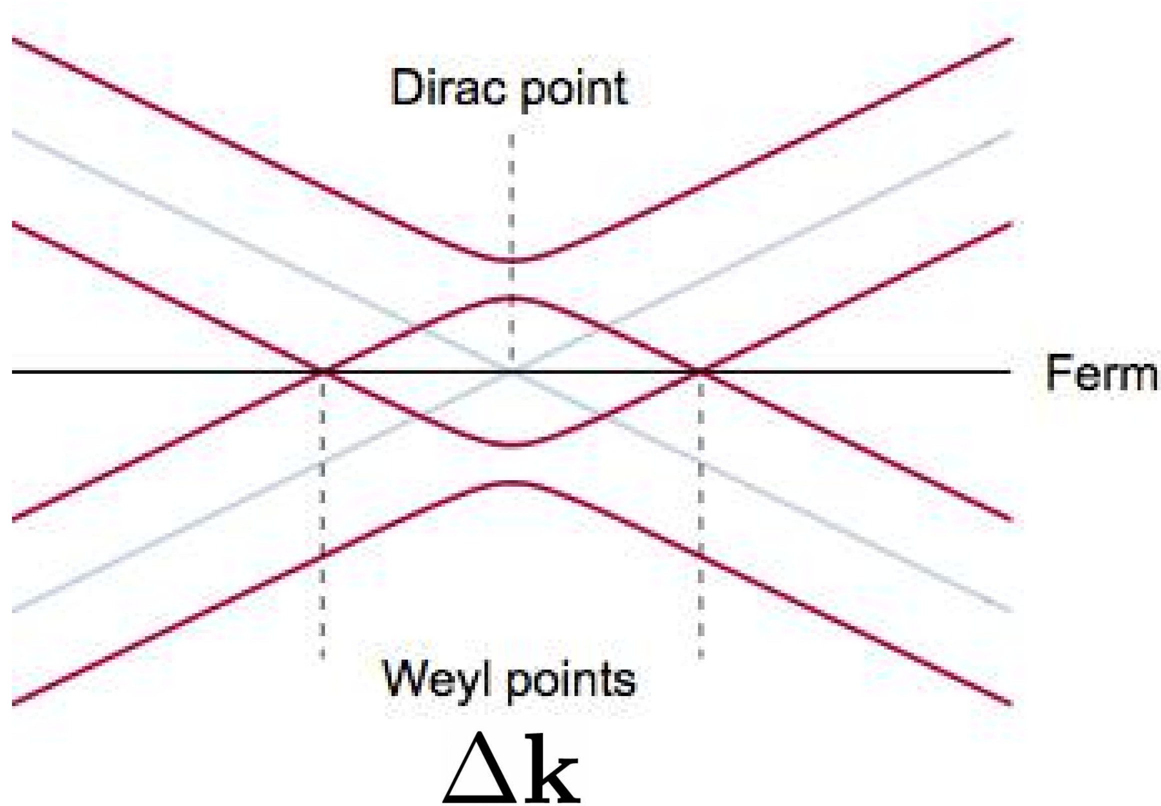


Z. K. Liu et al., Science 343, 864 (2014)

Light-induced Floquet topological states

3D Dirac material Na_3Bi

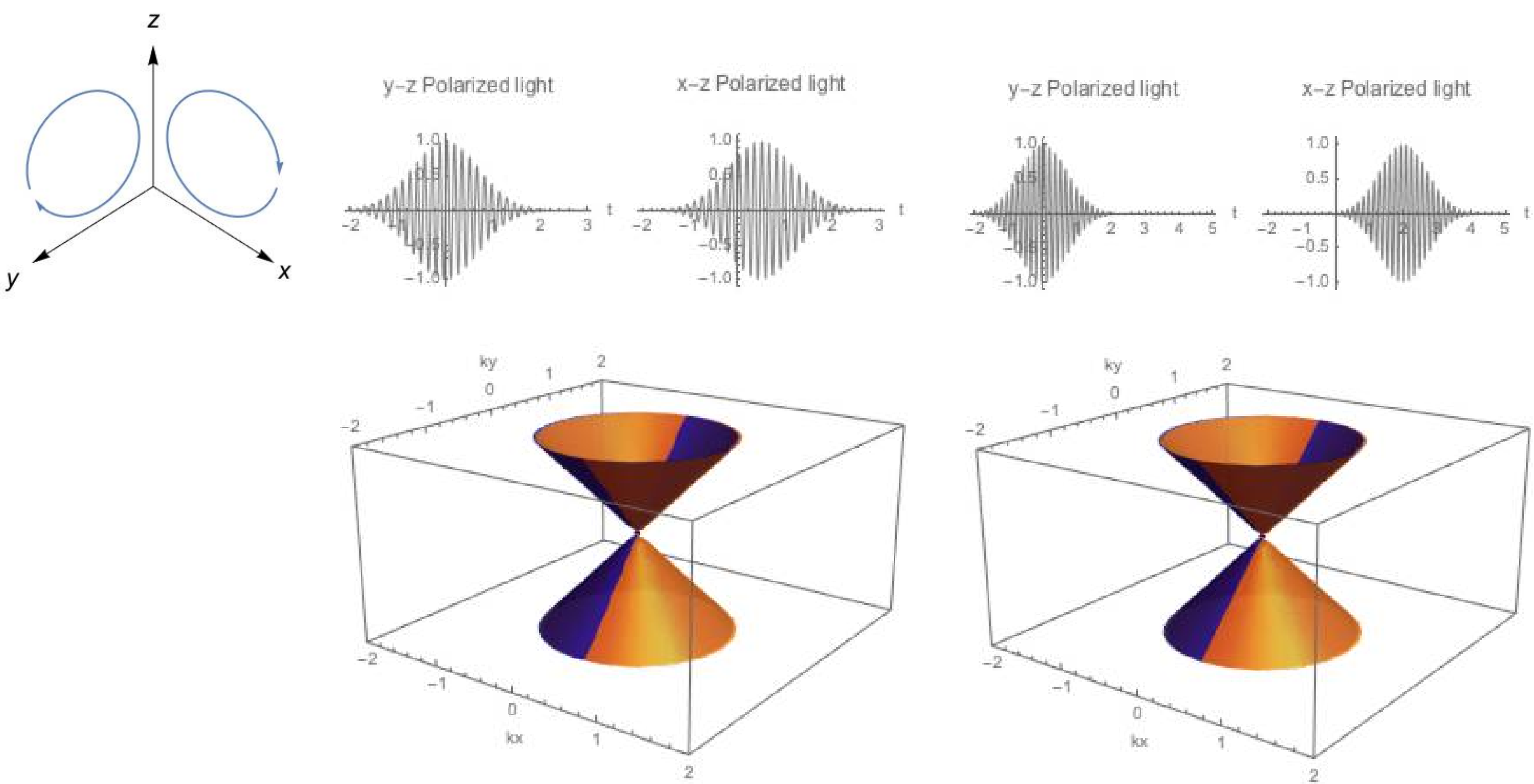
$$\Delta \mathbf{k} = 2 v_{\mathrm{F}} \frac{(\frac{e}{c} A)^2}{\Omega}$$



Engineering of fictitious gauge fields with real laser fields laser control of topological states of matter

H. Hübener, M. Sentef, A. F. Kemper, U. de Giovannini, AR   Nature Communications (2017)

Light-induced Floquet topological states
3D Weyl semimetal Na_3Bi



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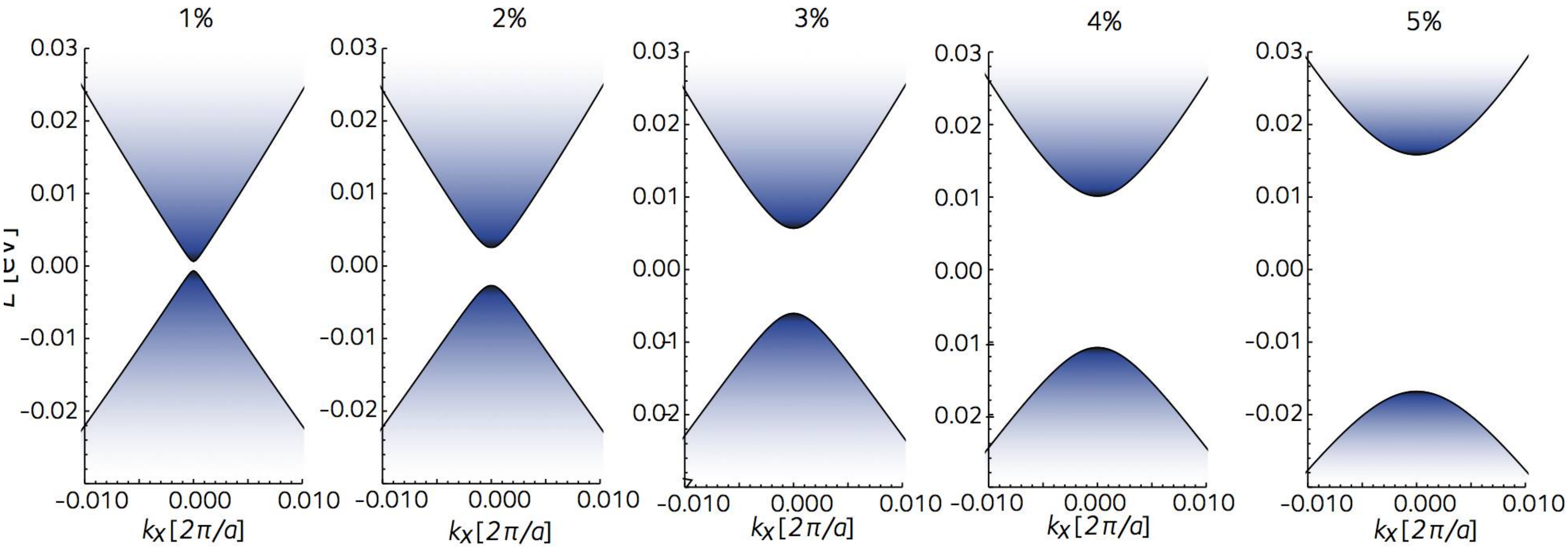
Light-induced Floquet topological states
3D Dirac material Na_3Bi

Topological protection

- apply strain



- topological insulator

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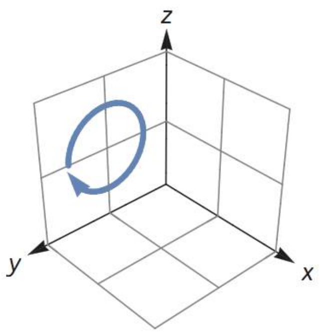
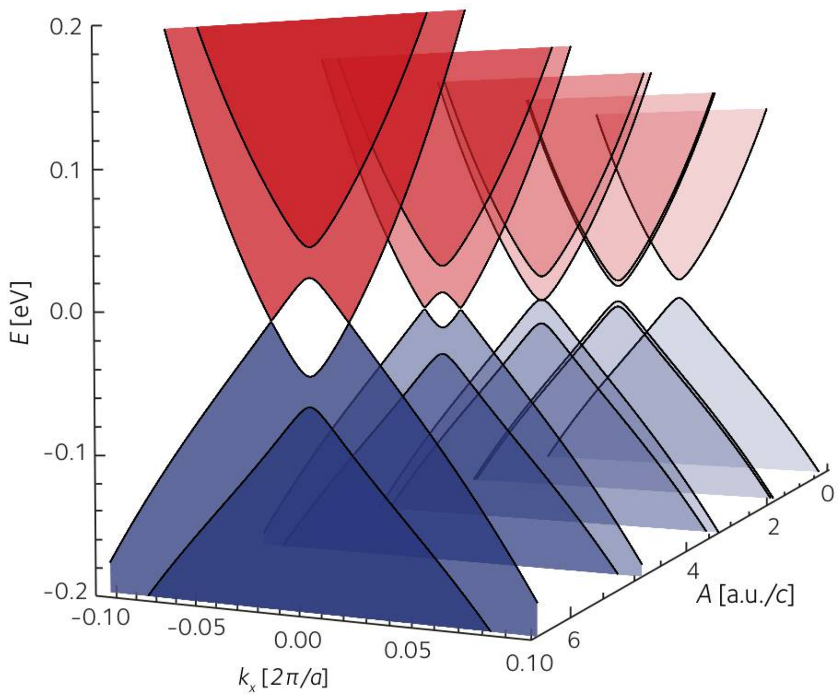
Light-induced Floquet topological states
3D Dirac material Na_3Bi

mpsd

Topological protection





- Weyl semimetal restored

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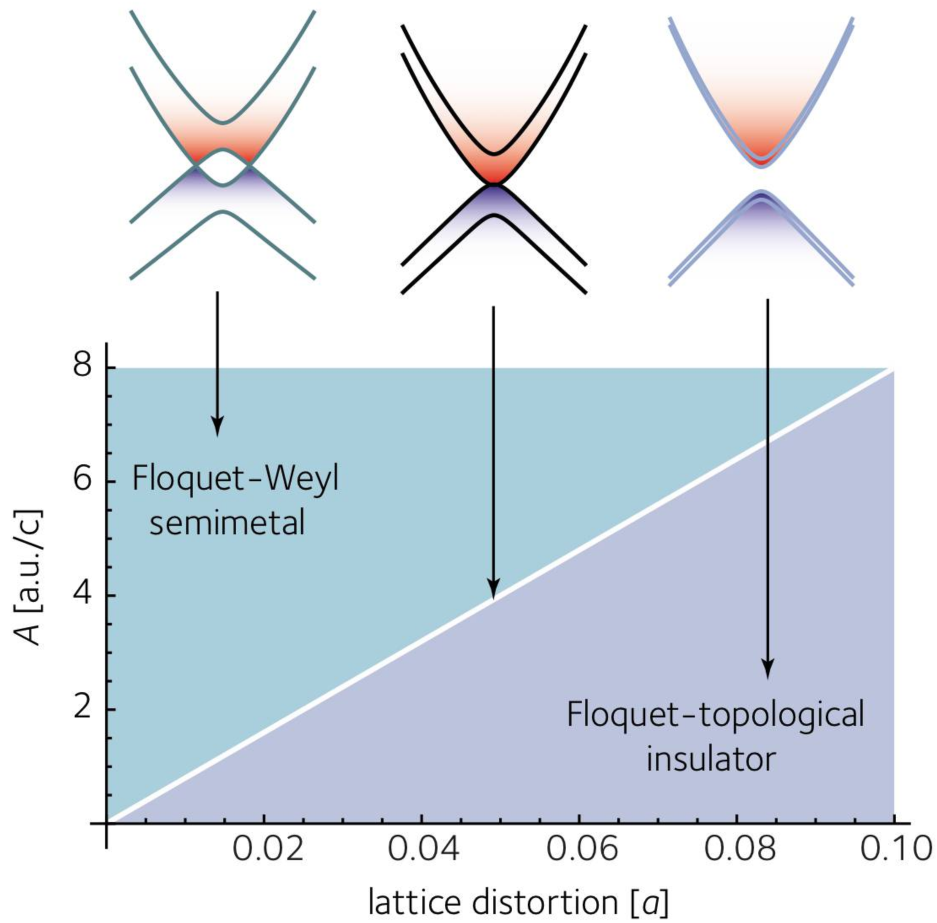
Light-induced Floquet topological states
3D Dirac material Na_3Bi

Floquet phase diagram



H. Hübener, M. Sentef, A. F. Kemper, U. de Giovannini, AR (Nature Communications 2016)

Acknowledgements

mpsd

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Experiments

FHI-group

Ralph Ernstorfer







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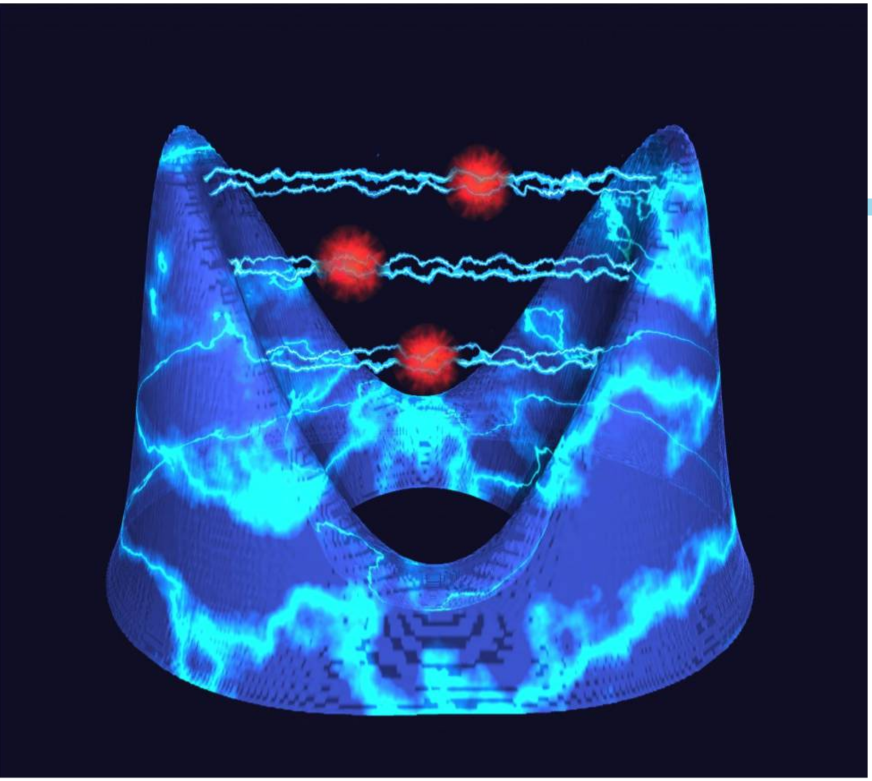
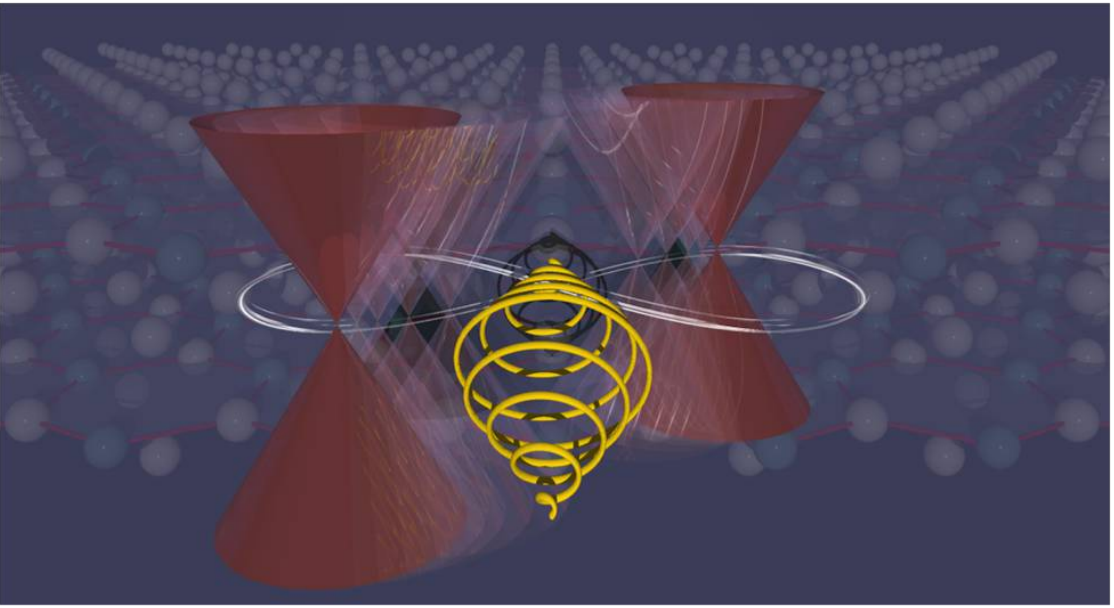


Thank You



Publications: (see group webpage)

J. Flick, M. Ruggenthaler, H. Appel, AR, PNAS (2015); and PNAS (2017)
H. Hübener, M. Sentef, A. F. Kemper, U. de Giovannini, AR Nature Comm. (2017)
U. de Giovannini, H. Hübener, AR NanoLetters (2016)
M. Ruggenthaler, J. Flick, C. Pellegrini, H. Appel, I.V. Tokatly, AR PRA (2014); PRL (2015)