U.S. DEPARTMENT OF ENERGY | Office of Science

PULSE

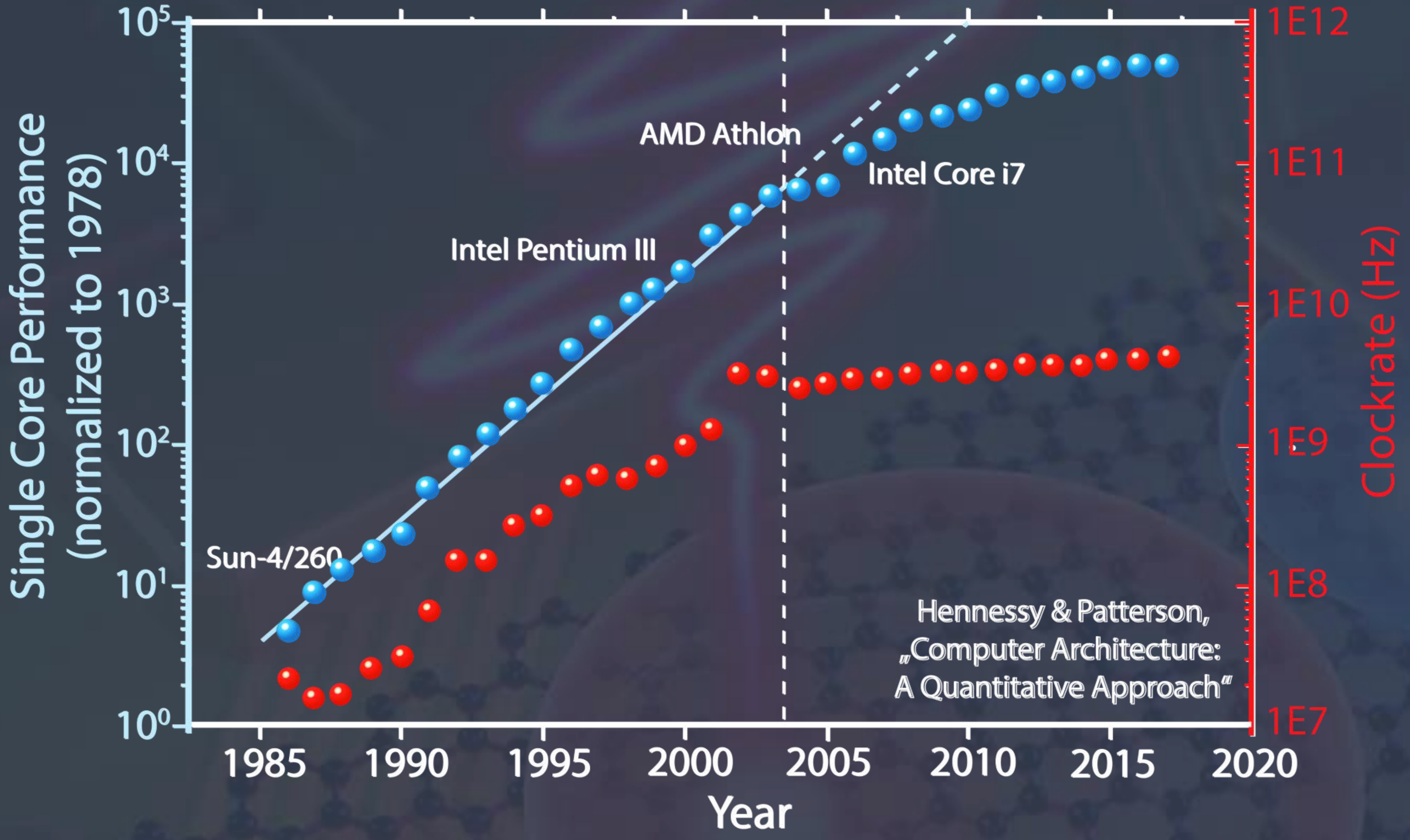
Steering Electrons with Light Fields:
from Ultrafast Electronics to Quantum Sensing

02/02/2023
SLAC
Panofsky Fellowship Seminar

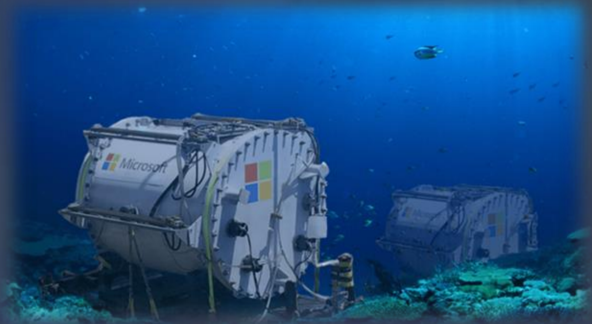
cheide@stanford.edu

Christian Heide
Stanford PULSE Institute

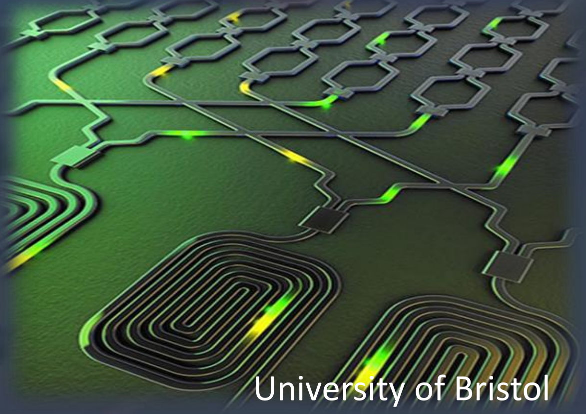
Can optics clear the CMOS bottleneck?



Data center at Microsoft



Silicon Photonics



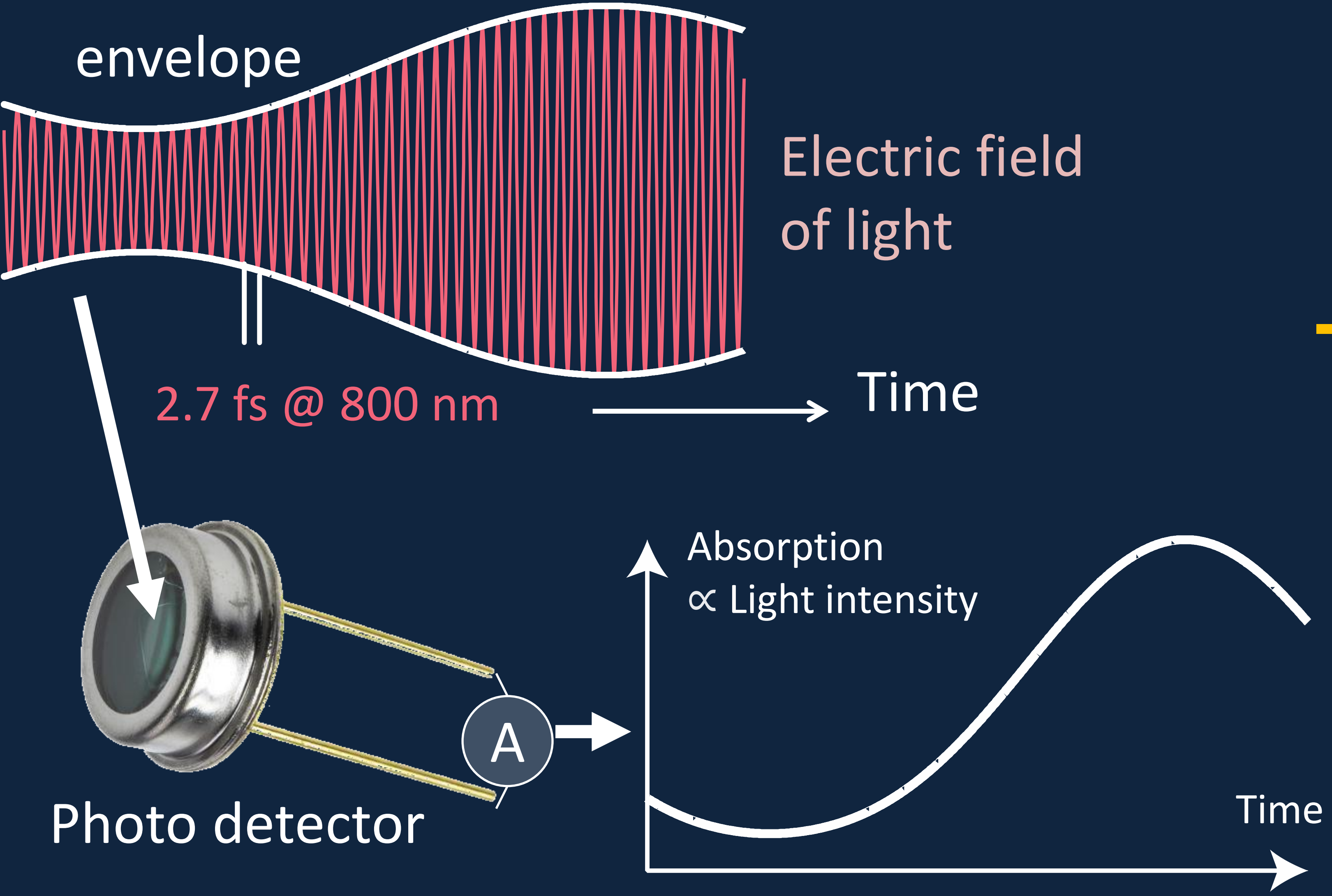
Field-driven dynamics
(non-perturbative)

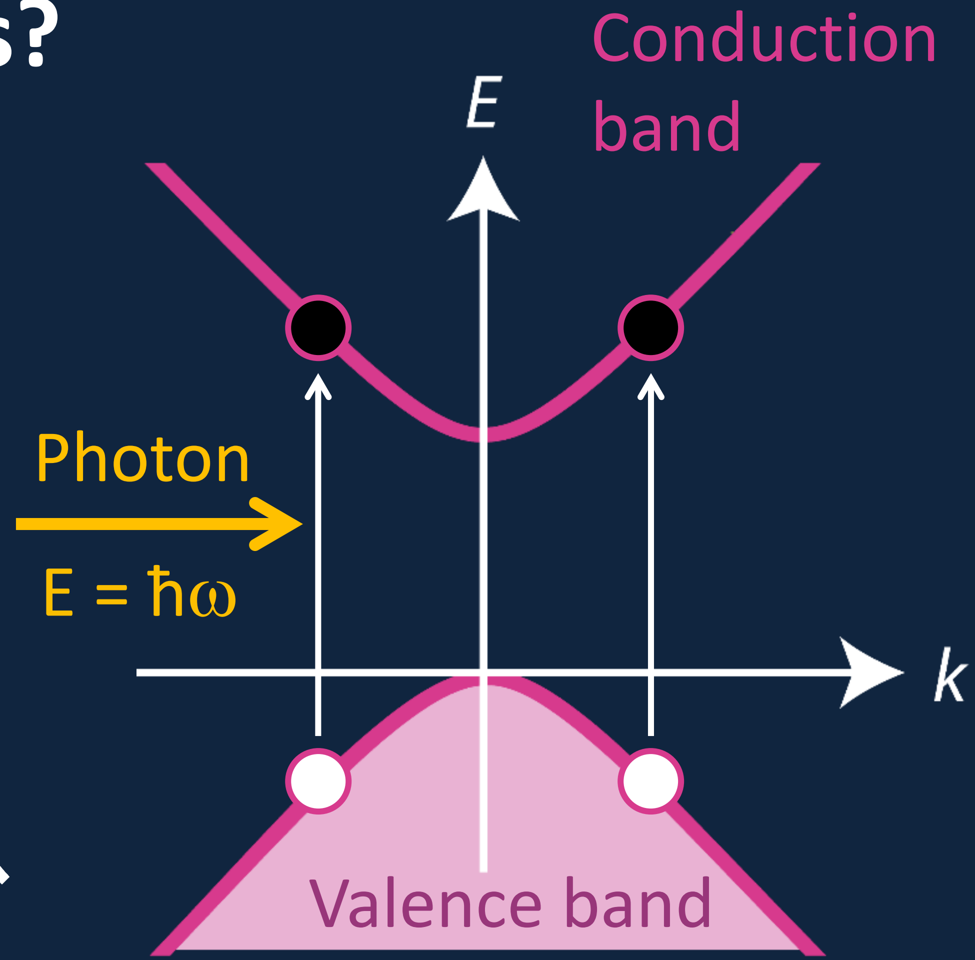
Photon-based
(Perturbative)
absorption

Light-field-driven (quantum) electronics

1 GHz (10^9 Hz)	1 THz (10^{12} Hz)	1 PHz (10^{15} Hz)
1 ns	1 ps	1 fs

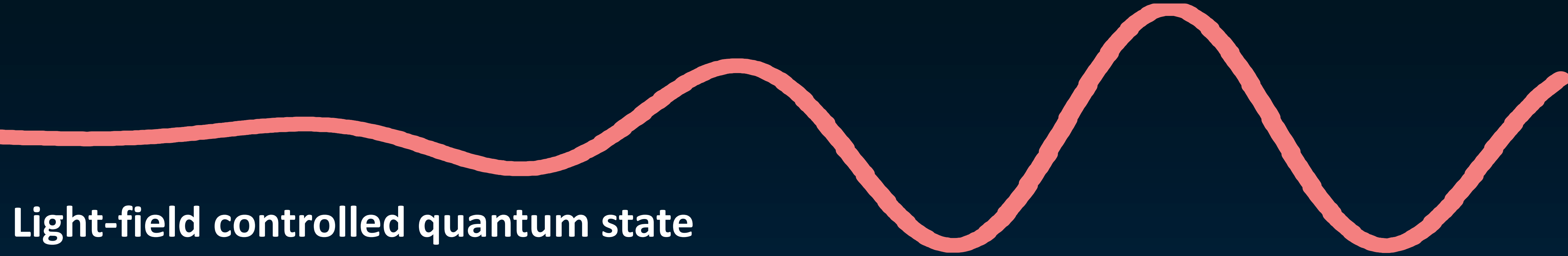
How does light interact with solids?



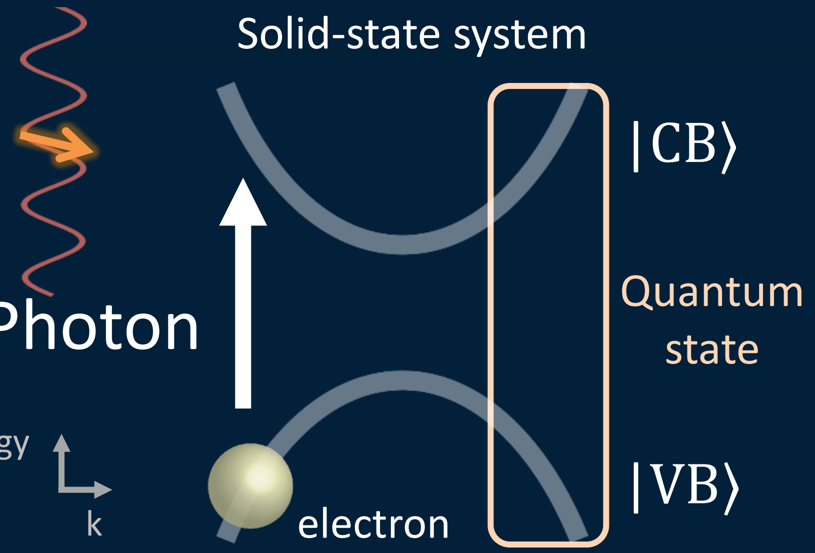


Good description when the light field is weak (perturbative)

Light-field controlled Electron dynamics



Light-field controlled quantum state



Time scale: 1 fs = 1×10^{-15} s

Is this state **coherent** and **observable**?

Ultrafast electronics
Ultrafast coherent spectroscopy

Electron wavevector

$$\dot{\mathbf{k}}(t) = \hbar^{-1} e\mathbf{E}(t)$$

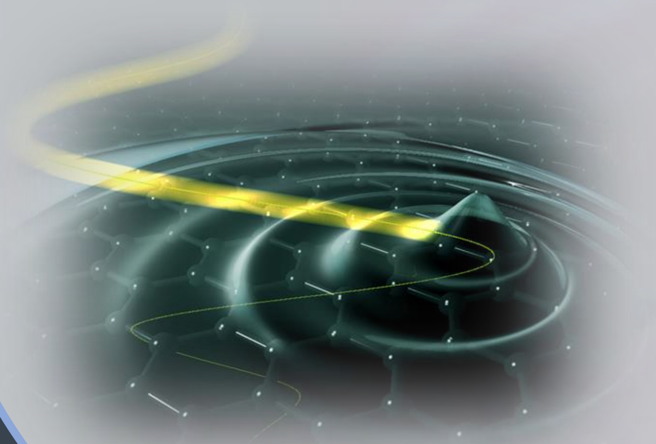
Relevant for $E_0 \sim$ V/nm

F. Bloch, Zeitschrift f. Physik 52, 555 (1928), C. Zener, Proc. R. Soc. A 145, 523 (1934)

Steering Electrons with Light Fields

Coherent Control

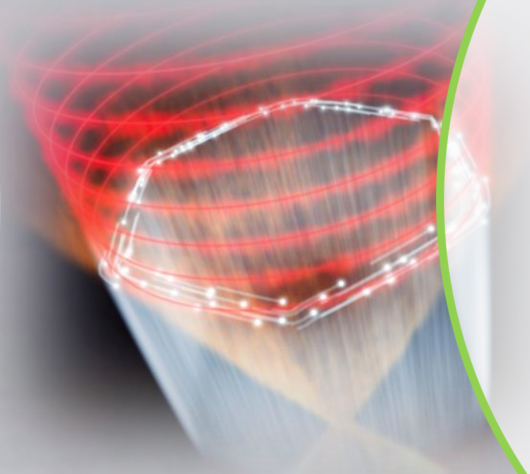
Driving electrons coherently inside of solids



Light

Material Science

Material properties on demand



Matter

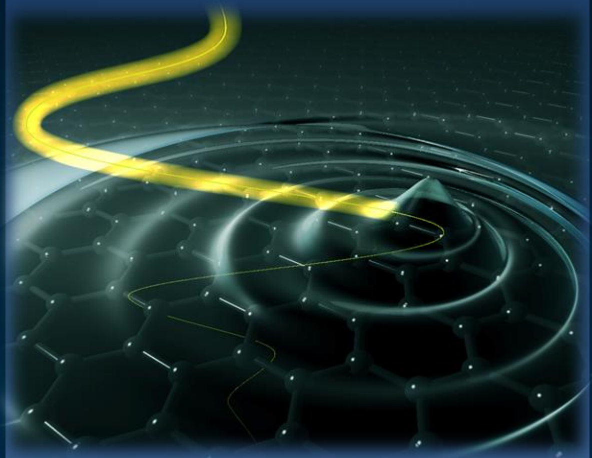
Attosecond Science

X-Ray probing of coherent electron motion

Probe

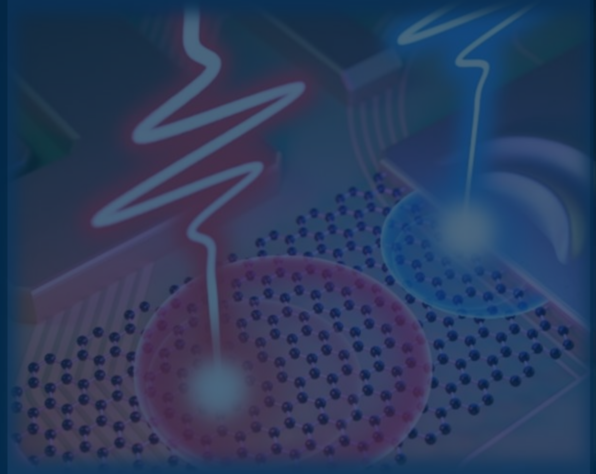
Playground: Light-field driven electron dynamics in solids

Light-field driven quantum dynamics



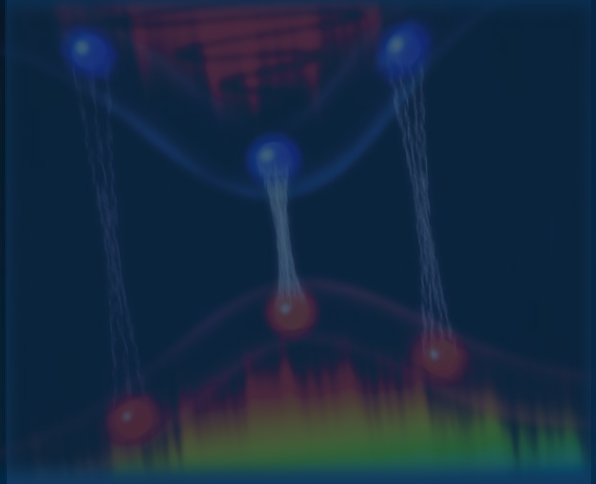
CH et al., **Nano Letters** 21, 9403 (2021)
CH et al., **PRL** 121, 207401 (2018)
TH, **CH** et al. **Nature** 550, 224 (2017)

Light-field driven dynamics at interfaces



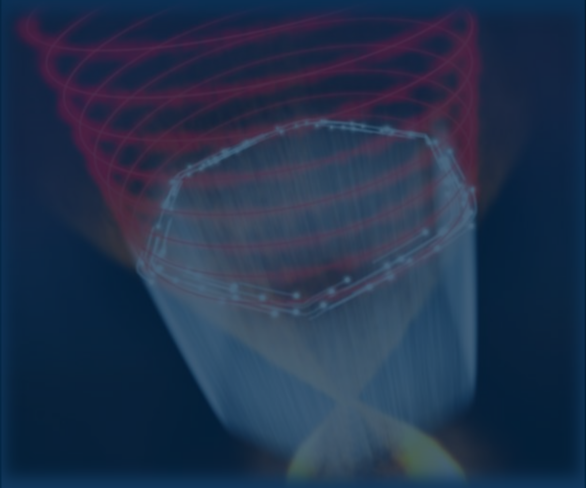
TB, **CH** et al., **Nature** 605, 251 (2022)
CH et al. **Nature Photon.** 14, 219 (2020)

Role coherence



CH, YK et al., **Optica**, 9, 5 (2022),
CH et al., **Nano Letters** 21, 22 (2021)

Topologically protected materials



CH et al., **Nature Photonics** 16, 620 (2022)

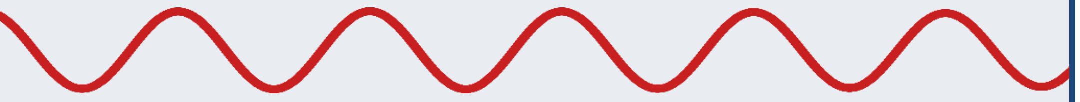
Light-field driven current

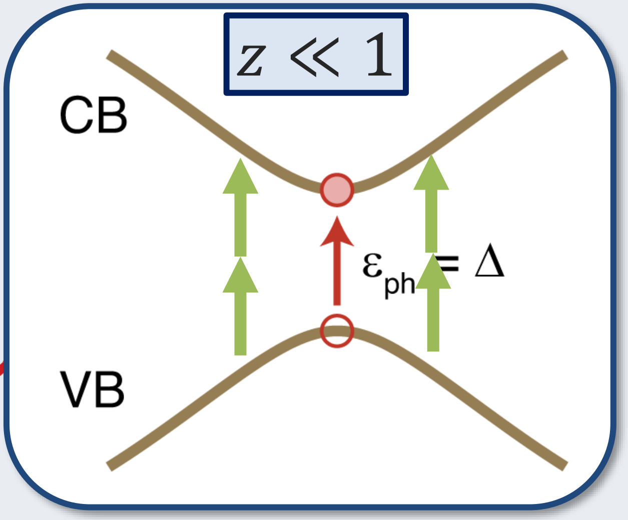
High-harmonic generation

Key ingredients of light-field driven electron dynamics

Linear and nonlinear response (multi-photon driven)

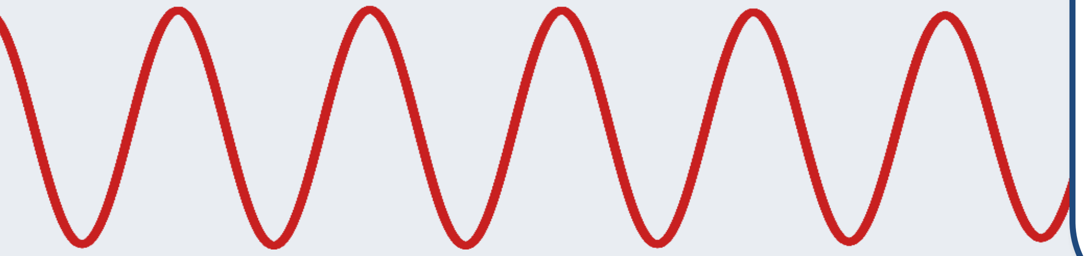
- Resonant absorption
- Linear and non-linear optics

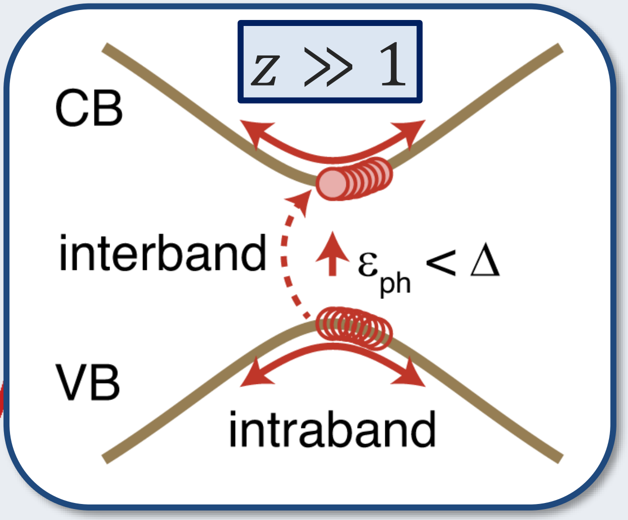




Optical-field-driven non-perturbative sub-cycle dynamics

- Non-resonant absorption
- Inter- & intraband motion





Polarization of the medium

$$P = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \cdots.$$

Non-perturbative intensity parameter

$$z = \frac{U_P}{E_{Ph}} = \frac{e v E_0}{\omega^2}$$

$z = 1$;
E_0 ~ V/nm

Heide et al., PRA 104, 023103 (2021)
Corkum & Krausz, Nat. Phys. **3**, 381 (2007)
Krausz & Ivanov, Rev. Mod. Phys. **81**, 163 (2009)
Krausz & Stockman, Nat. Photon. **8**, 205 (2014)

Key ingredients of light-field driven electron dynamics

Envelope Carrier

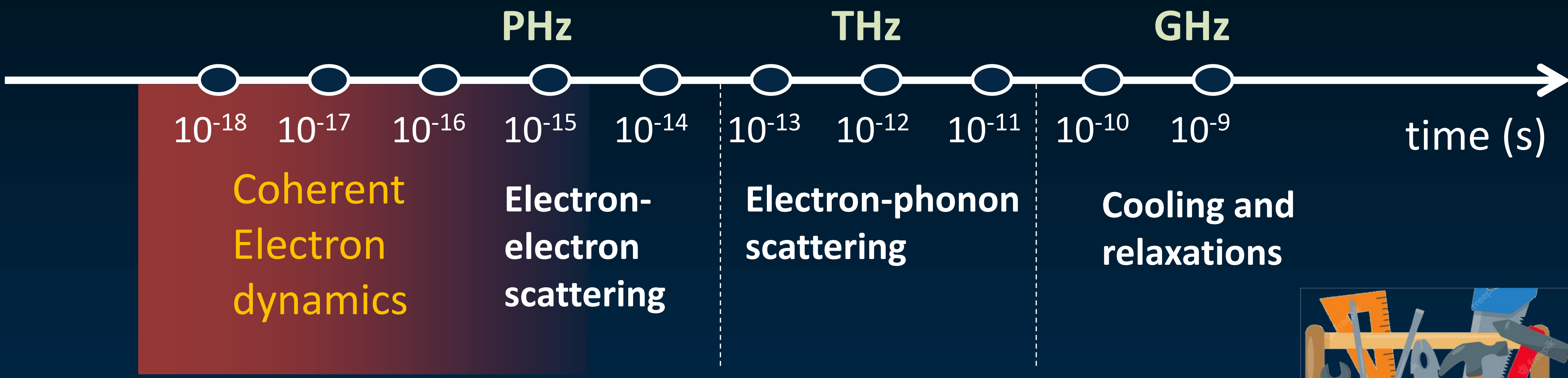
$$E(t) = f(t)\cos(\omega t + \varphi_{\mathrm{CE}})$$

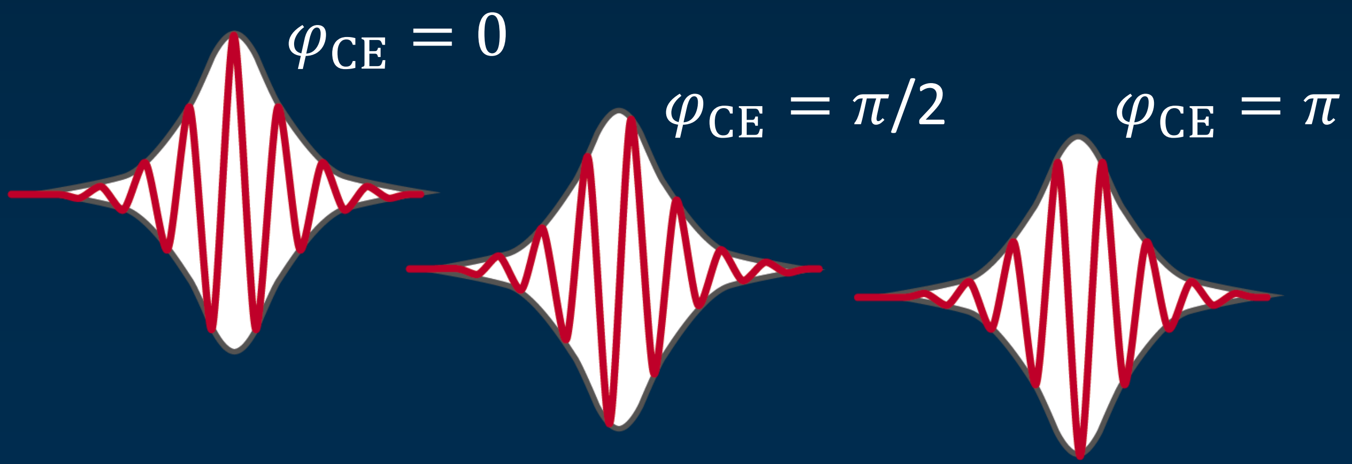
Carrier-envelope phase (CEP)

Attosecond timing control:
Phase stability $\Delta\varphi_{\mathrm{CE}} = 20$ mrad
corresponds to 8 as time delay (@ 800 nm)
(8 as = 8 x 10^{-18}s = 0.000000000000000008 s)

Review: Th. Udem, R. Holzwarth, T.W. Hänsch, **Nature 416**, 233-337 (2002)
T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000).

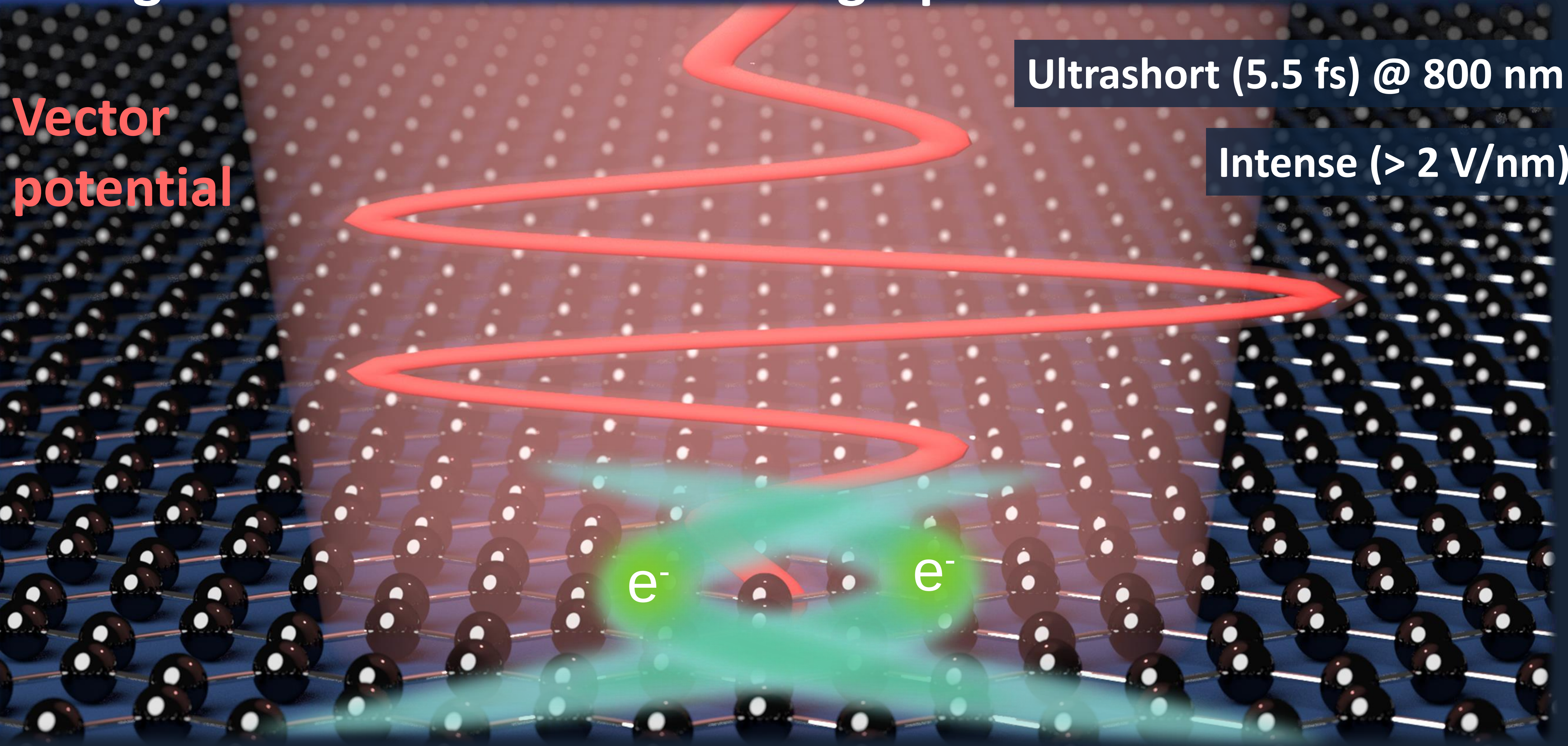
Key ingredients of light-field driven electron dynamics





Well-controlled optical waveforms oscillating at mid-IR frequencies (λ ~1-5 μm, E ~ V/nm)

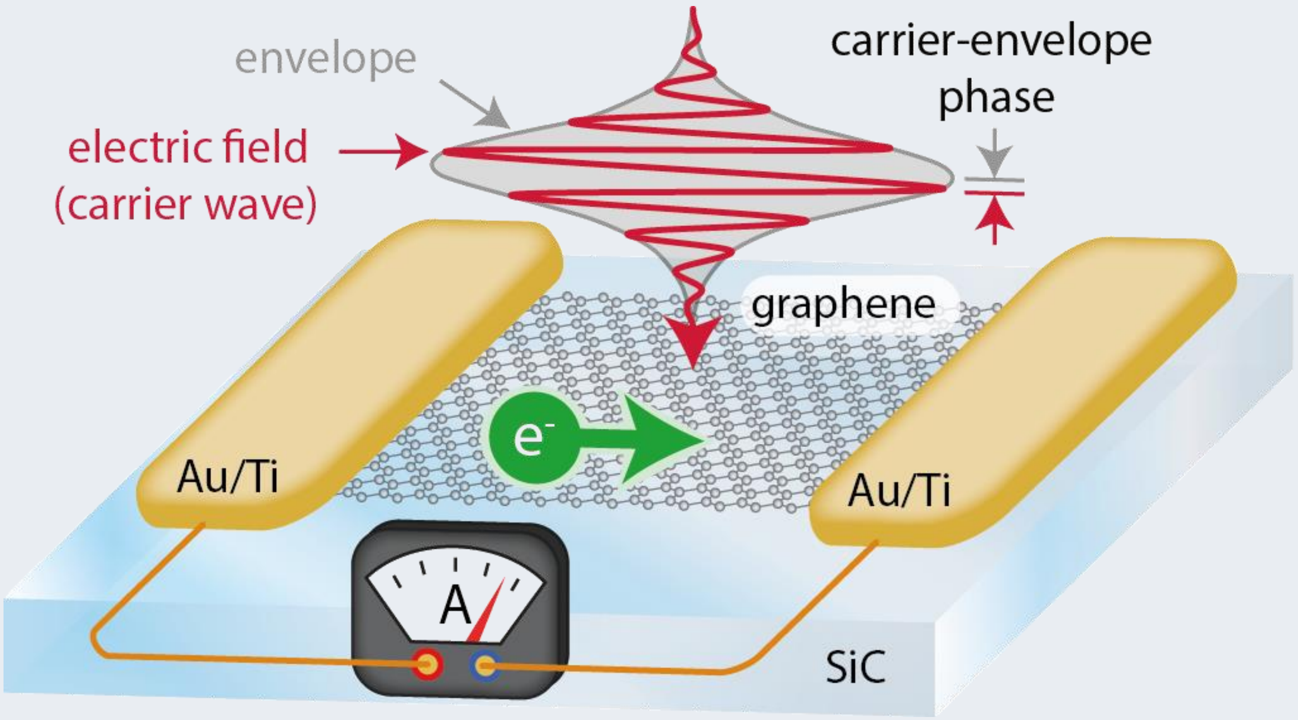
Light-field-driven currents in graphene

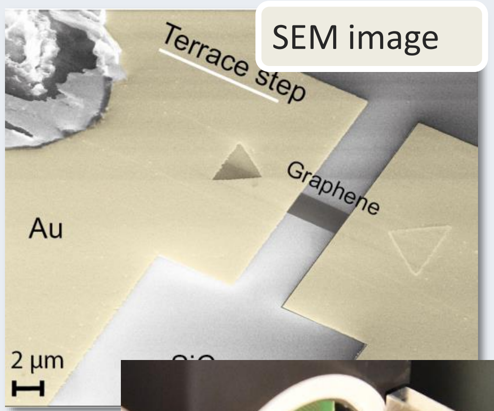


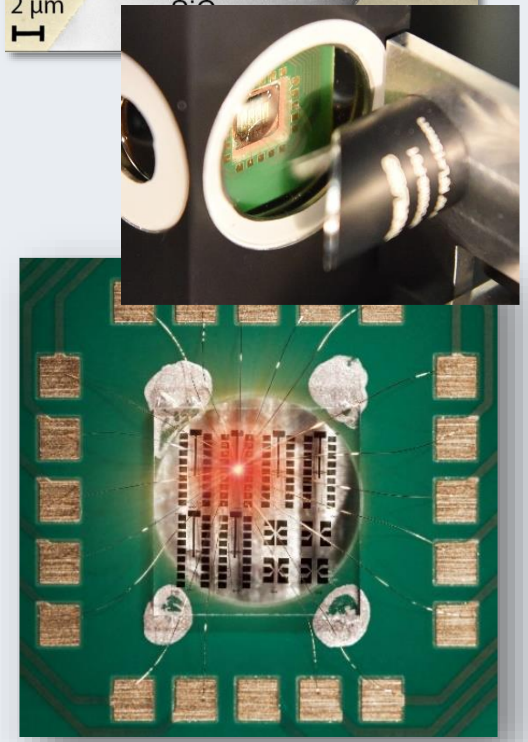
T. Boolakee*, C. Heide*, A. Garzon-Ramírez, HB Weber, I. Franco, P. Hommelhoff, **Nature** 605, 251 (2022)
T. Higuchi, C. Heide, K. Ullmann, HB Weber, P. Hommelhoff, **Nature** 550, 224 (2017)

CEP-dependent current generation in graphene

f_{rep} = 80 MHz, τ = 6 fs, λ_0 = 800 nm





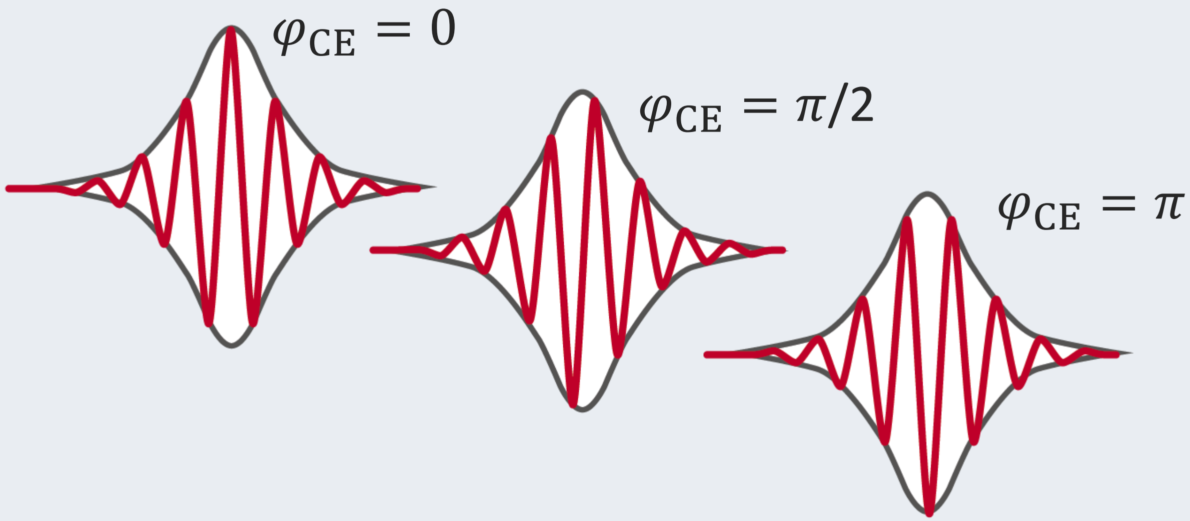


No external bias voltage

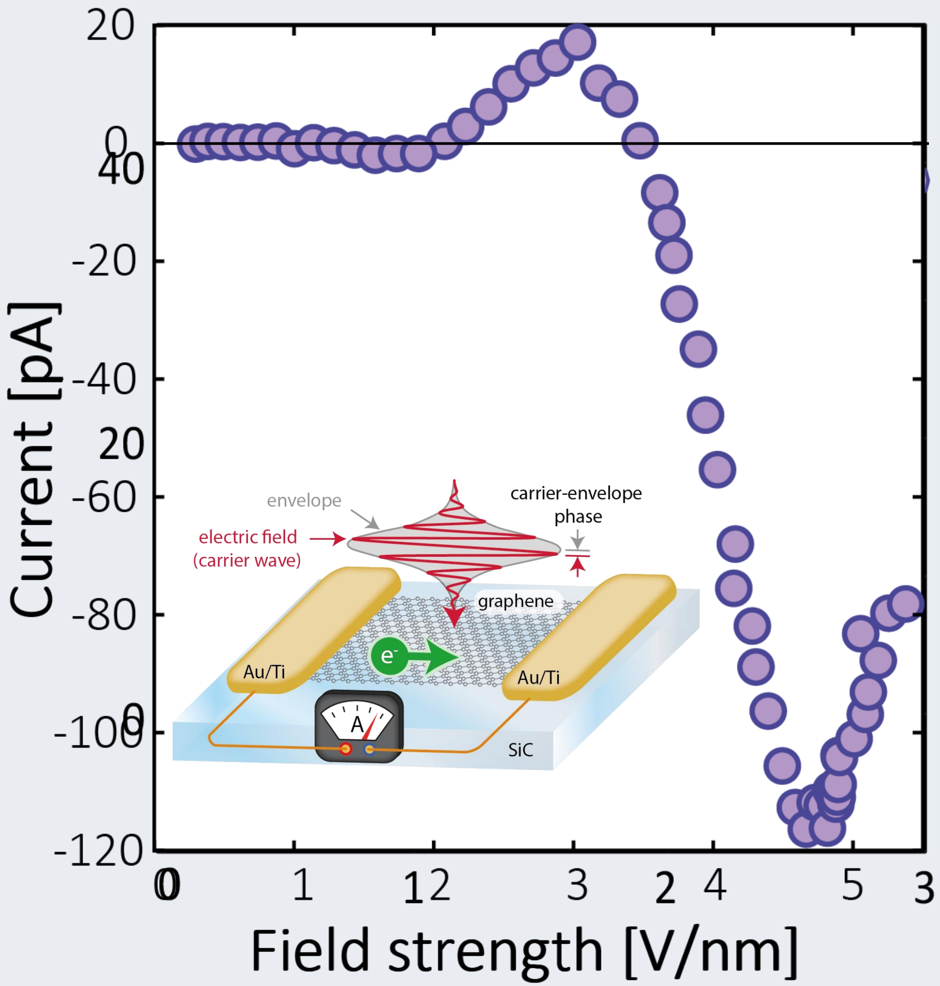
T. Boolakee*, C. Heide*, A. Garzon-Ramírez, HB Weber, I. Franco, P. Hommelhoff, **Nature** 605, 251 (2022)
C. Heide, T. Higuchi, H.B. Weber, P. Hommelhoff, **PRL** 121, 207401 (2018)
T. Higuchi, C. Heide, K. Ullmann, HB Weber, P. Hommelhoff, **Nature** 550, 224 (2017)

CEP-dependent photocurrent

Lock-in detection via modulating the carrier-envelope phase



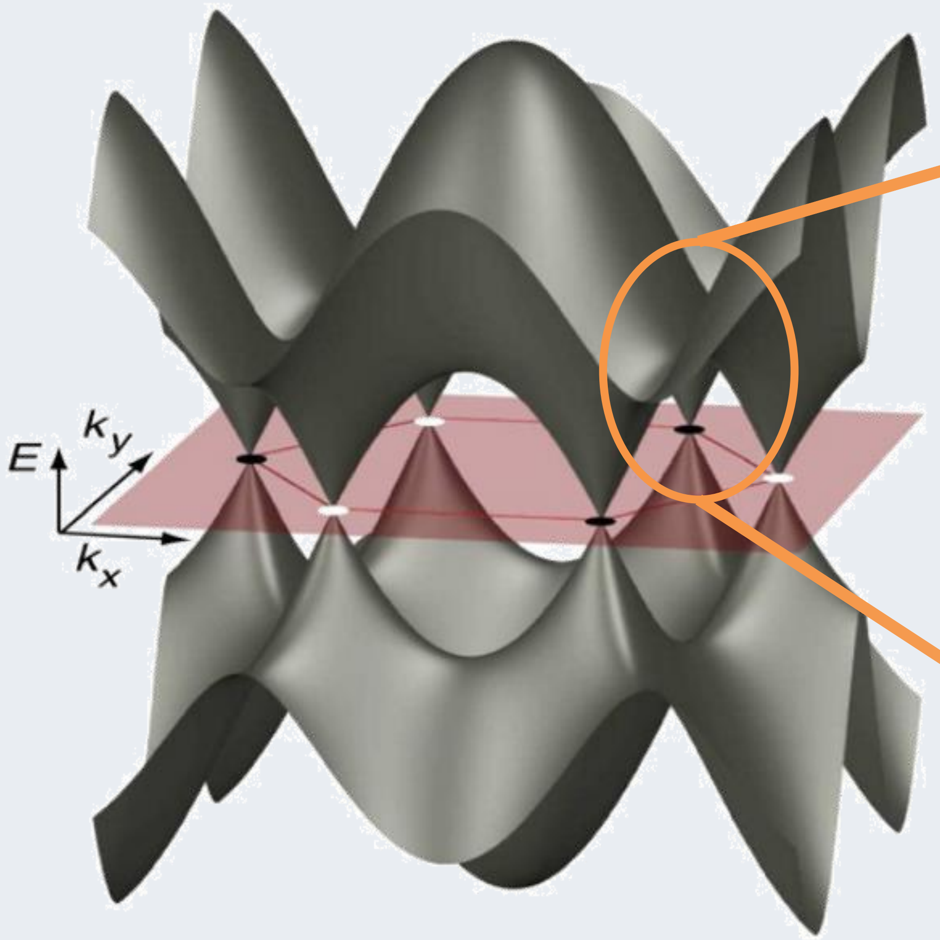
Current oscillates as a function of field strengths



Graphene‘s band structure

Momentum space

(tight-binding model)

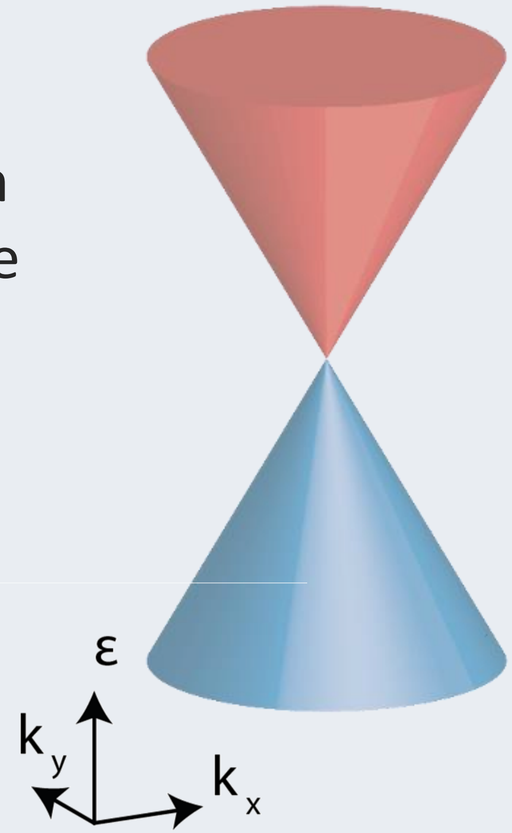


A. H. C. Neto *et al.*, Rev. Mod. Phys. 81, 109 (2009)

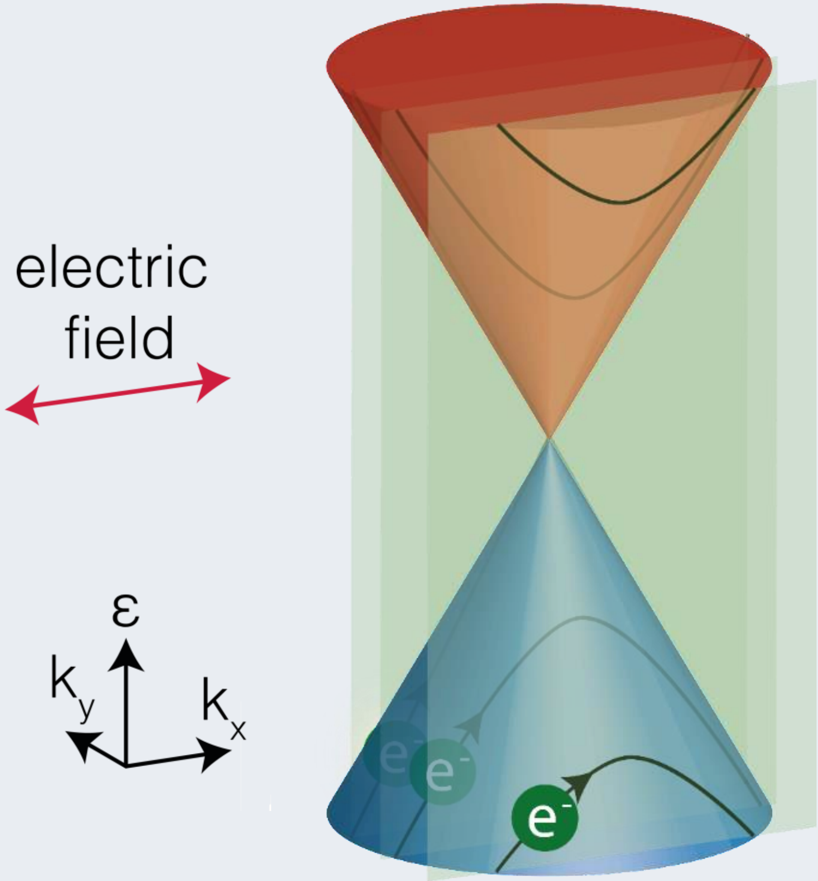
Dirac cone

(linear energy dispersion)

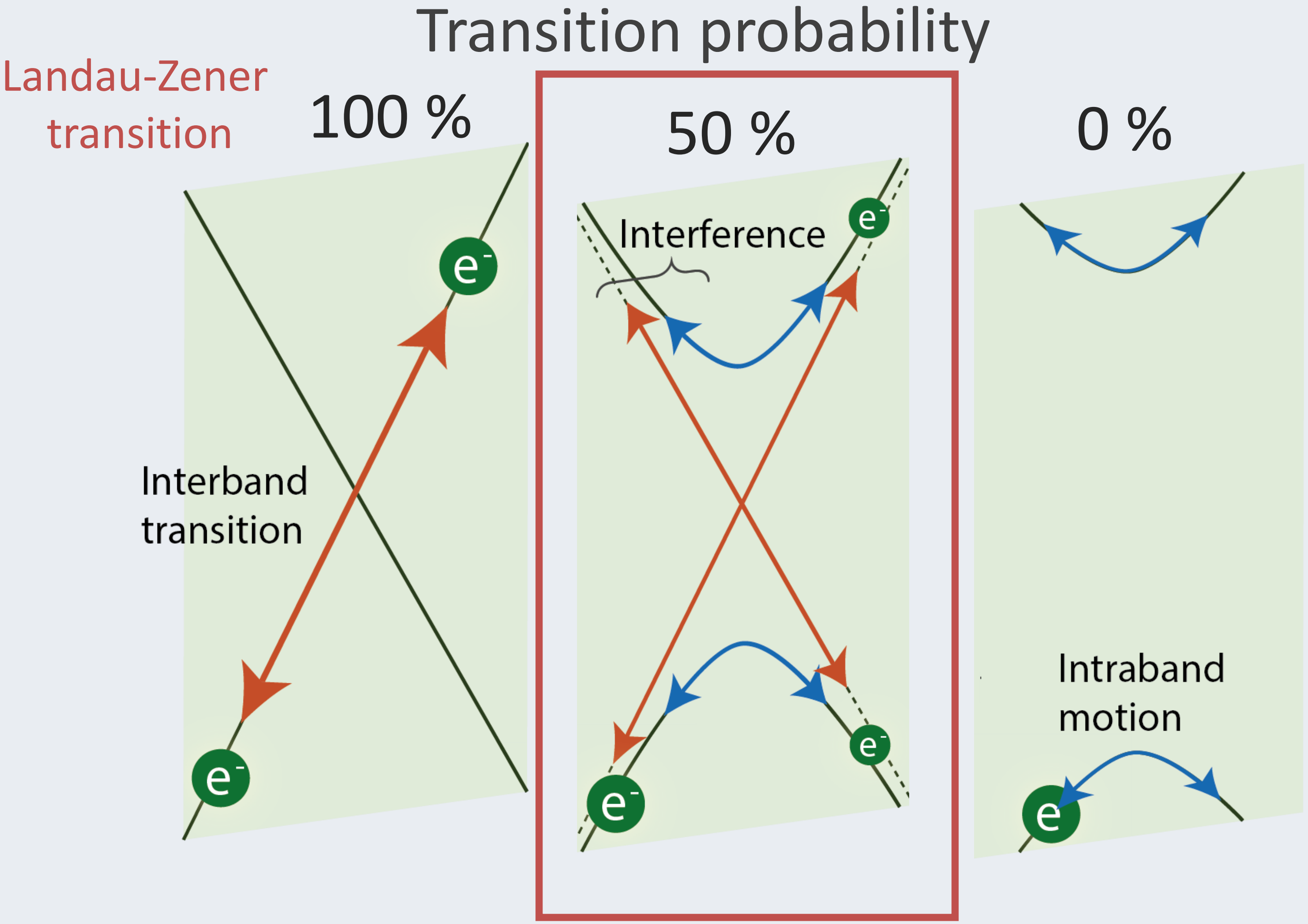
Conduction and valence band; π-orbitals



Light-field driven electron dynamics



$$\dot{\mathbf{k}}(t) = \hbar^{-1} e\mathbf{E}(t)$$

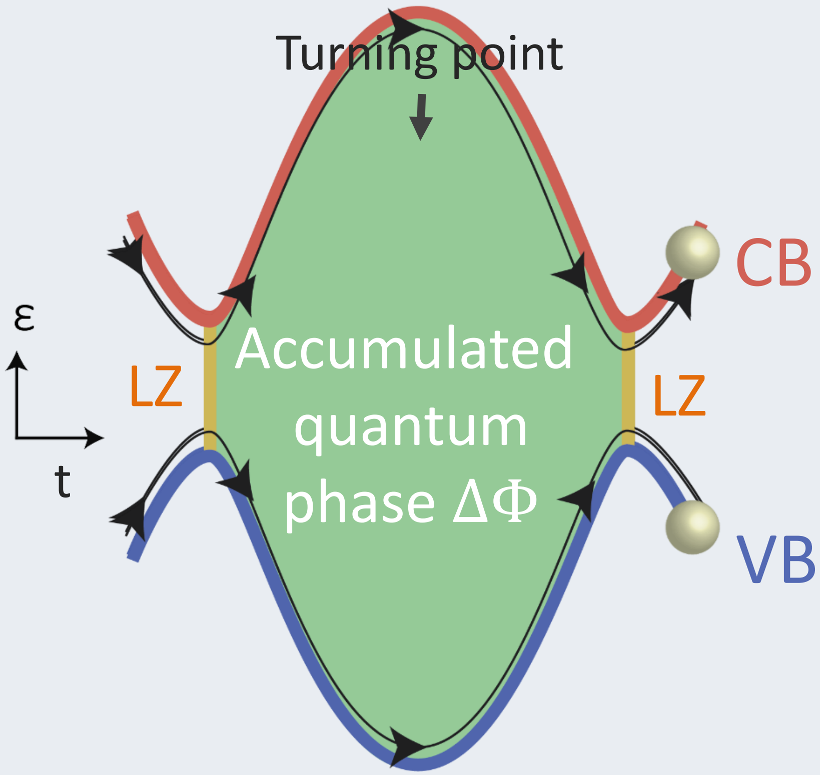


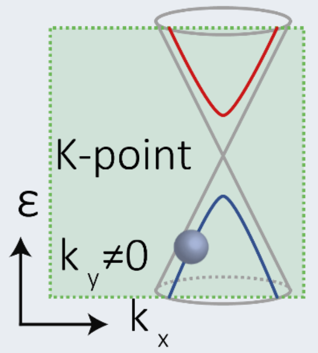
Theory: K. L. Ishikawa, PRB 82, 201402 (R) (2010), H. K. Kelardeh *et al.*, PRB 91, 045439 (2015), Wachter *et al.*, NJP 17, 123026 (2015), Kruchinin *et al.* Rev. Of Modern Physics (2018), C. Heide *et al.*, NJP 21, 045003 (2019), C. Heide *et al.*, PRA 104, 023103 (2021),
Review: O.V. Ivakhnenko *et al.*, Phys. Rep. 995, 89 (2023)

Landau-Zener-Stückelberg interferometry

$$\dot{\mathbf{k}}(t) = \hbar^{-1} e\mathbf{E}(t)$$

Light-field-driven electron interferometer





Michelson-Interferometer

Light analogue

Temporal evolution of the wavefunction

$$|\Psi(t)\rangle = e^{i\phi_n(t)}|\Psi_n(t)\rangle$$

$$\phi_n(t) = -\frac{1}{\hbar}\int_{t'}^{t} E_n(t)\,\mathrm{d}t + \oint A_n dR$$

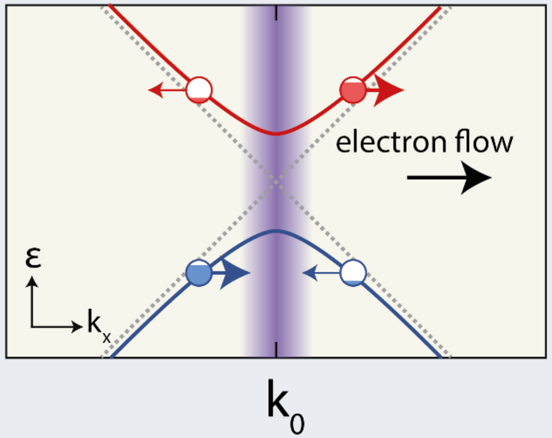
Dynamical phase

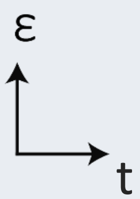
geometrical phase

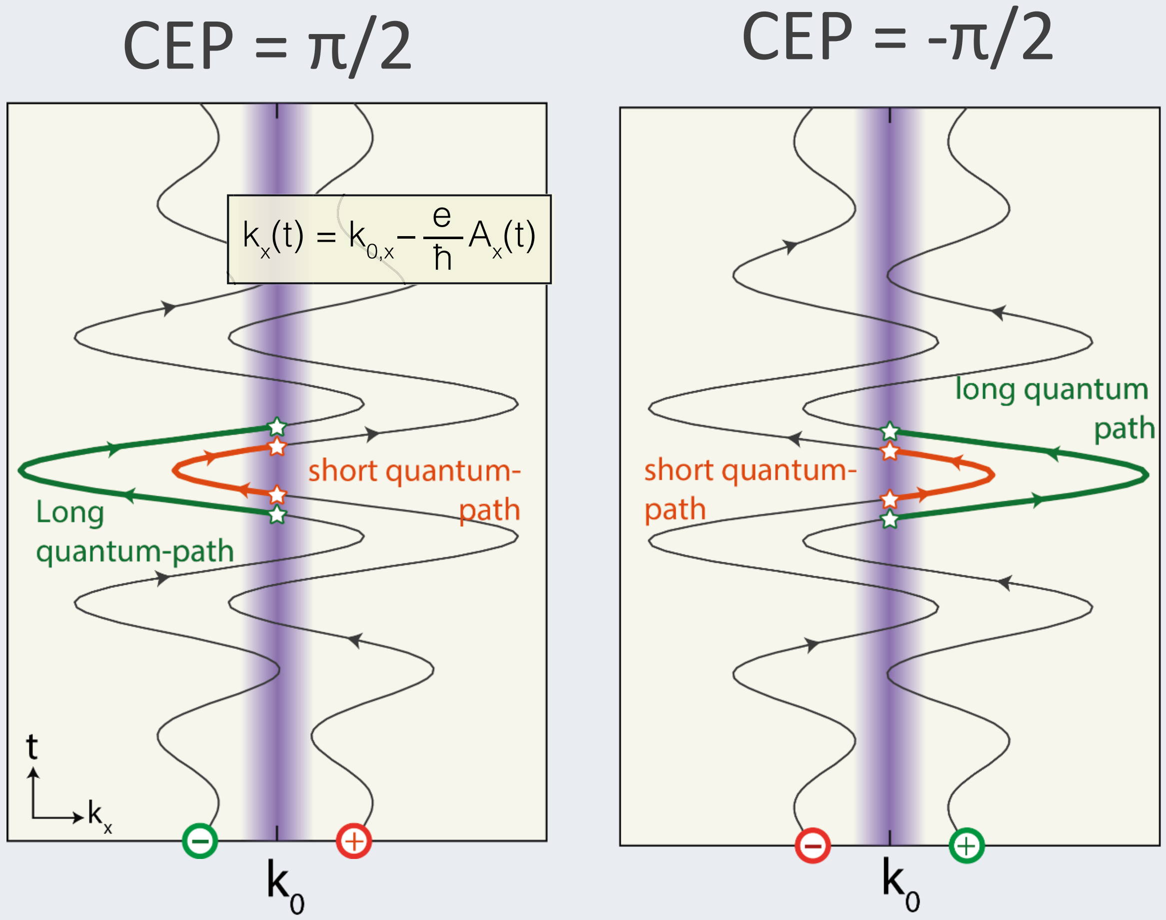
Probing of quantum phases via light-field-driven interferometry

CEP-dependent phase evolution

Residual ballistic current

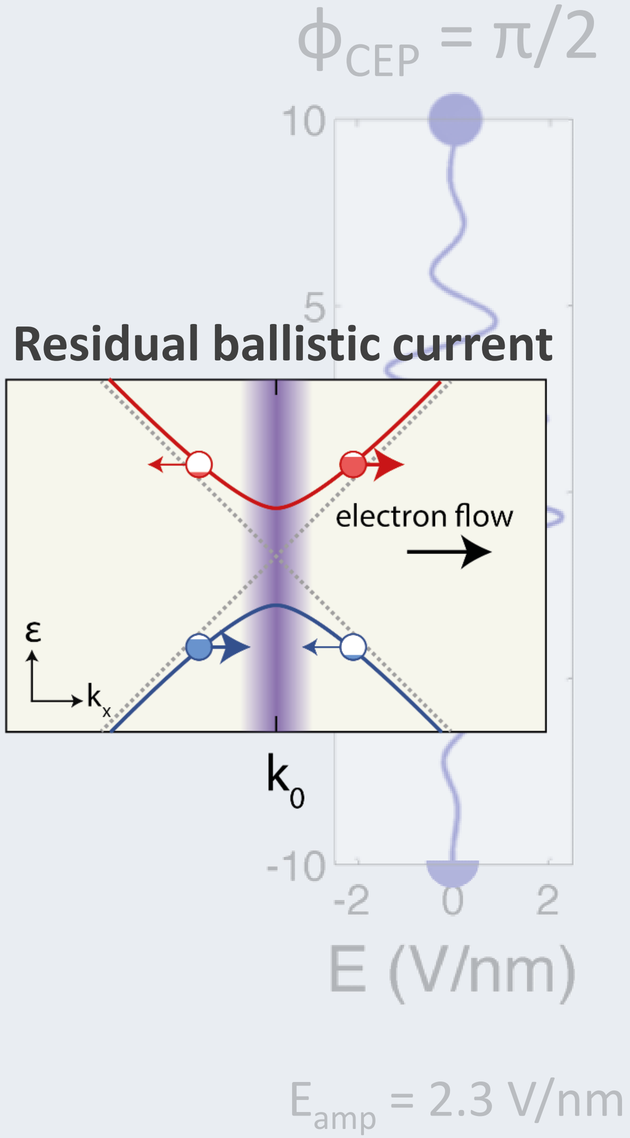


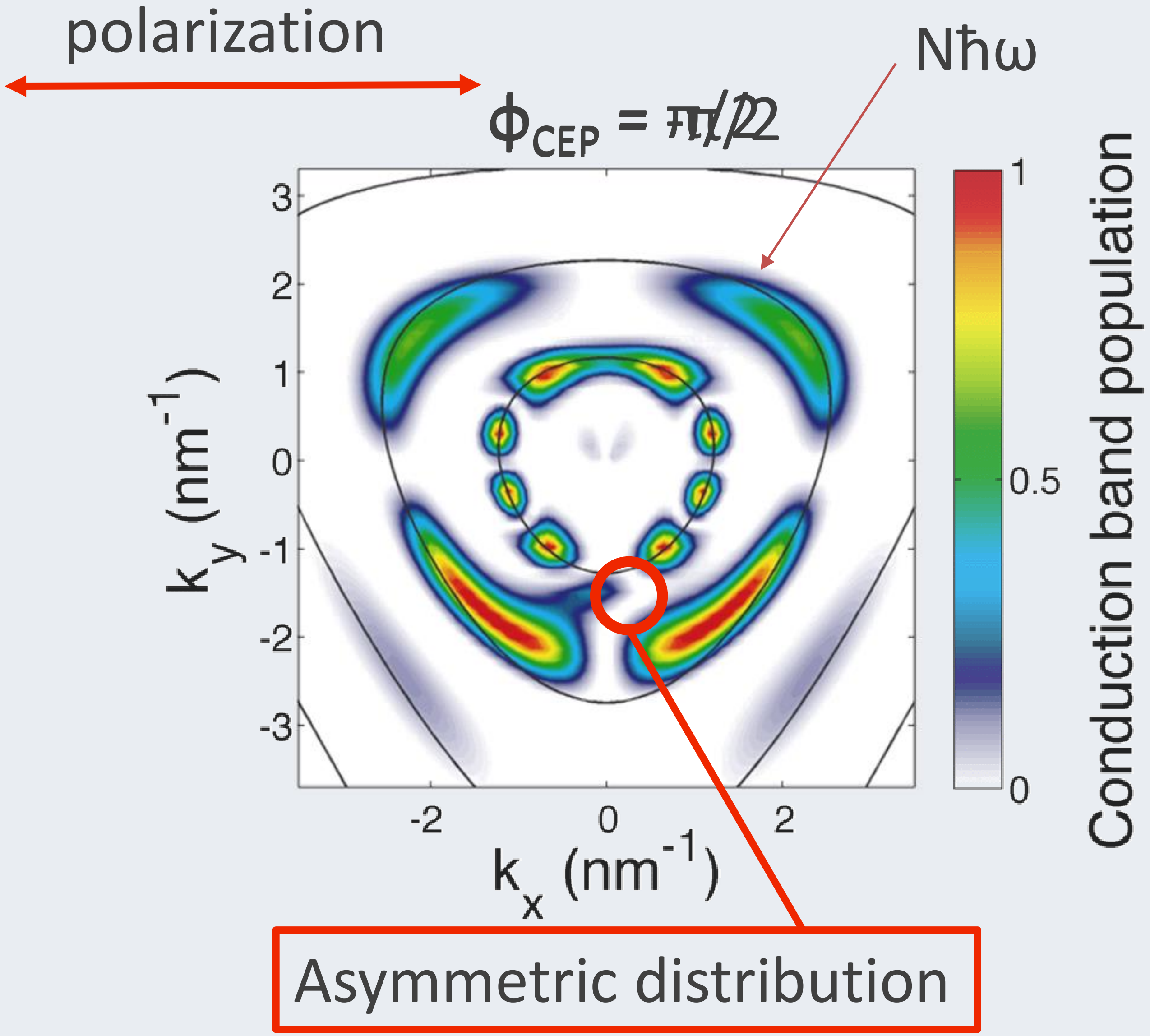




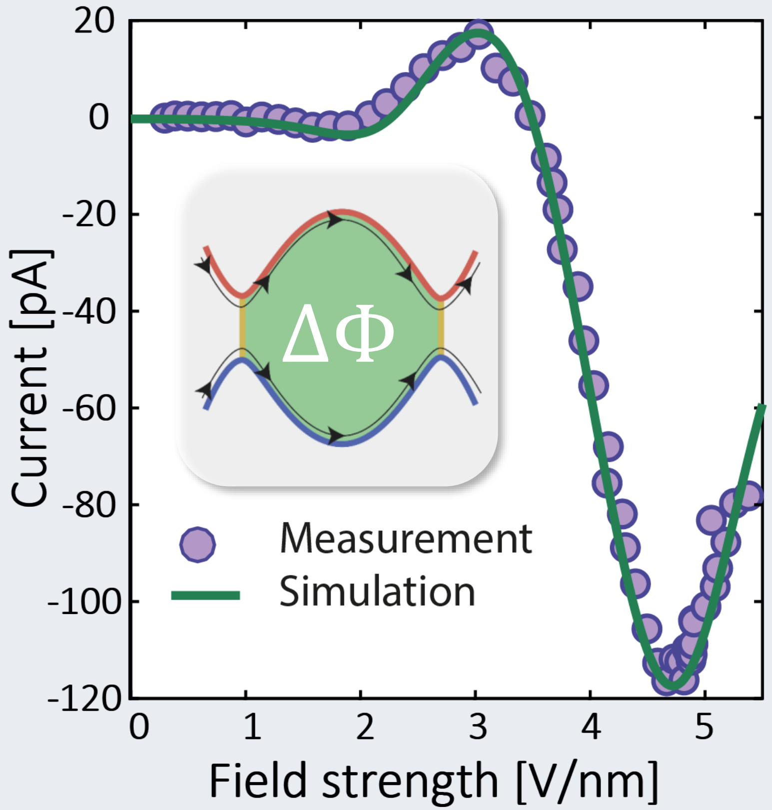
CEP-dependent current

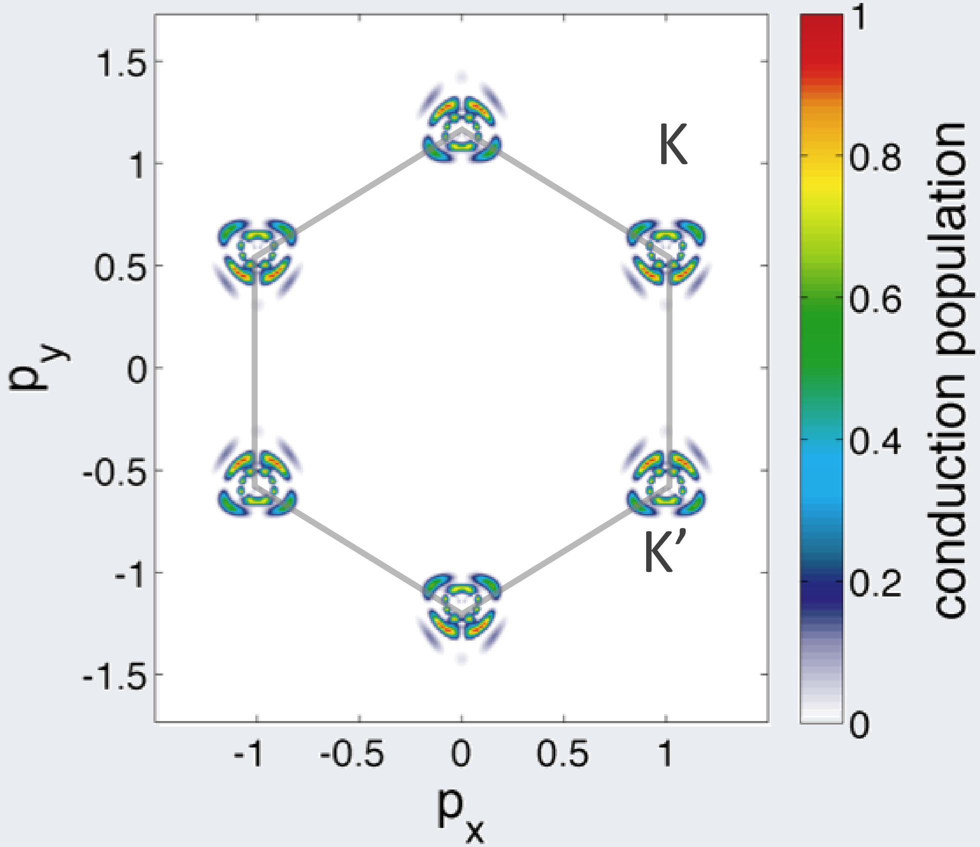
Numerical simulation (2 bands, tight binding)





Landau-Zener-Stückelberg interferometry





Sensing of quantum phases and coherence via current measurement

$$j_{\mathrm{el}}^{\mathrm{TLS}} = \frac{2e}{(2\pi)^2} \int_{-\infty}^{\infty} \frac{1}{\hbar} \frac{\mathrm{d}\mathcal{E}(k)}{\mathrm{d}k} \Delta f(k,t) \mathrm{d}k$$

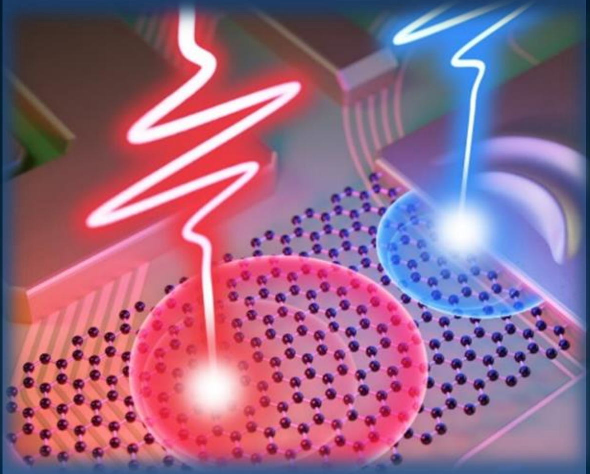
C. Heide, T. Boolakee, P. Hommelhoff in preparation

Playground: Light-field driven electron dynamics in solids

Light-field driven quantum dynamics

CH et al., **Nano Letters** 21, 9403 (2021)
CH et al., **PRL** 121, 207401 (2018)
TH, **CH** et al. **Nature** 550, 224 (2017)

Light-field driven dynamics at interfaces



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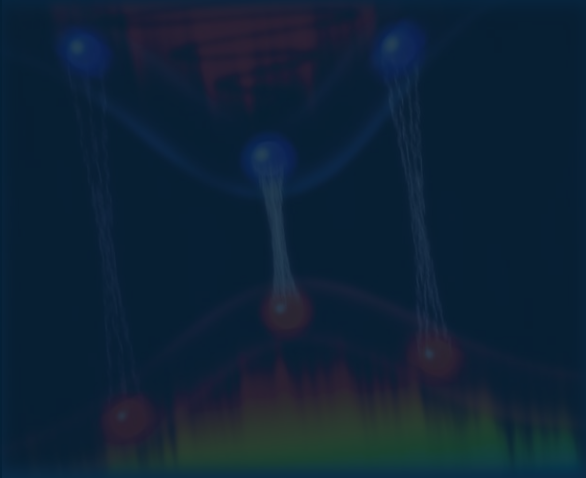
Role coherence



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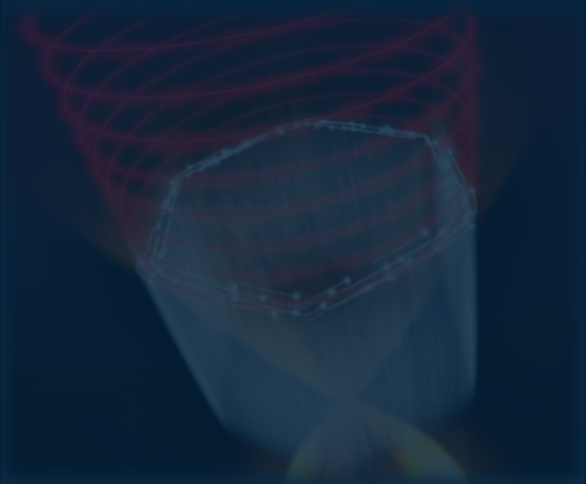
Topologically protected materials

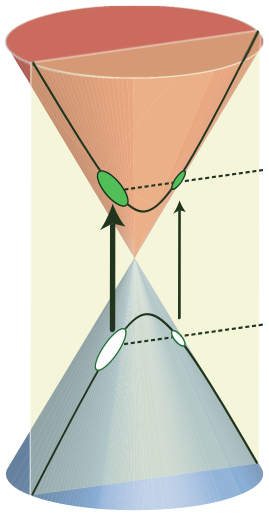


CH, YK et al., **Nature Photonics** 16, 620 (2022)

Light-field driven current

High-harmonic generation

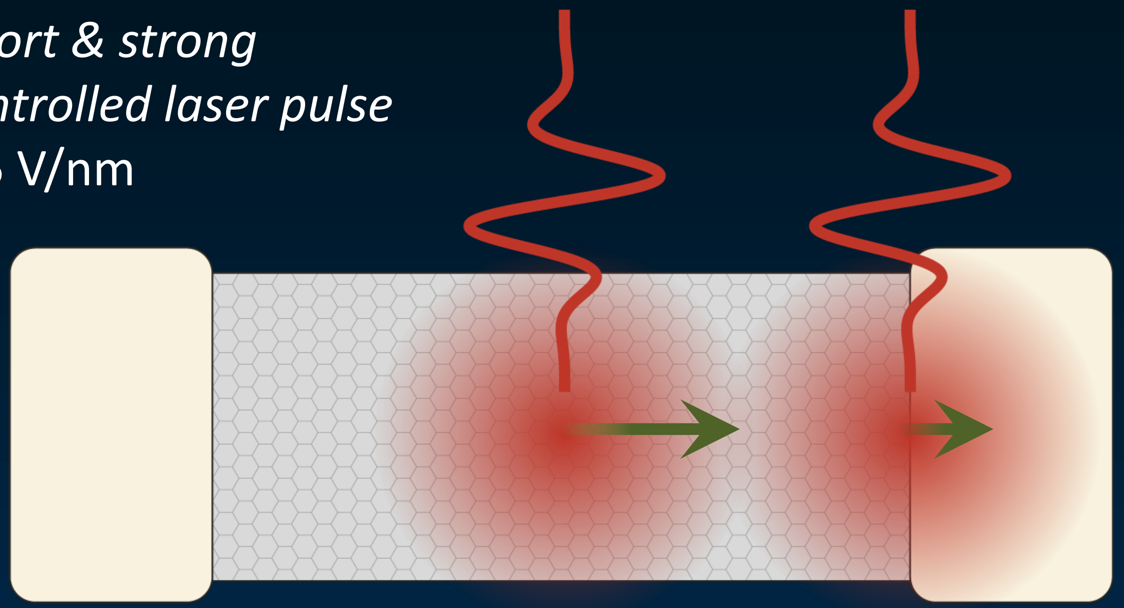
Light-field control of real and virtual charge carriers



Maximized for

$\varphi_{CE} = \pm\pi/2$

Ultrashort & strong
CEP-controlled laser pulse
6 fs, 2.5 V/nm



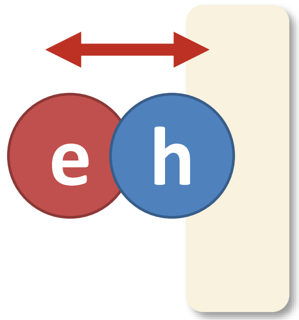
Real charge carriers

→ travel through the graphene

Virtual charge carriers

→ captured by the gold interface

For which waveform does the current peaks?

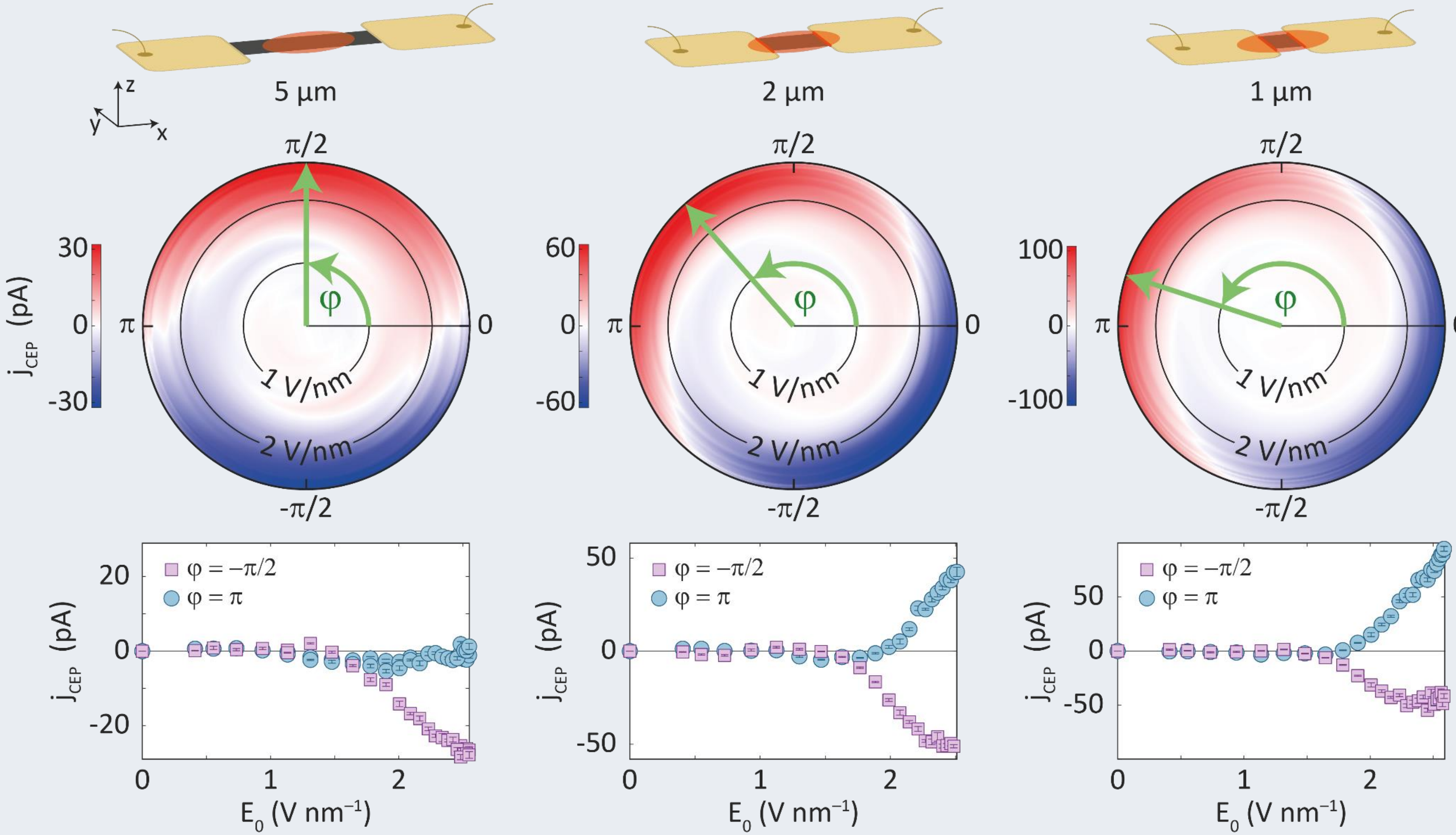


Maximized for

$\varphi_{CE} = 0, \pi$

T. Boolakee*, C. Heide*, et al. Nature **605**, 251 (2022)

CEP-resolved current

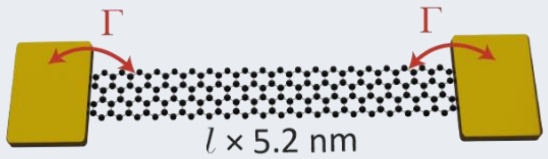


TB, CH, et al. **Nature 605**, 251 (2022)

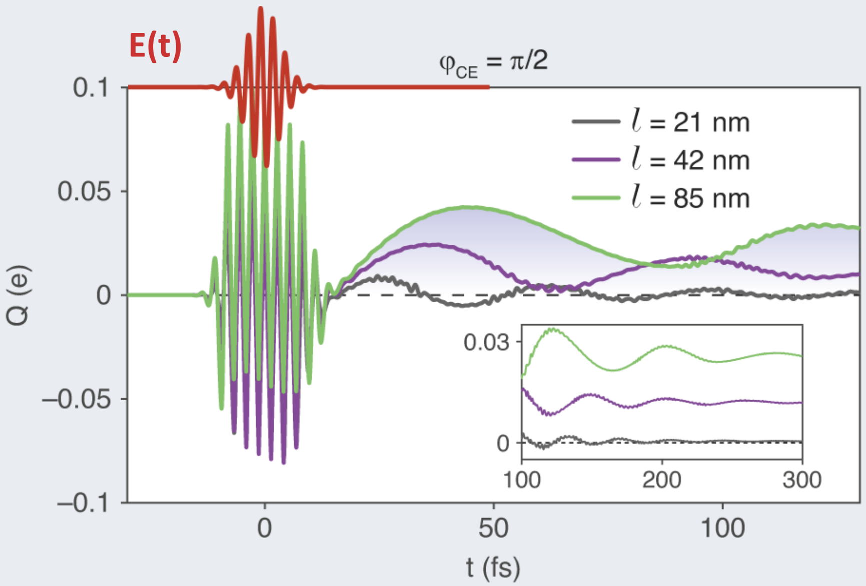
Real charge carriers virtual charge carriers

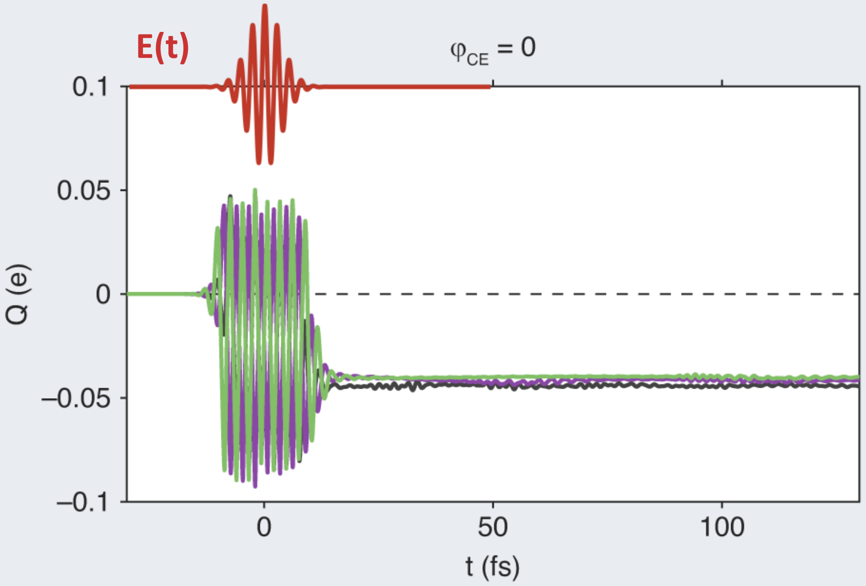
Ab initio charge transfer simulations

- Excitation: Graphene tight-binding model
- Propagation: Time-dependent non-equilibrium Green‘s function method



$$\underbrace{H(t) = H_G(t)}_{\text{graphene}} + \underbrace{H_M(t)}_{\text{metal}} + \underbrace{H_{GM}(t)}_{\text{interface coupling}}$$





Ignacio Franco

Antonio Garzon-Ramirez

Theory: X. Zheng *et al.*, Phys. Rev. B **75**, 195127 (2007), Y. Zhang, S. Chen, and G. Chen, Phys. Rev. B **87** (2013), L. Chen, Y. Zhang, G. Chen, and I. Franco, Nat. Commun. **9**, 2070 (2018)

Playground: Light-field driven electron dynamics in solids

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CH et al., PRL 121, 207401 (2018)
TH, CH et al. Nature 550, 224 (2017)

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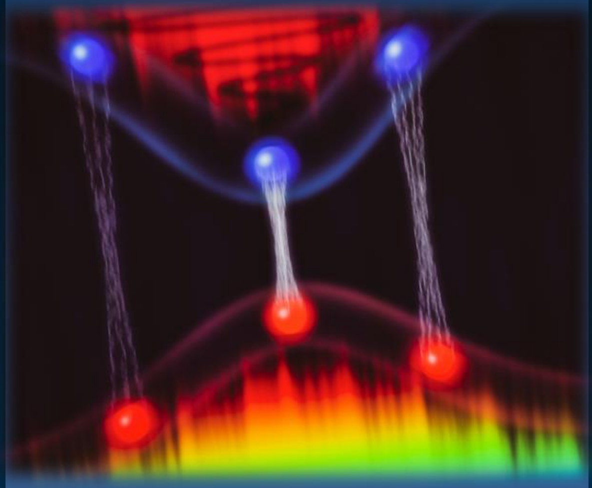
CH et al. Nature Photonics 14, 219 (2020)

Light-field driven current

Role coherence



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CH et al., **Nano Letters** 21, 22 (2021)

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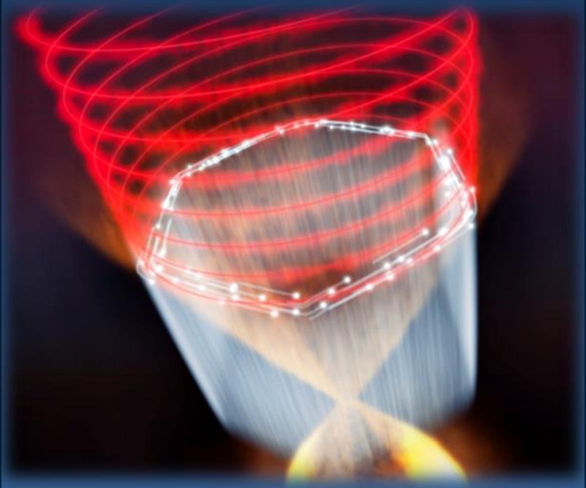


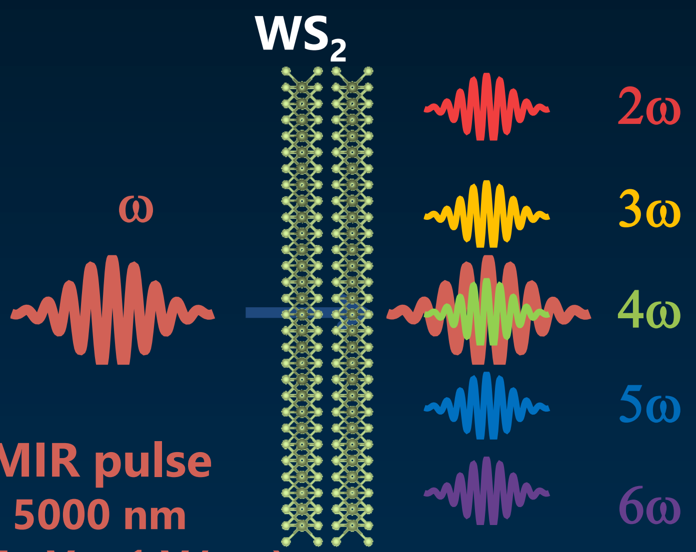
CH, YK et al., **Nature Photonics** 16, 620 (2022)

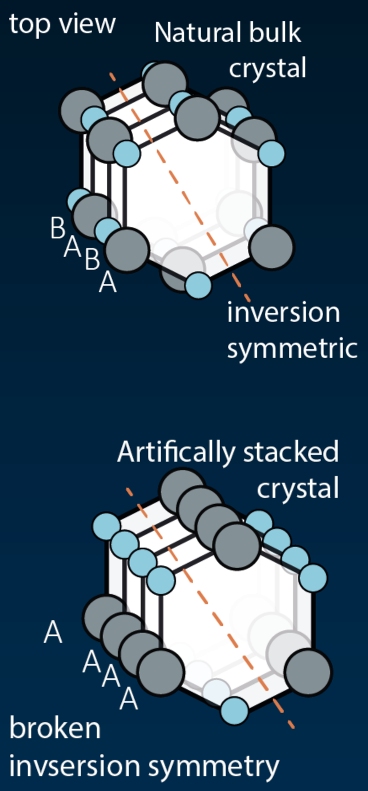
High-harmonic generation

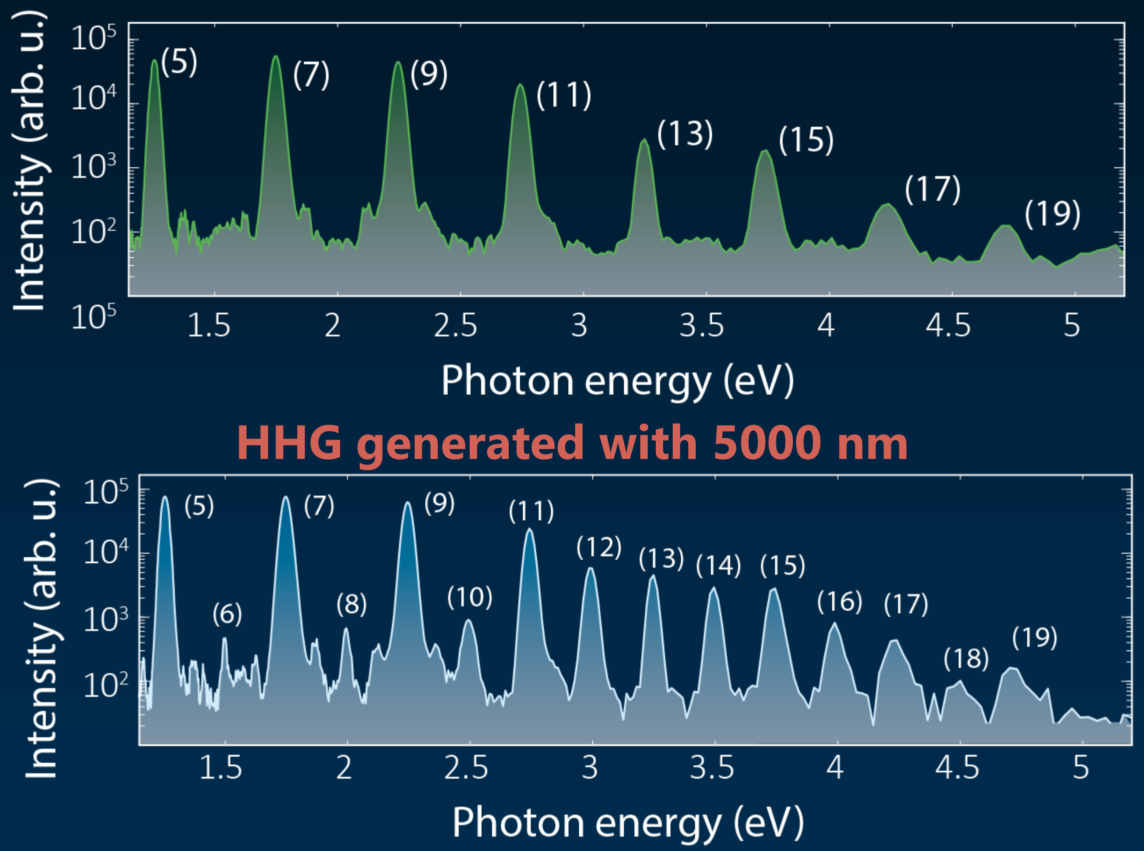
High-harmonic generation solids

Transition-metal dichalcogenide (TMD)

MoS_2, $MoSe_2$, WS_2, WSe_2, …

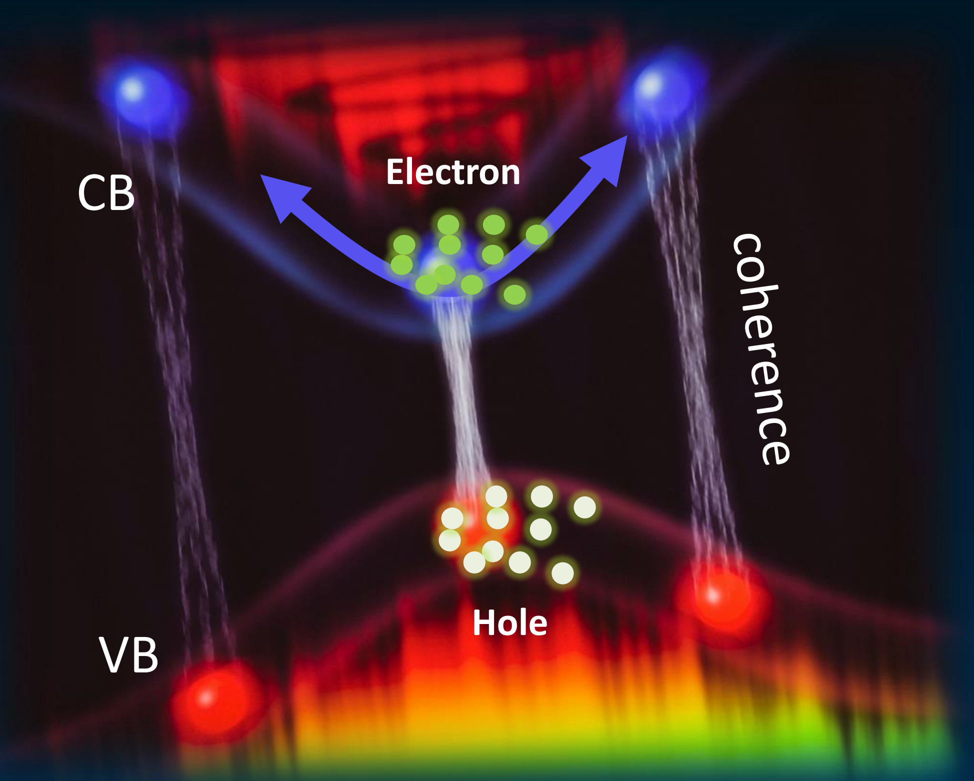






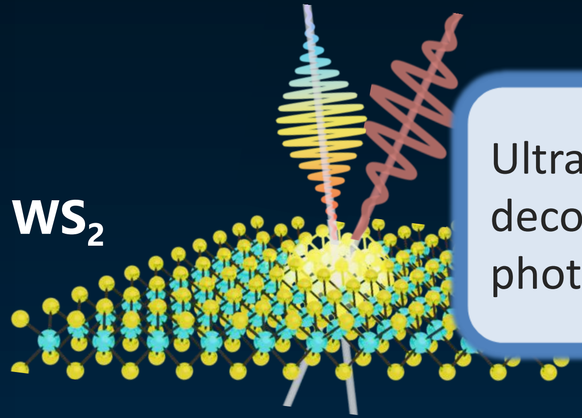
S. Ghimire, AD DiChiara, E Stistrunk, P Agostini, LF DiMauro, DA Reis, Nature Physics 7, 138 (2011)
C. Heide, Y. Kobayashi, AC Johnson, F. Liu, DA Reis, TF Heinz, S. Ghimire, **Nanophotonics** (2023)
C. Heide, Y. Kobayashi, AC Johnson, F. Liu, DA Reis, TF Heinz, S. Ghimire, **Optica** 9, 512 (2022)

Probing electron-hole coherence in MoS_2 via HHG

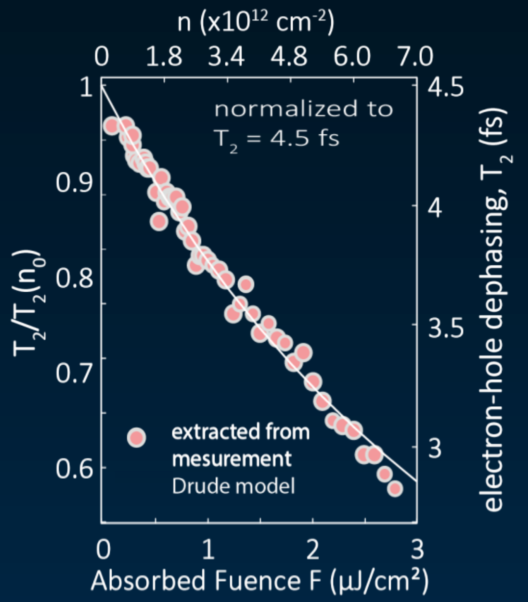


1) intraband motion
2) interband polarization

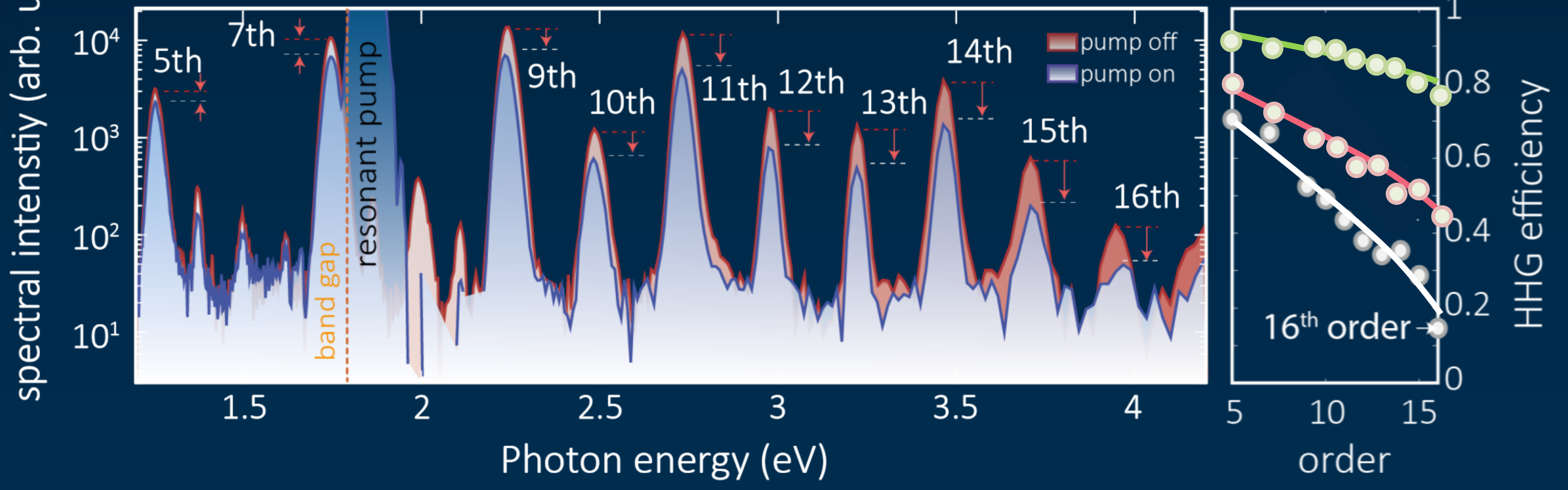
For 2) e-h coherence required!
(typ. Timescale 1 - 10s of fs)



Ultrafast, sub-cycle decoherence under high photodoping



HHG generated with 5000 nm

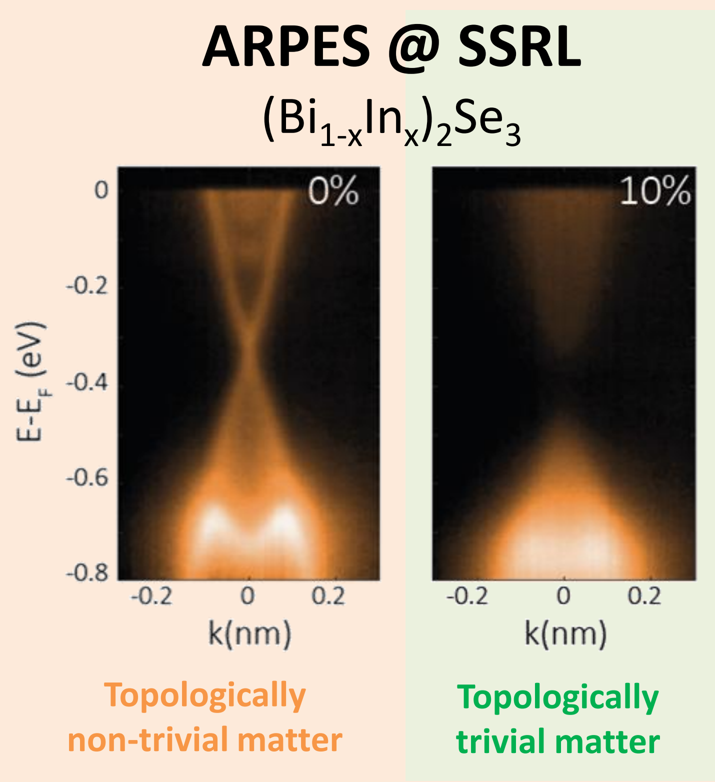


C. Heide, Y. Kobayashi, AC Johnson, F. Liu, DA Reis, TF Heinz, S. Ghimire, **Optica** 9, 512 (2022)

Limitations of lightwave electronics

PROBLEM: Ultrafast scattering (τ<10 fs) leads decoherence

SOLUTION: Topological insulators, e.g. SB_2Te_3, Bi_2Se_3 …



C. Heide et al., **Nature Photonics** 16, 620 (2022)

Seongshik Oh
Deepti Jain
Rutgers University

Ballistic acceleration through Dirac point

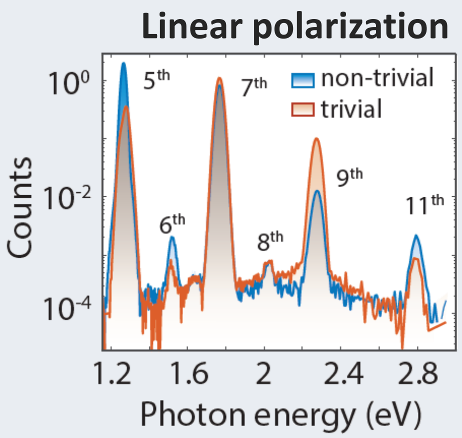
Bauer et al., PRL 120, 177401 (2018)
Silva et al., Nat. Photonics 13, 849 (2019)
Baykusheva et al., PRA 103, 023101 (2021)

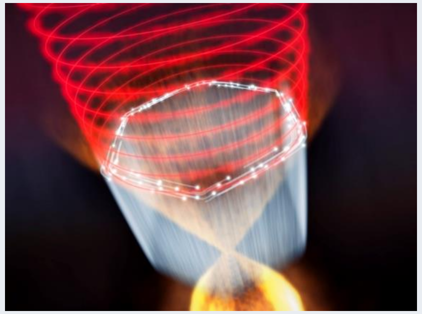
Can this be observed?

Havez et al., Nature 561, 507 (2018)
Kovalev et al., Nat. Com. 11, 2451 (2020)
Bai et al., Nat. Phys. 17, 311 (2021)
Schmid et al., Nature 593, 385 (2021)

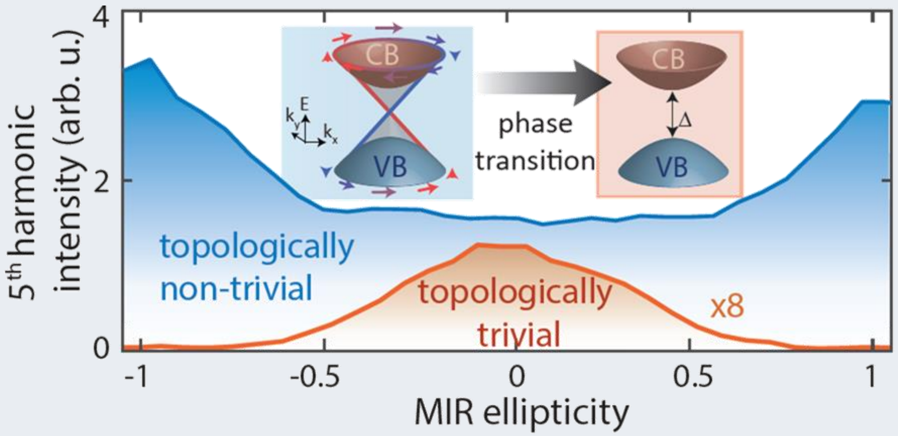
Enhanced light-matter interaction from top. surface state

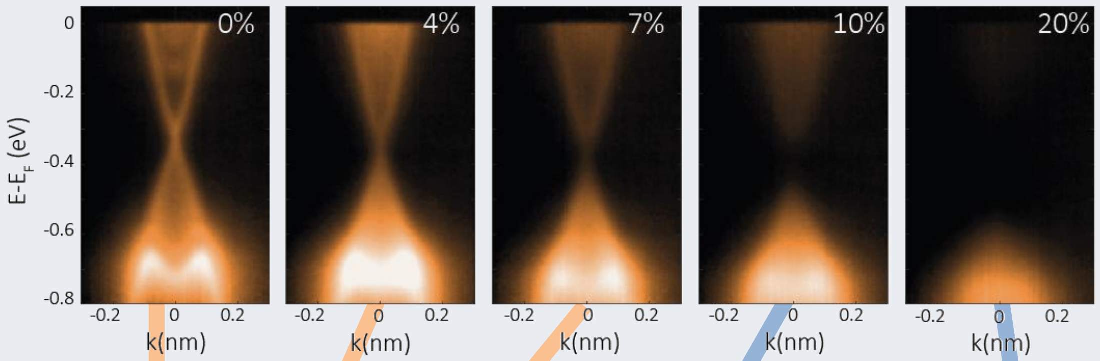
Laser parameter: 5000 nm, 100 fs, 0.21 V/nm

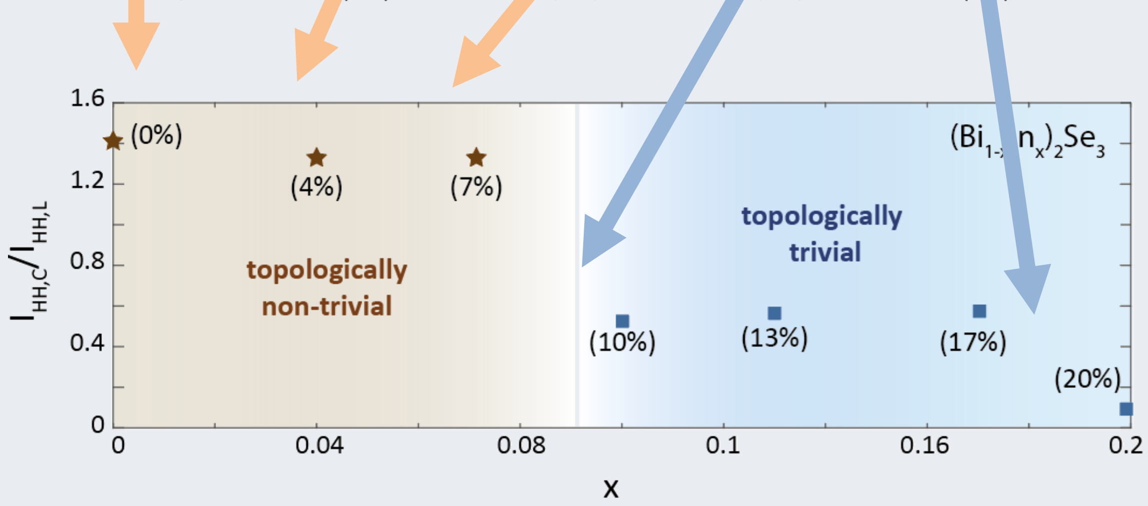




5th harmonic order







Enhanced response from topologically protected surface state under circular excitation

CH et al., **Nature Photonics** 16, 620 (2022)

Playground: Light-field driven electron dynamics in solids

atoms, molecules

condensed matter

2D materials

Topological insulators

Functional electronics

Light-field driven quantum dynamics

Light-field driven dynamics at interfaces

Role coherence

Topologically protected materials

CH et al., Nano Letters 21, 9403 (2021)
CH et al., PRL 121, 207401 (2018)
TH, CH et al. Nature 550, 224 (2017)

TB, CH et al., Nature 605, 251 (2022)
CH et al. Nature Photonics 14, 219 (2020)

CH, YK et al., Optica, 9, 5 (2022),
CH et al., Nano Letters 21, 22 (2021)

CH, YK et al., Nature Photonics 16, 620 (2022)

Light-field driven current

High-harmonic generation

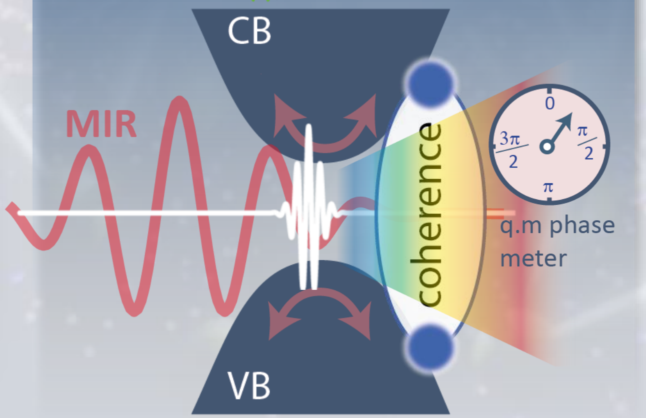
Wavetronics: Light-field driven quantum electronics on a chip

1. Sub-cycle-resolved X-ray scattering from light-field-driven electronic systems

Tracking electron motion in real/momentum space

2. Attosecond wave-packet interferometry

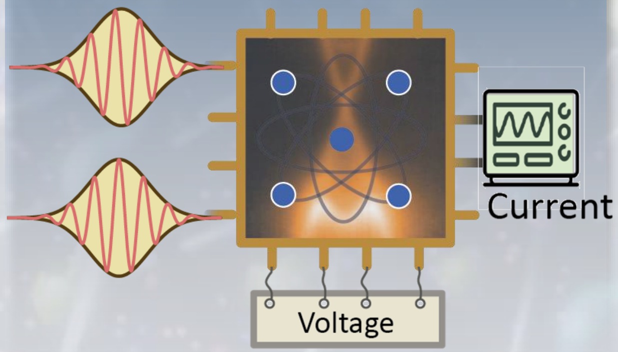
Probing the full quantum nature of the electrons



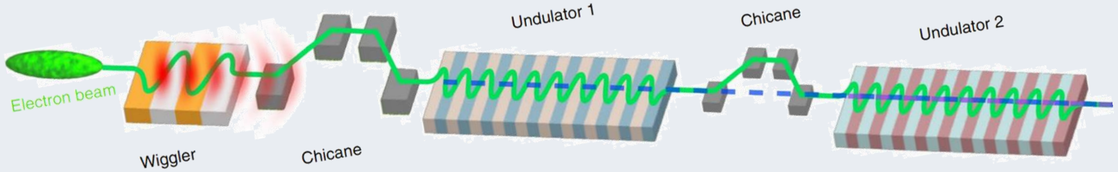
3. Wavetronics

"Petahertz electronics"

Strong-field laboratory on a chip



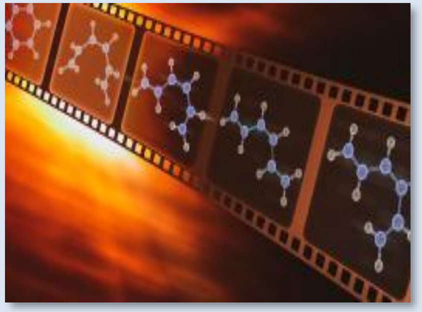
1. Sub-cycle-resolved X-Ray scattering from light-field-driven electronic systems



J. Duris, S. Li *et al.*, Nature Photonics **14**, 30-36 (2020)

Attosecond XFEL for pump-probe experiments for research on the sub-femtosecond timescale quantum-mechanical motion of electrons

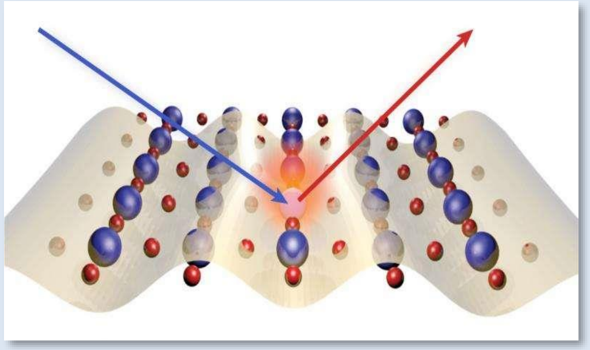
Attosecond science in AMO



Molecular movies

S. Li, T. Driver *et al.* Science **375**, 6578 (2022)

Light-induced processes in condensed matter



Instruments: **XPP, UED**

THz resolved dynamics, phonon dynamics, Correlations, phase Transitions, charge density waves

L. Chaix et al., Nature Physics 13, 952-956 (2017)

Developing coherent and sub-cycle science with XFELs on electronic systems

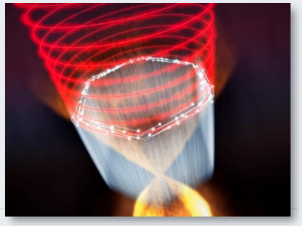
Questions to be address:

- Nanoscale material dynamics (i.e., charge separation at interfaces)
- Transient non-equilibrium and meta-stable phases
- Emergent phenomena in quantum materials

1. Sub-cycle-resolved X-Ray scattering from light-field-driven electronic systems

Challenges: Jitter between x-ray and optical beam

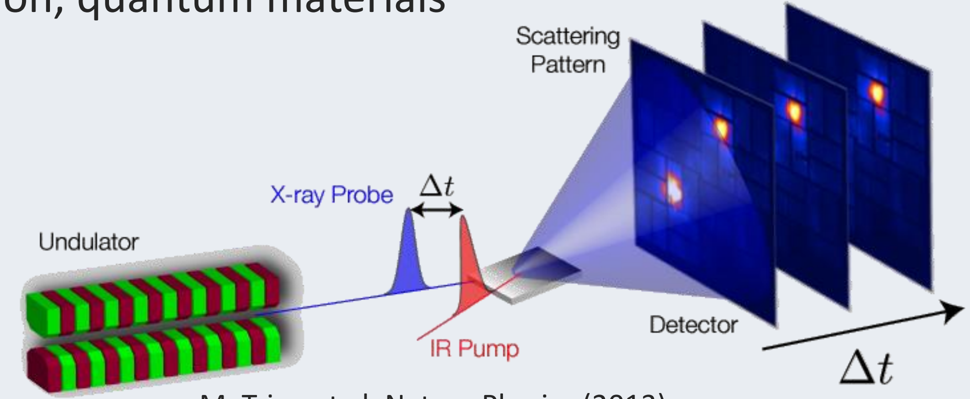
Sub-cycle resolved transient absorption spectroscopy



Electron dynamics: excited sates, non-equilibrium, transient structure
Materials: topol. materials, valley excitation, quantum materials

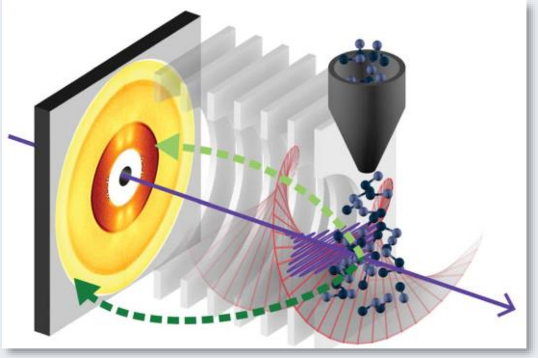
Sub-cycle resolved x-ray diffraction

Benefit: Spatial resolution
I.e., charge separation at material interfaces



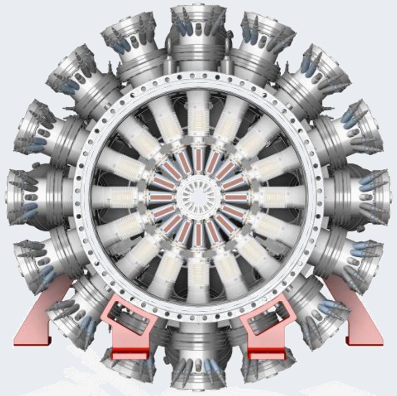
M. Trigo et al. Nature Physics (2013)

Attoclock



S. Li, T. Driver *et al.* Science **375**, 6578 (2022)

LCLS II

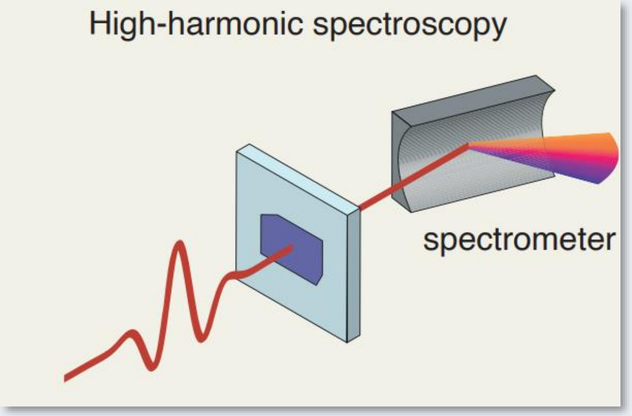


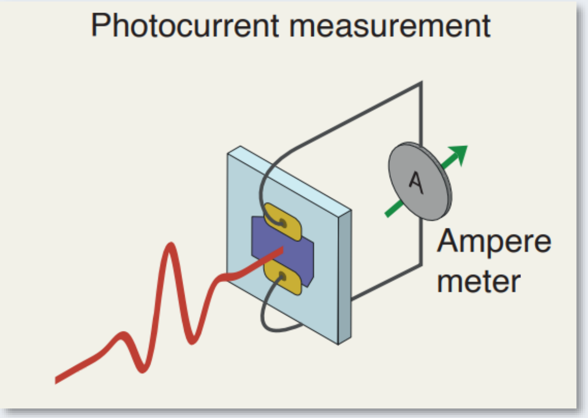
P. Walter *et al.*, JSR, 28, 5 (2021)

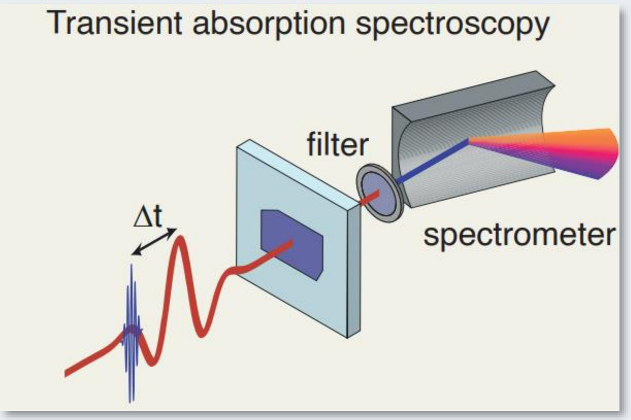
- How do we **control** material processes at the level of electrons?
- Ultimate **speed and efficiency** of charge separation?
- Characterize control matter under **extreme non-equilibrium conditions**?
- **Harnessing coherence** in light and matter
- Study **quantum materials**: high-temperature superconductivity, colossal magnetoresistivity, and topologically protected phases

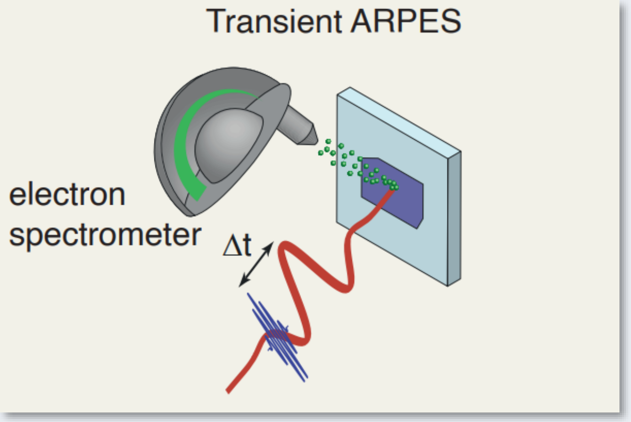
2. Attosecond wave-packet interferometry

Tracking light-field driven processes



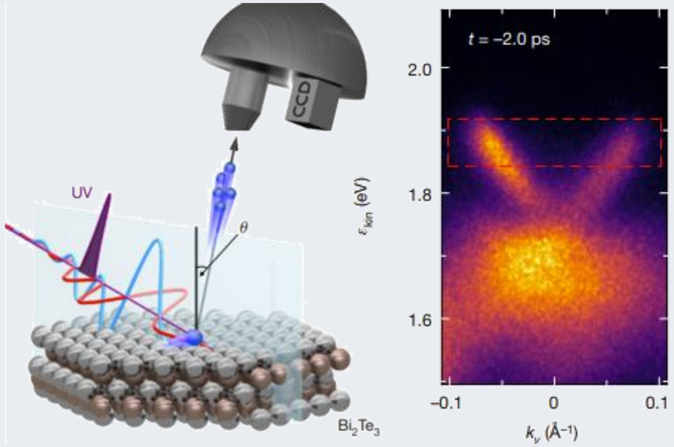






Phase information of the electrons gets lost!

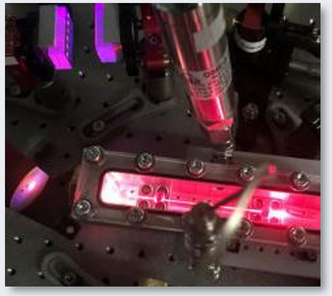
Development of **attosecond wave packet interferometry**

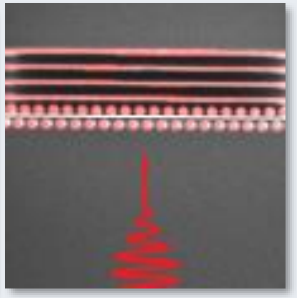


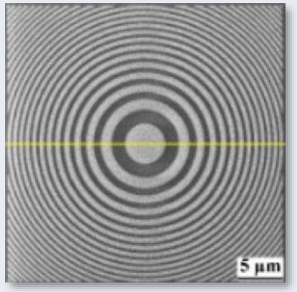
Reimann *et al.*, **Nature 562**, 396-400 (2018)

2. Attosecond wave-packet interferometry: electron's phase.

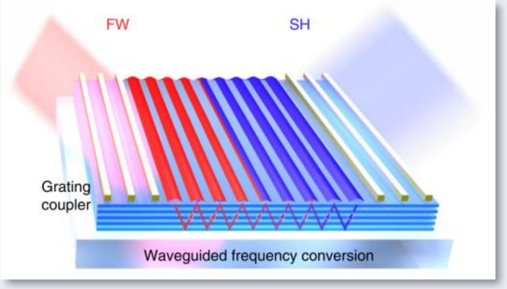
Source development (Photonics)


AE, **CH**, et al., Optics Let. 44, 5005 (2019)


ACHIP, Moore foundation



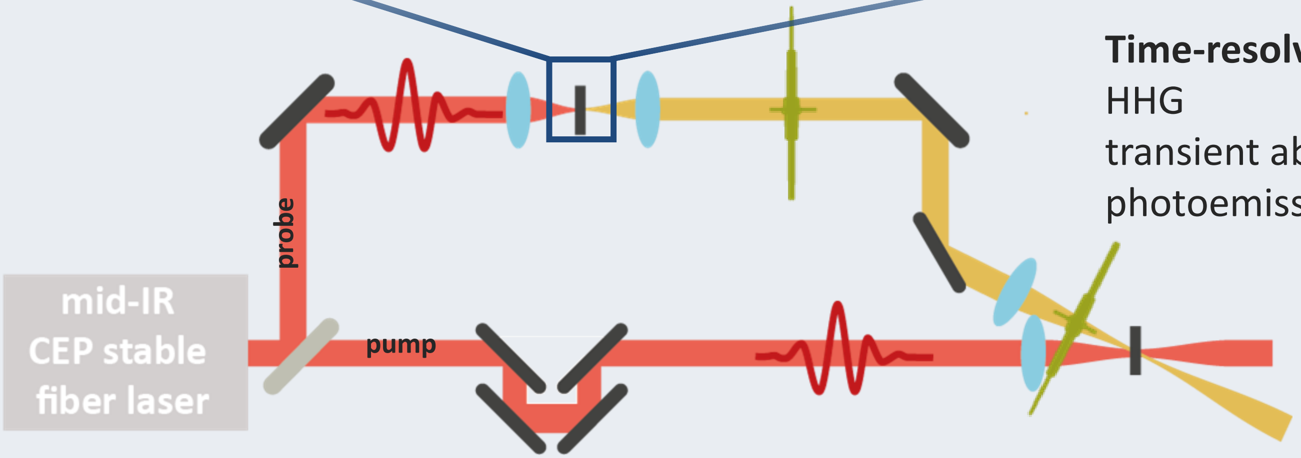
AK, **CH**, *et al.*, PRX **12**,041036 (2022)



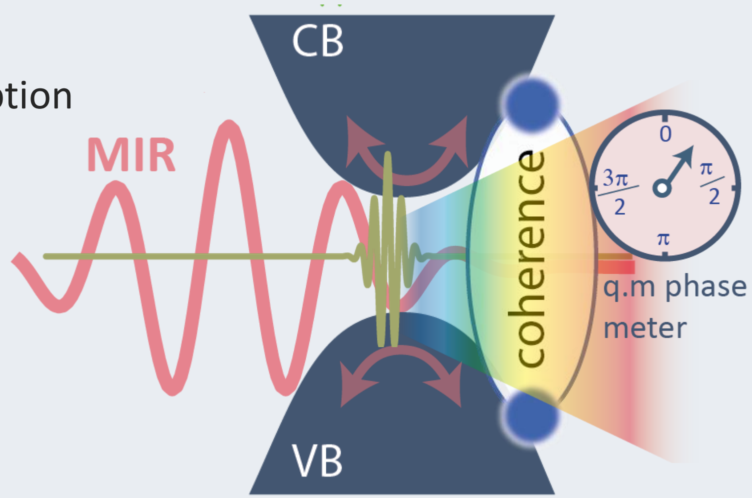
Xu, *et al.*, Nat. Photon. **15**, 6-10 (2022)

- sub-cycle resolved coherent spectroscopy
- Reconstruction of full quantum mechanical wavefunction via electron wave interference
- Probing of dynamical and geometrical quantum phases

Interesting for: Programmable quantum materials and topologically protected materials



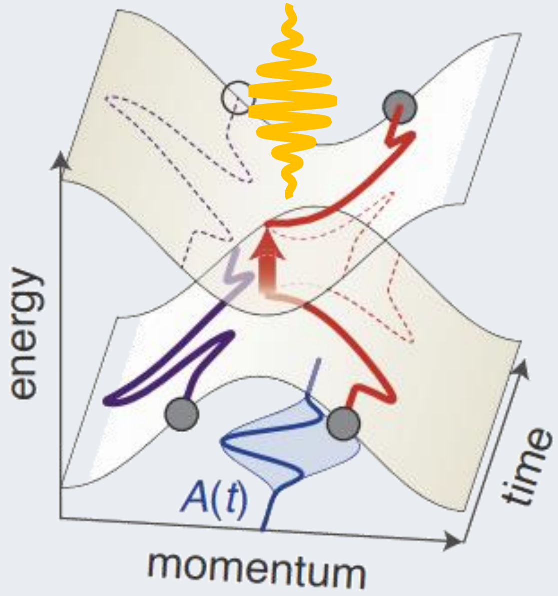
Time-resolved
HHG
transient absorption
photoemission



3. Lightwave electronics – light-field-induced states

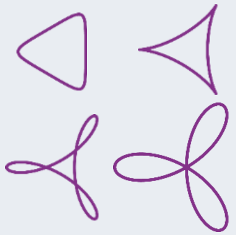
Advanced light-field driven electron interferometry

Coherent seeding

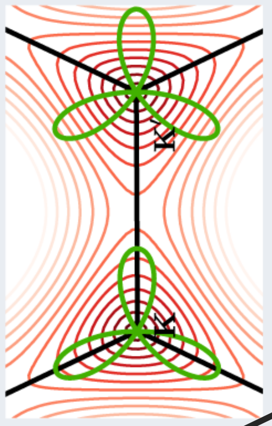


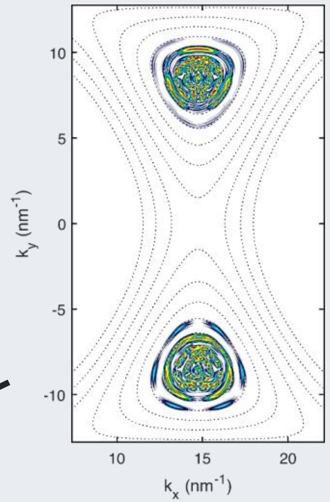
Electron trajectory control and coherent seeding for band structure and phase reconstruction (*closed-loop control*)

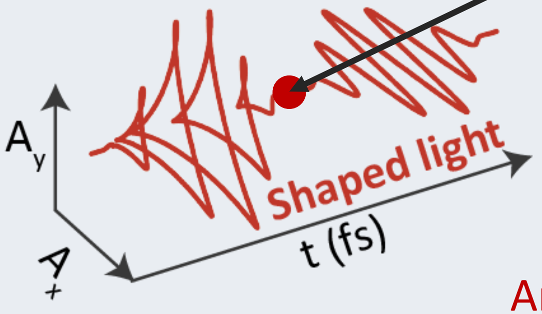
Multicolor and all-optical anomalous Hall effects



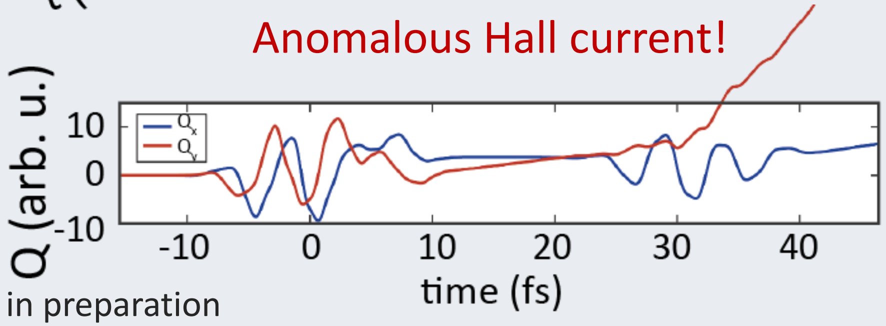
M. S. Mrudul *et al.*, Optica **8**, 422 (2021)
S.A. O. Motlagh et al., Nanophotonics 10, 3677 (2021)





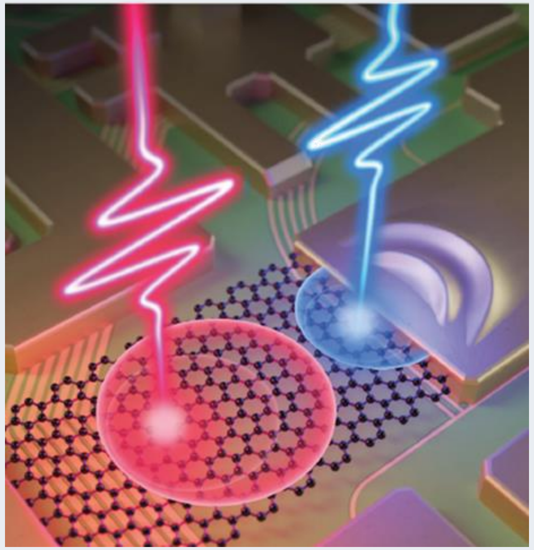


Light-induced valleytronics in pristine graphene

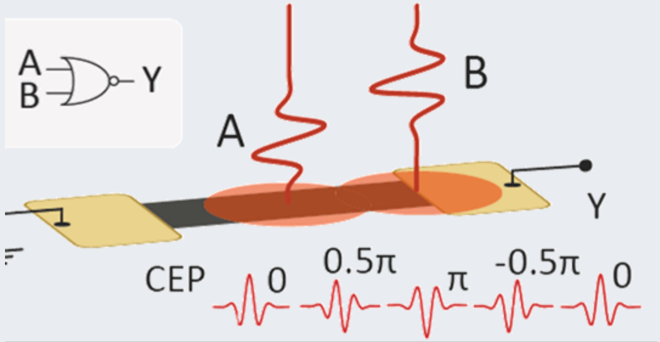


C. Heide *et al.*, in preparation

3. Lightwave electronics: optical logic gates

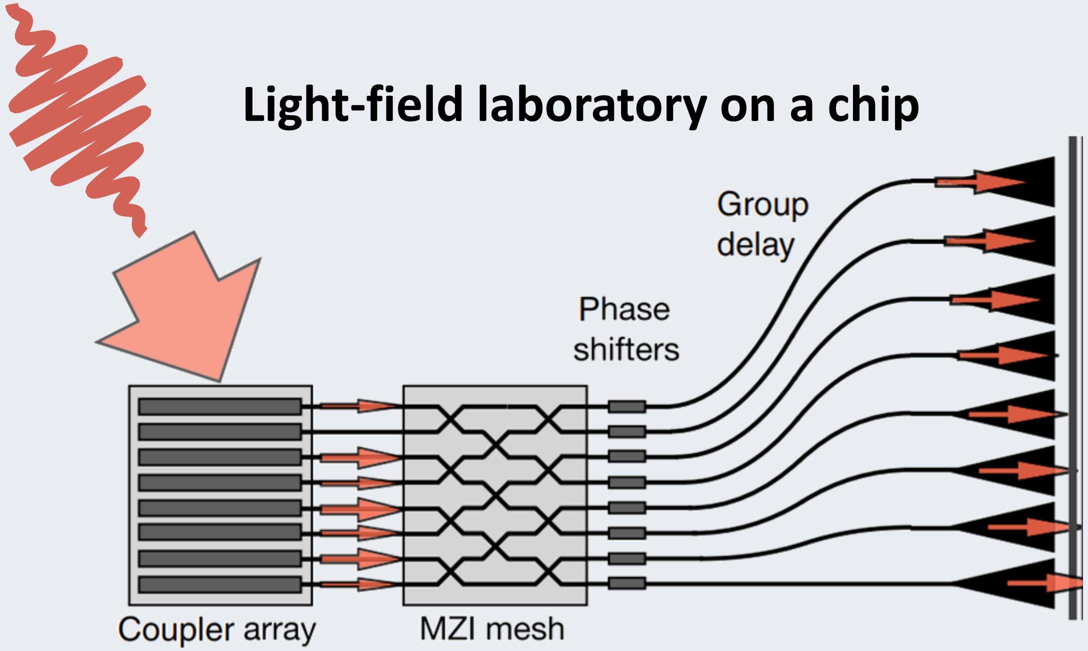


Lightwave logic



$\Delta\varphi_{CE}$	φ_A	φ_B	A	B	Y = A ▽ B
0	0.5π	0.5π	0	0	1
0.5π	0.5π	0	0	1	0
π	-0.5π	0.5π	1	0	0
-0.5π	-0.5π	0	1	1	0

Light-field laboratory on a chip



Hughes et al. PRA 11, 064014 (2019)

Transfer information from electric field waveform to current to build optical logic gates/transistors

„Power“ photonic circuits on a chip for light-field driven electronics

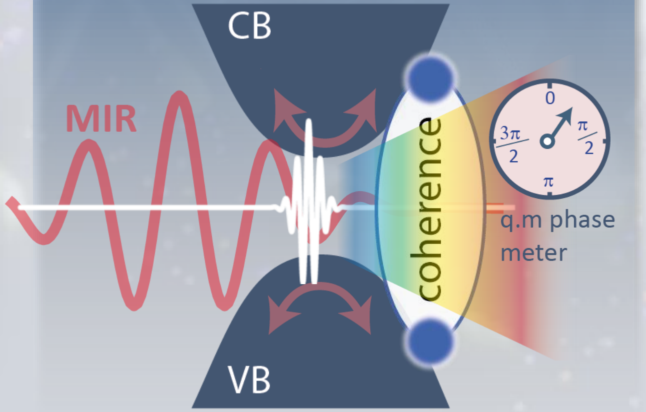
Wavetronics: Light-field driven quantum electronics on a chip

1. Sub-cycle-resolved X-ray scattering from light-field-driven electronic systems

Tracking electron motion in real/momentum space

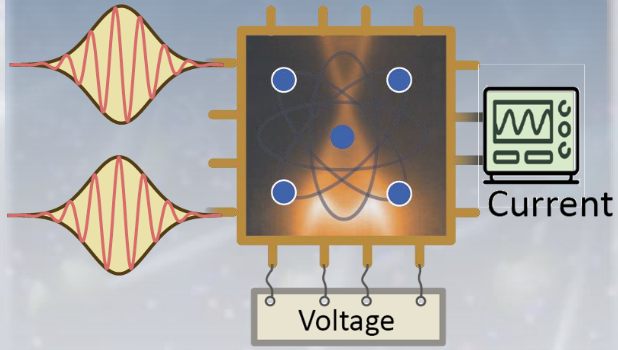
2. Attosecond wave-packet interferometry

Probing the full quantum nature of the electrons



3. Wavetronics
"Petahertz electronics"

Strong-field laboratory on a chip



SLAC

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SUNCAT

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