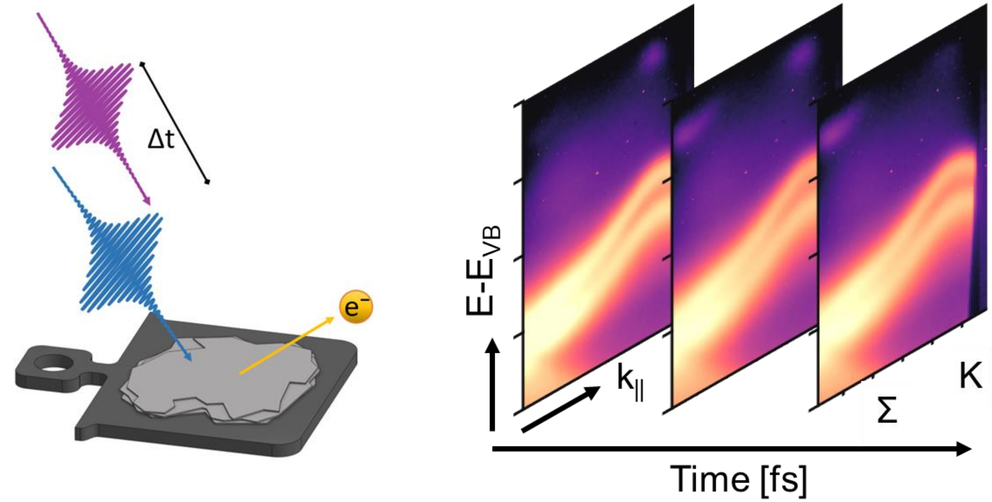
Universität
Augsburg
University

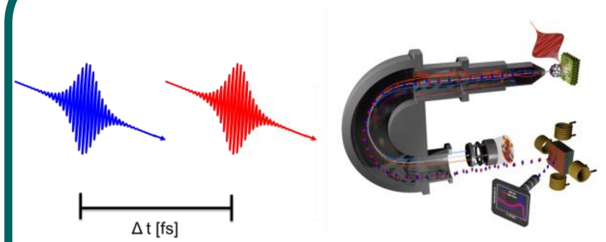


Time-resolved photoemission spectroscopy – ***An ultrafast camera to image charge and spin carrier dynamics in condensed matter***

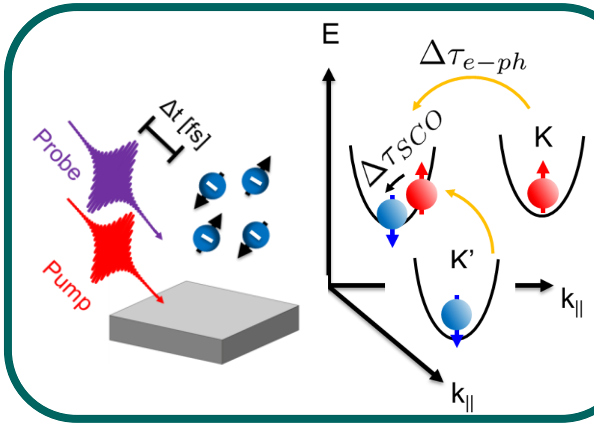
Benjamin Stadtmüller

Experimentalphysik II, Institute of Physics, Augsburg University, Germany

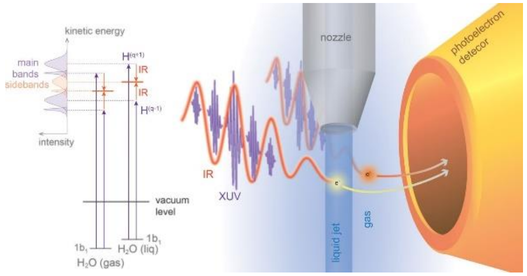
Outline



Ultrafast science in solids
… electrons in solids
… time-resolved photoemission spectroscopy



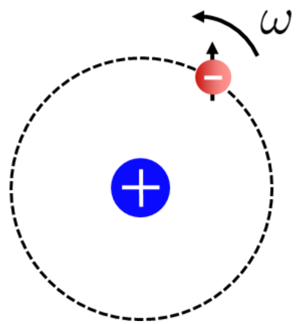
Monitoring….
…. the population dynamics of charge and spin carriers in momentum space
… the nature and spatial distribution of charge carriers in direct space
… interlayer charge separation across interfaces



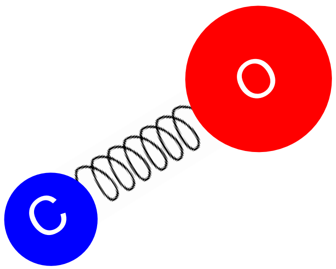
Perspectives and challenges of attosecond surface science

The characteristic timescales of (condensed) matter

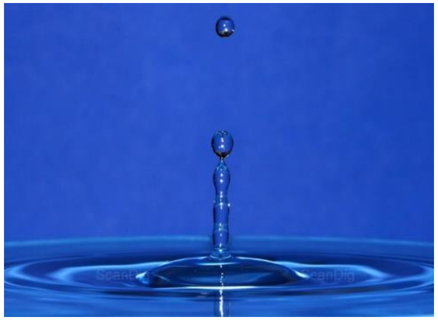
charge/spin carriers



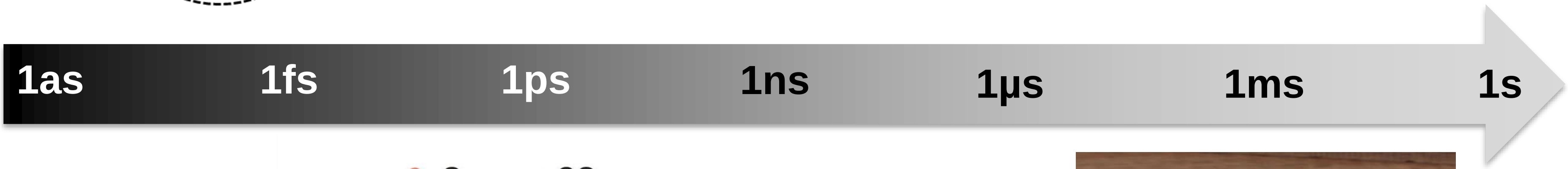
atoms/molecules



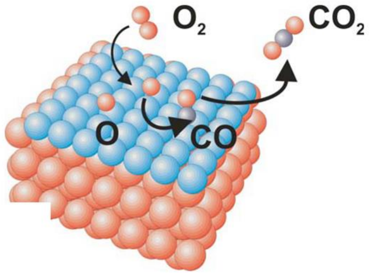
water droplets



humans



Shortest bursts of light



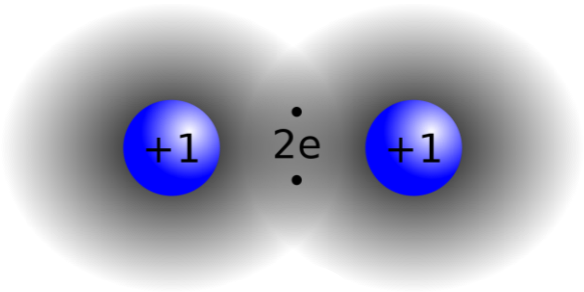
Chemical reactions

data processing in current technology

animal world

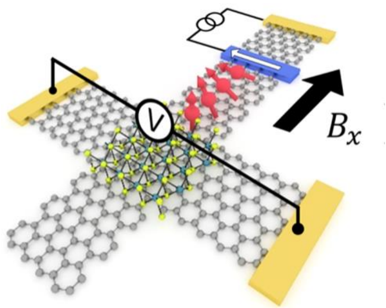
The Role of Charges and Spins in Condensed Matter

Bonding of atoms to molecules and solids



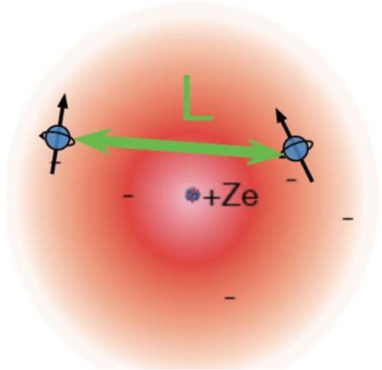
wikipedia.org

Charge and spin transport

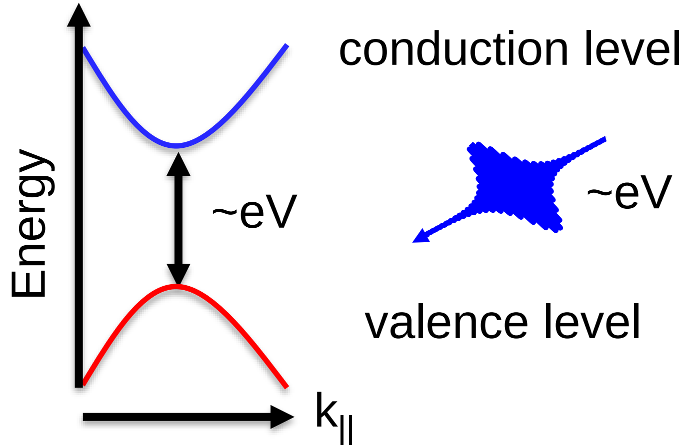


Nano Lett. 19,1074 (2019)

Order in materials

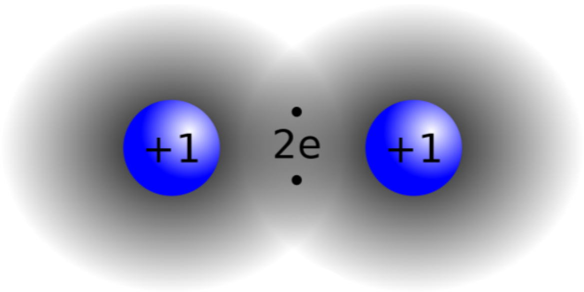


Direct interaction with visible light



The Role of Charges and Spins in Condensed Matter

Bonding of atoms to molecules and solids



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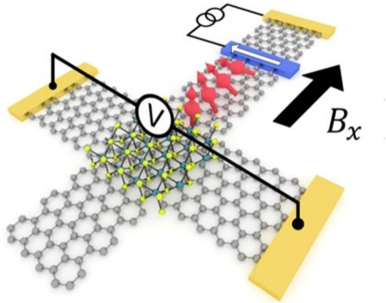
Charge and spin transport



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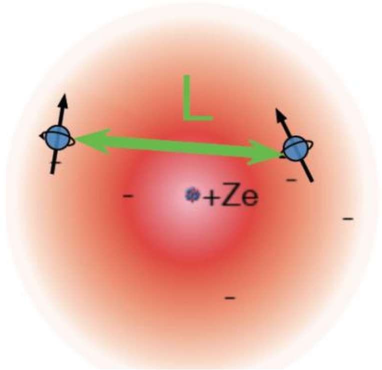

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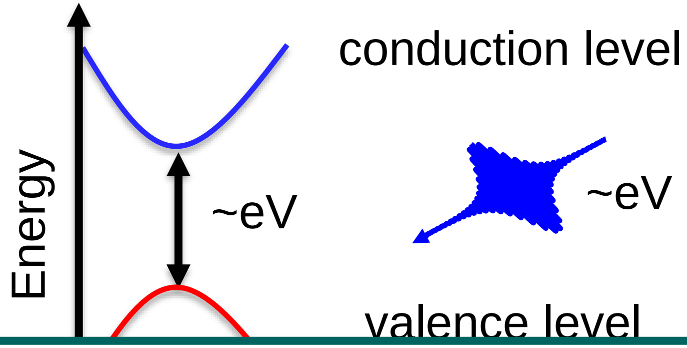
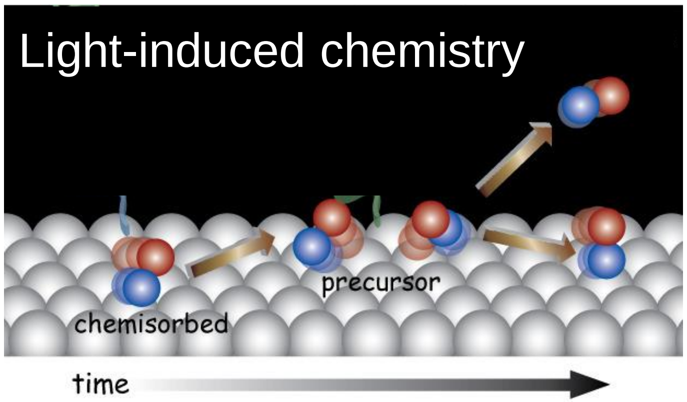


Light is the ideal tool to image and manipulate electrons in solids

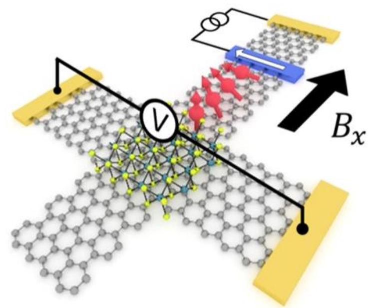
The Role of Charges and Spins in Condensed Matter

Bonding of atoms to molecules and solids



Science 339, 1302-1305 (2013)

Charge and spin transport



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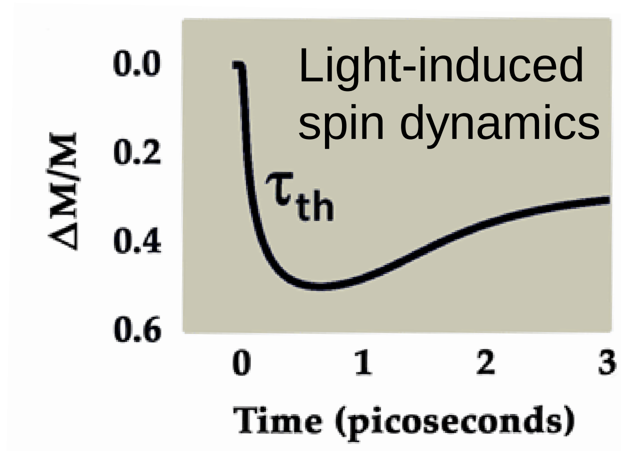

Phys. Rev. Lett. 76, 4250 (1996)

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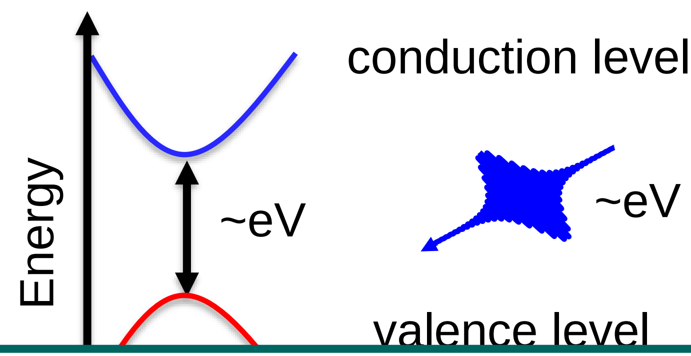
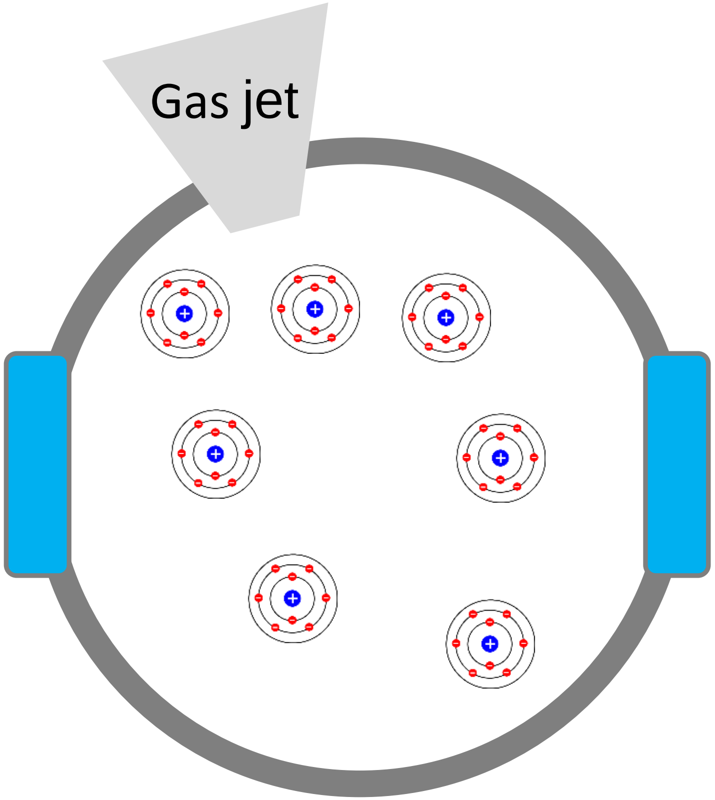


Light is the ideal tool to image and manipulate electrons in solids

Ultrafast Detectors for Electrons in Atoms and Molecules

Investigation of isolated, **non-interacting** particles

Atoms in gas phase



- Isolated non-interacting particles
- Localized wave functions on individual particles
- Well separated electronic states

Ultrafast Detectors for Electrons in Atoms and Molecules

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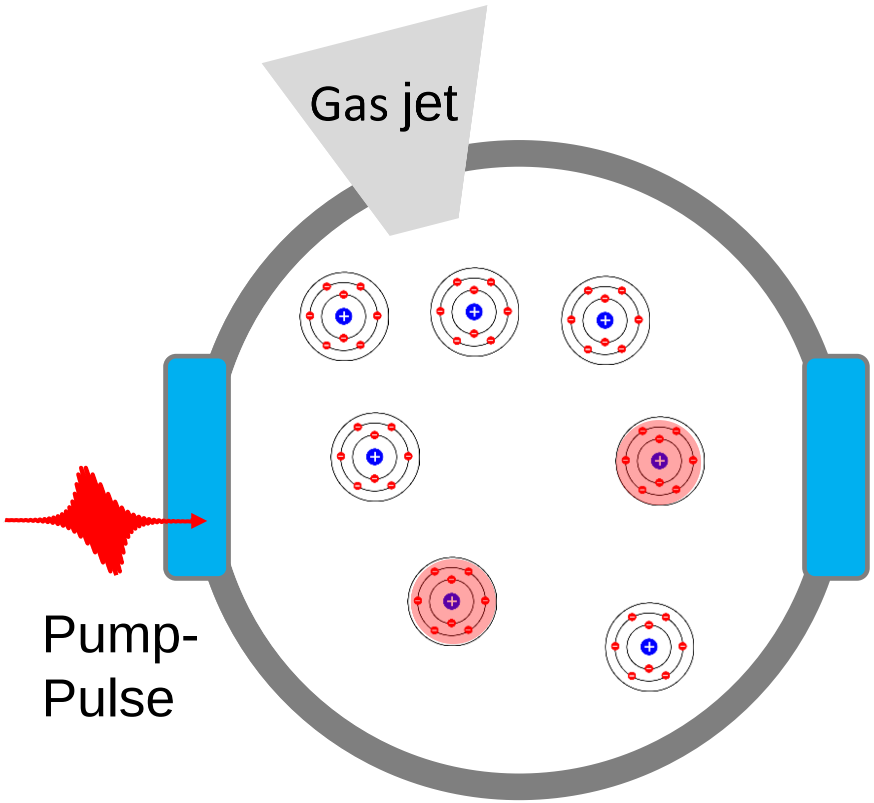
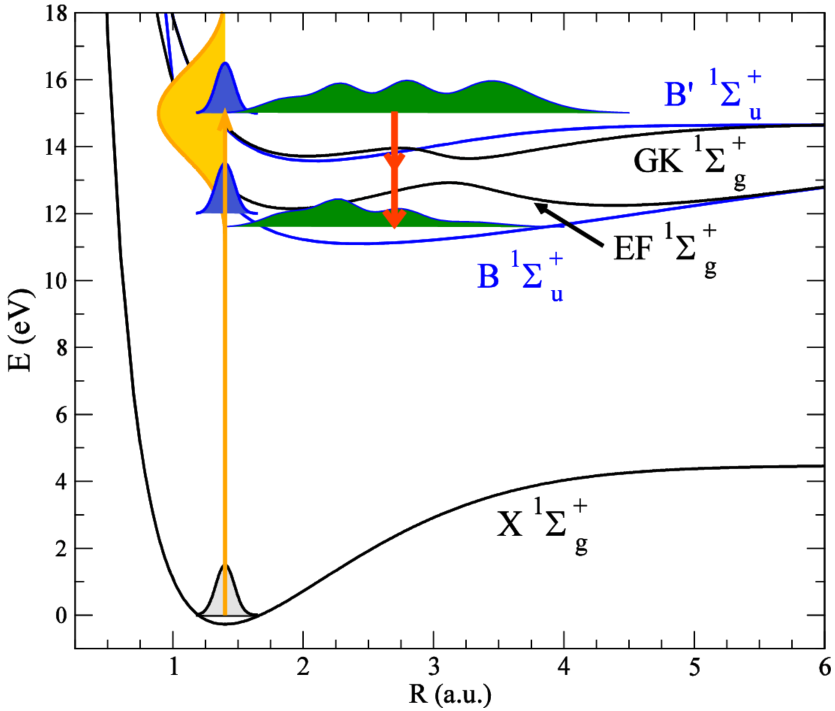


Ultrafast electron dynamics



Phys. Rev. A **94**, 023403 (2016)

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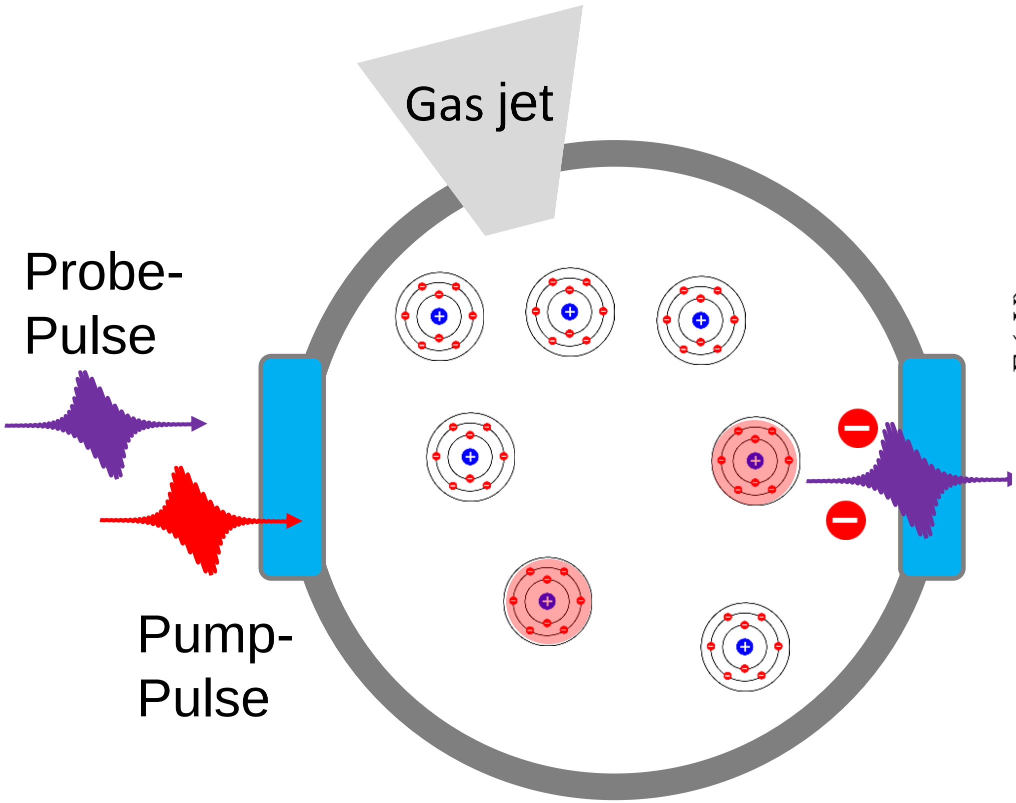

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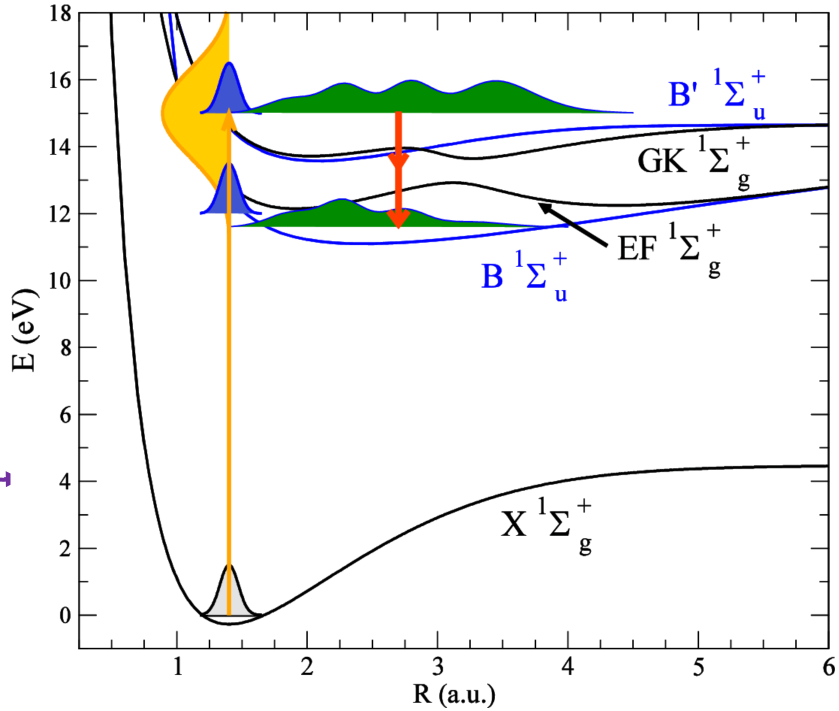

Phys. Rev. A **94**, 023403 (2016)

Ultrafast Detectors for Electrons in Atoms and Molecules

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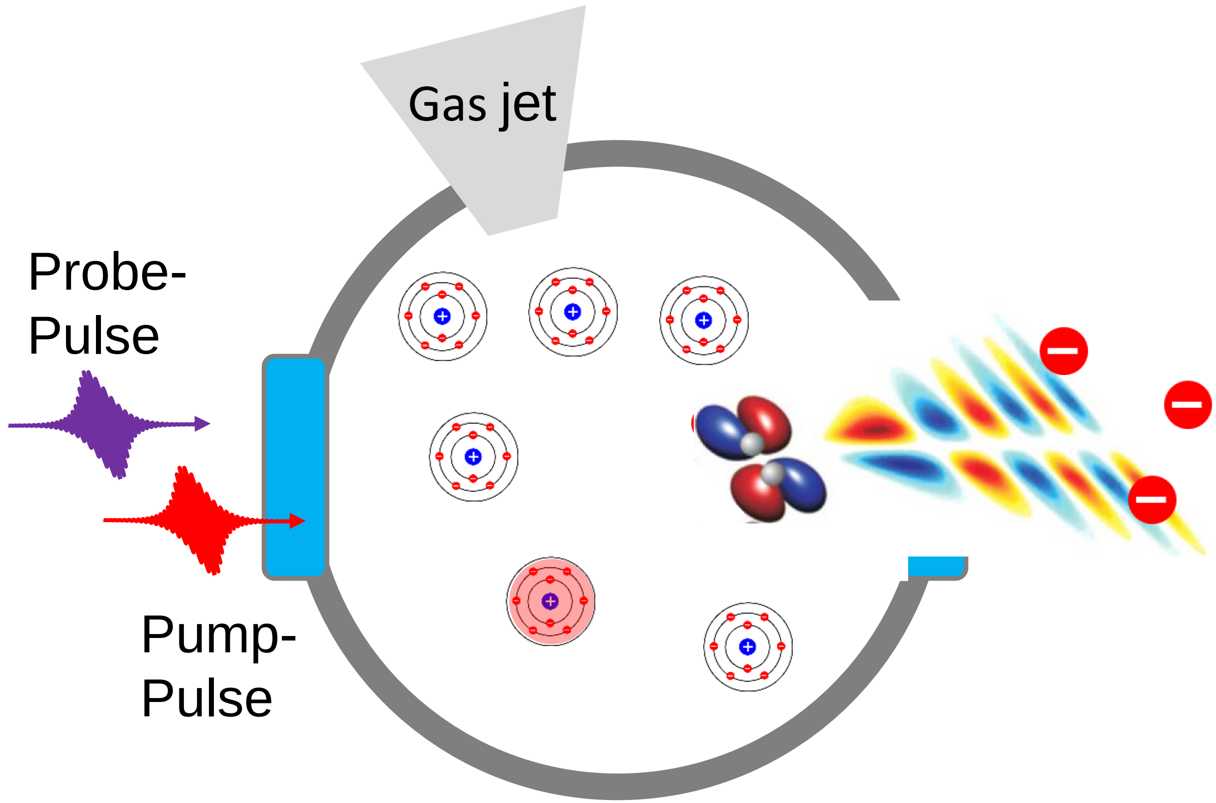
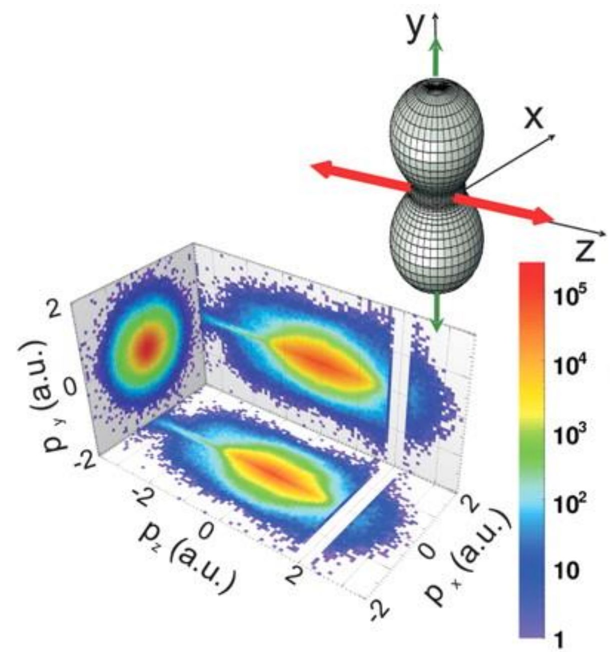
Orbital Tomography



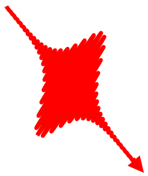


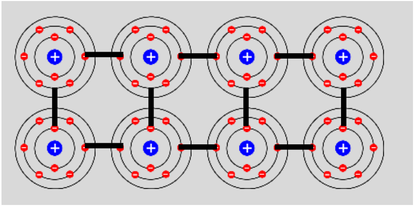
Electron and wave packet dynamics of individual molecules on their intrinsic as to fs timescale

Meckel M et al 2008 Science 320 1478–82

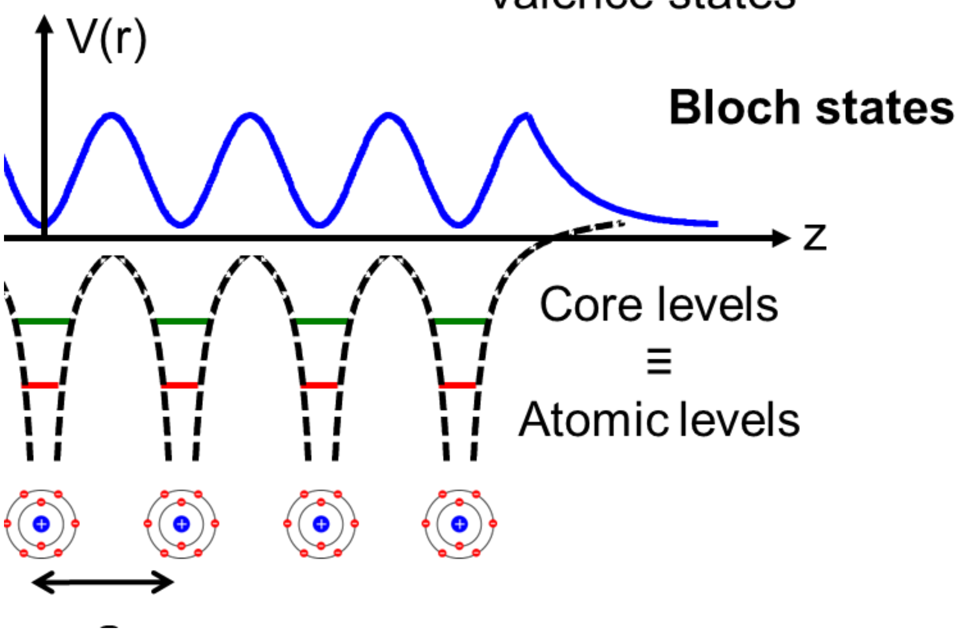
Ultrafast Dynamics in Solids

Optically excited solids





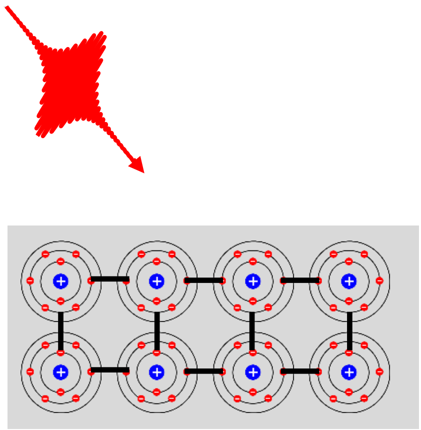
Electronic degree of freedom



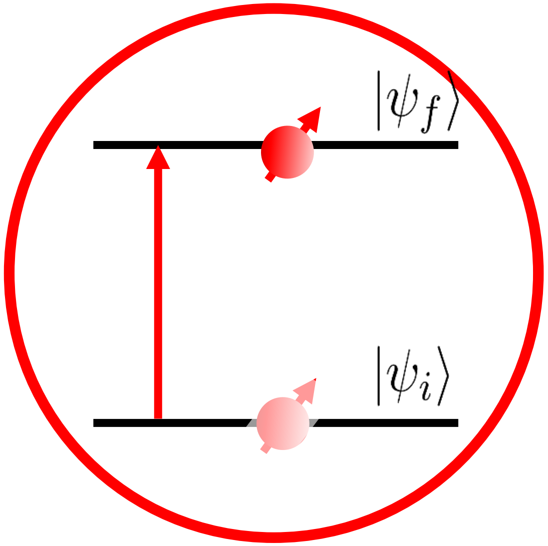
- Coupled atoms/particles
- Chemical bonding
- Coupling between different degrees of freedom

Ultrafast Dynamics in Solids

Optically excited solids



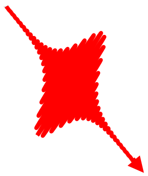
Electronic degree of freedom

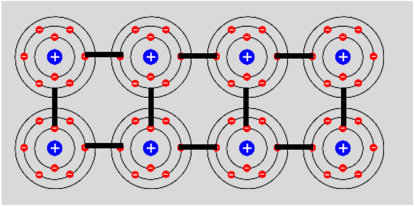


- Interaction of light with electronic degree of freedom

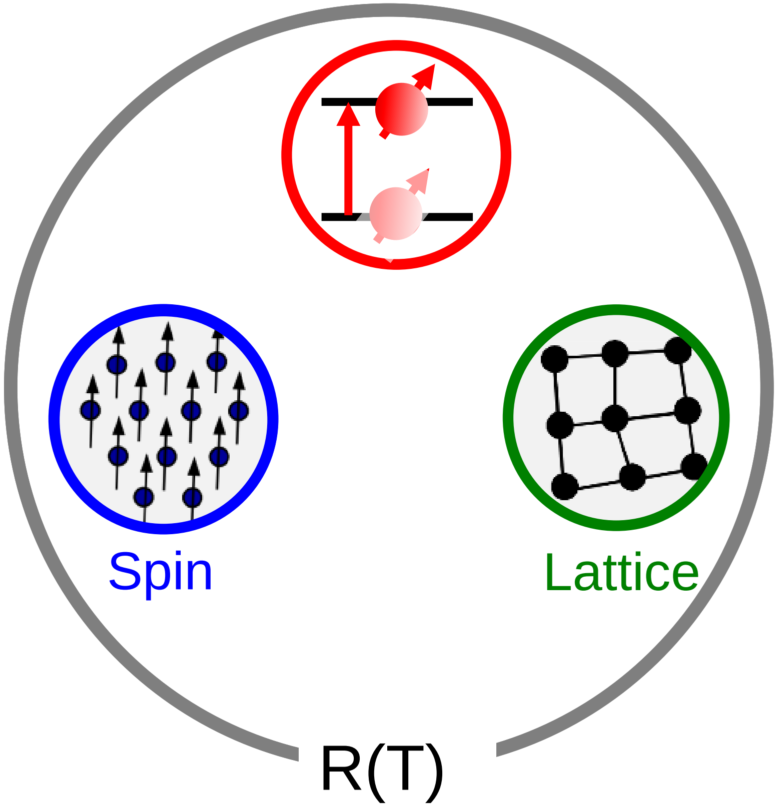
Ultrafast Dynamics in Solids

Optically excited solids





- Interaction of light with electronic degree of freedom
- Coupling of electrons to other degrees of freedom



Ultrafast Dynamics in Solids

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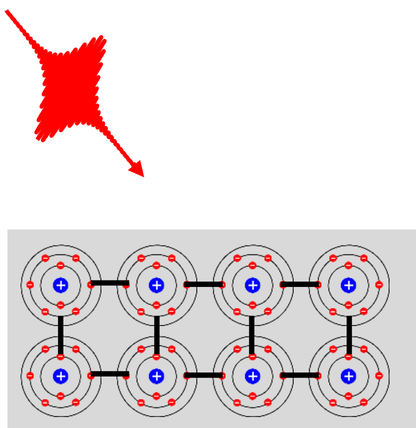

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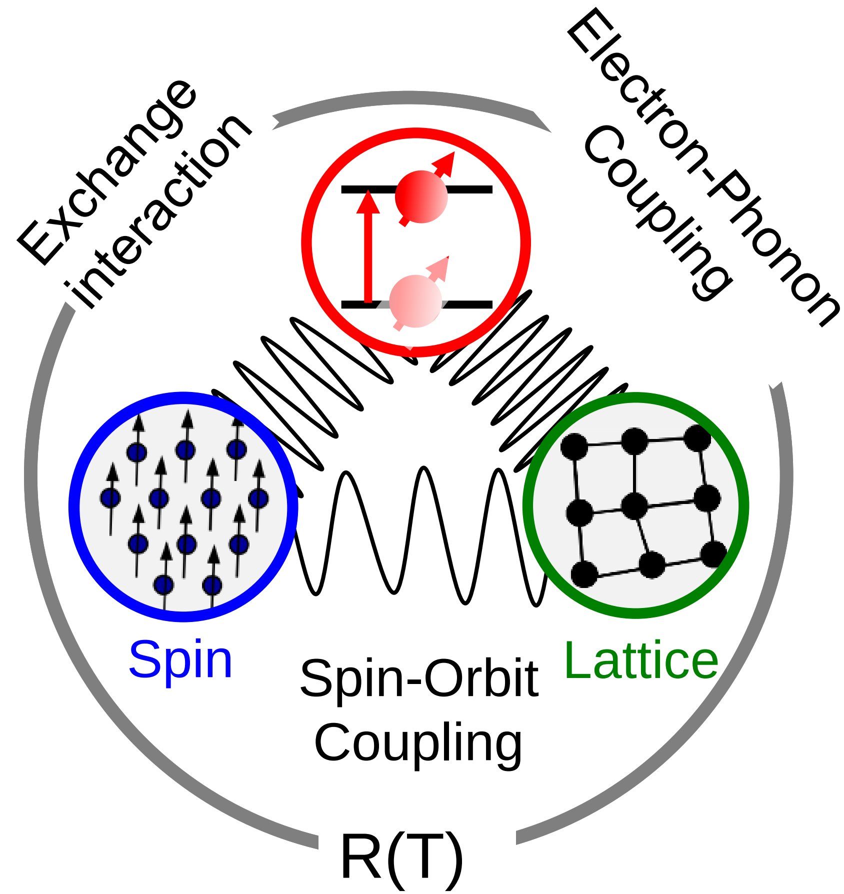

Ultrafast Dynamics in Solids

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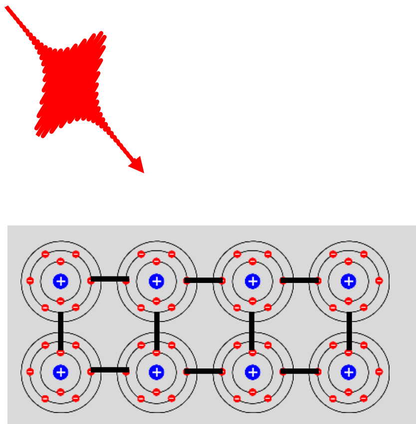
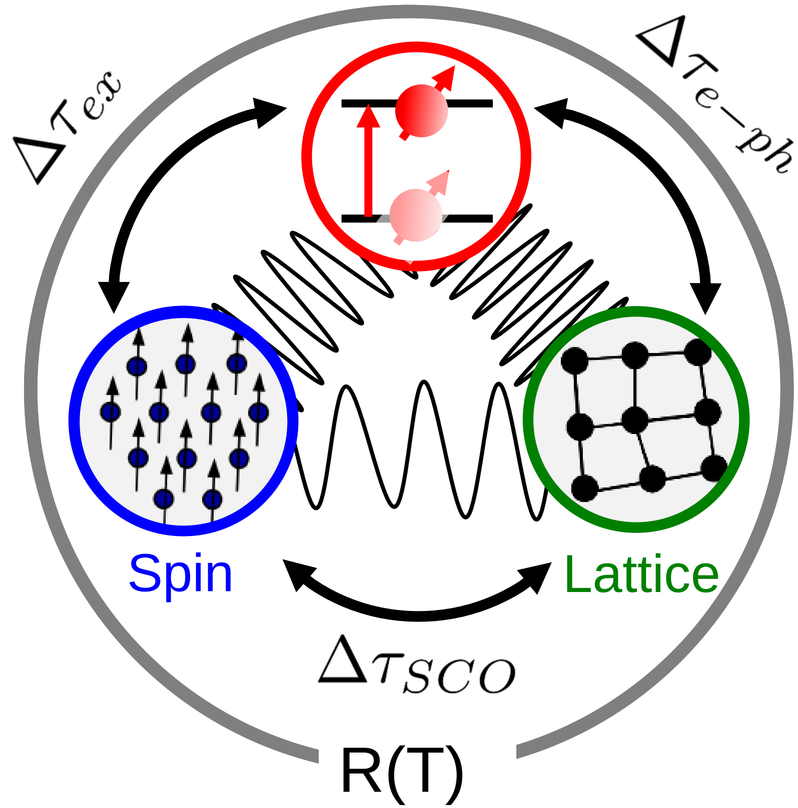
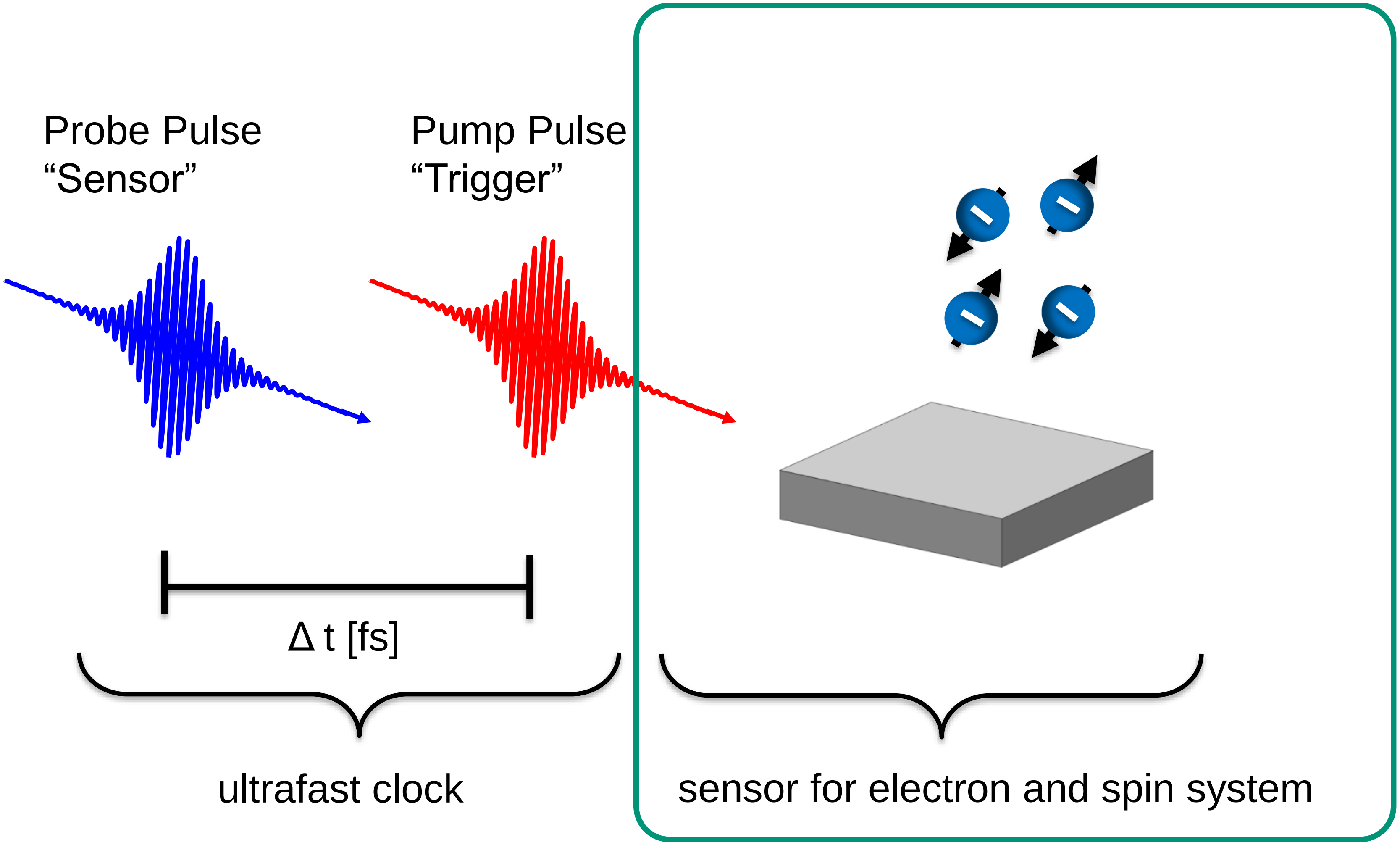




- Interaction of light with electronic degree of freedom
- Coupling of electrons to other degrees of freedom
- Coupling strength reflected by scattering rate $\Gamma = \frac{1}{\Delta\tau_i}$

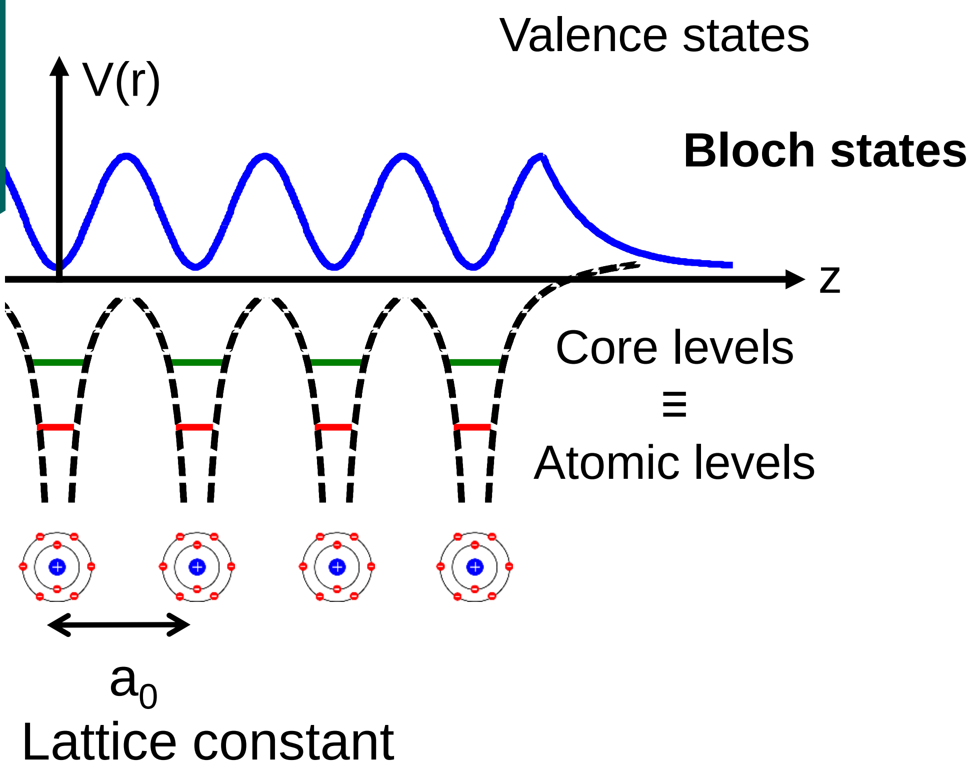
Ultrafast Spectroscopy



Pump-Probe Photoemission Spectroscopy

Signatures of Charge and Spin Carriers in Solids

Electronic wave functions in a solid



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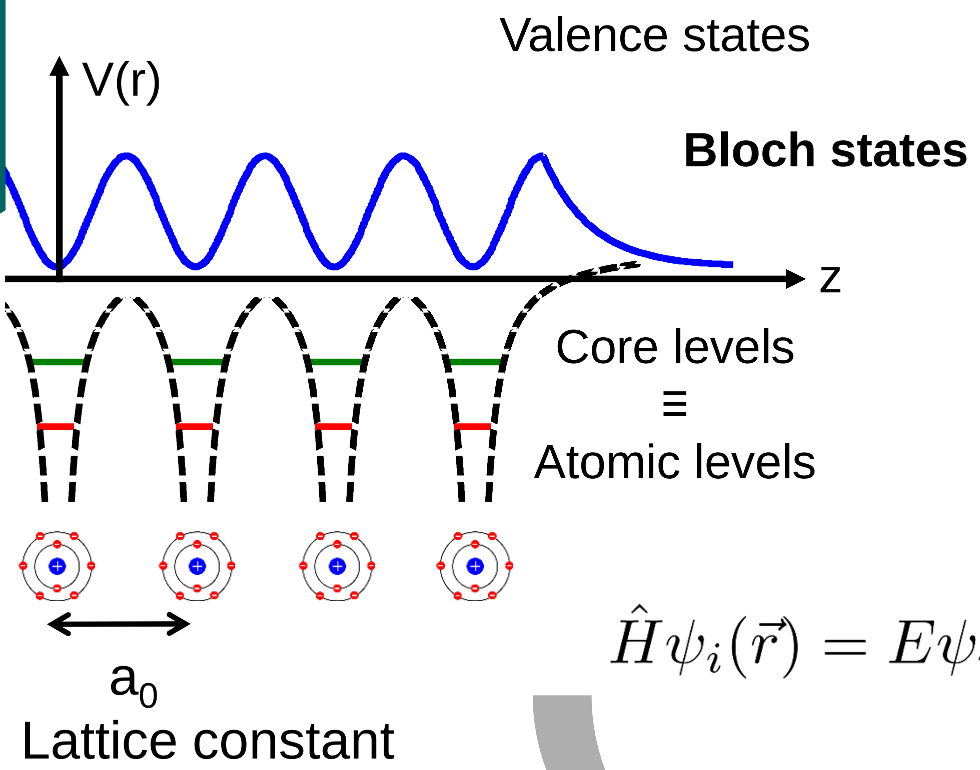
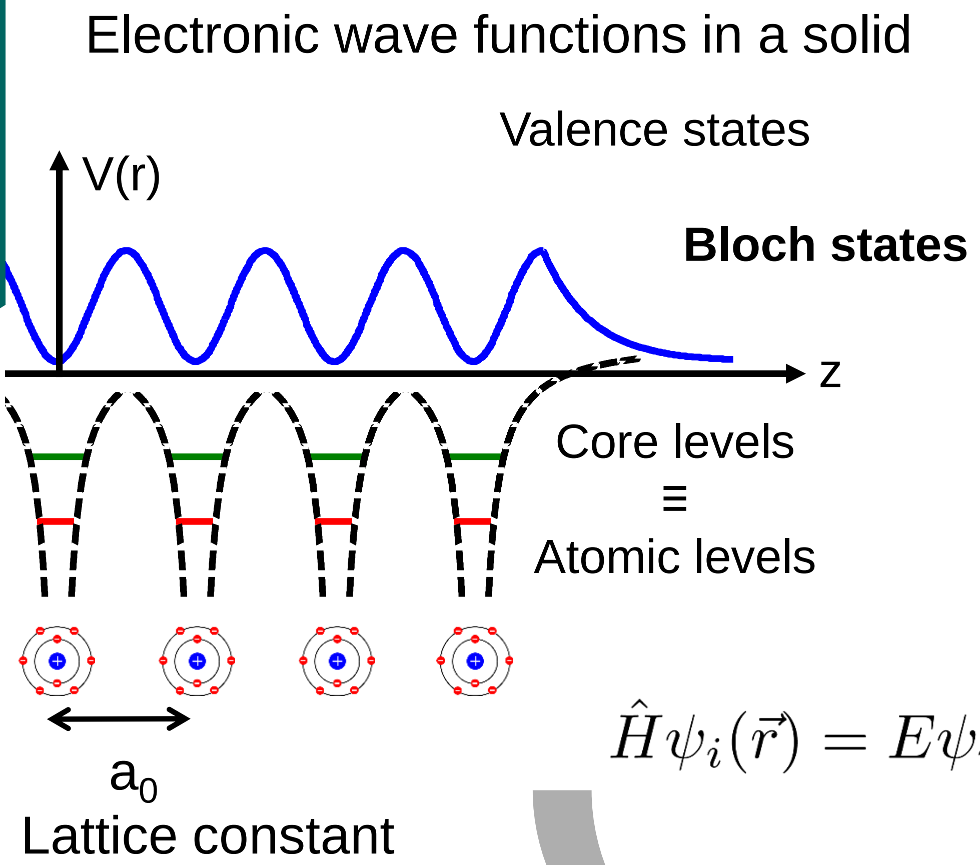


$$\hat{H}\psi_i(\vec{r}) = E\psi_i(\vec{r})$$

Signatures of Charge and Spin Carriers in Solids



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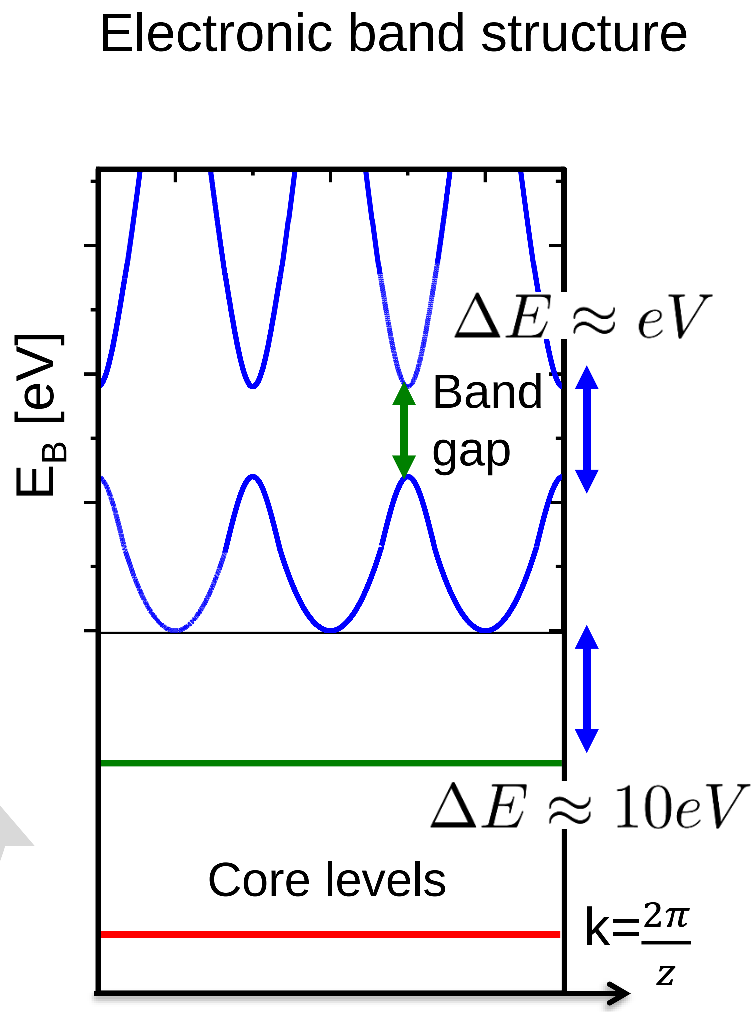
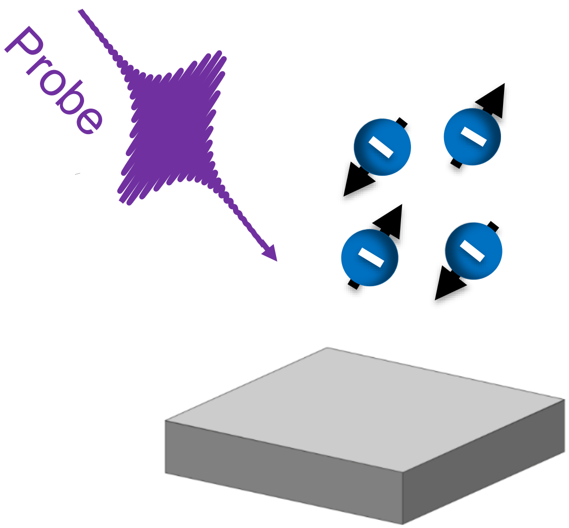
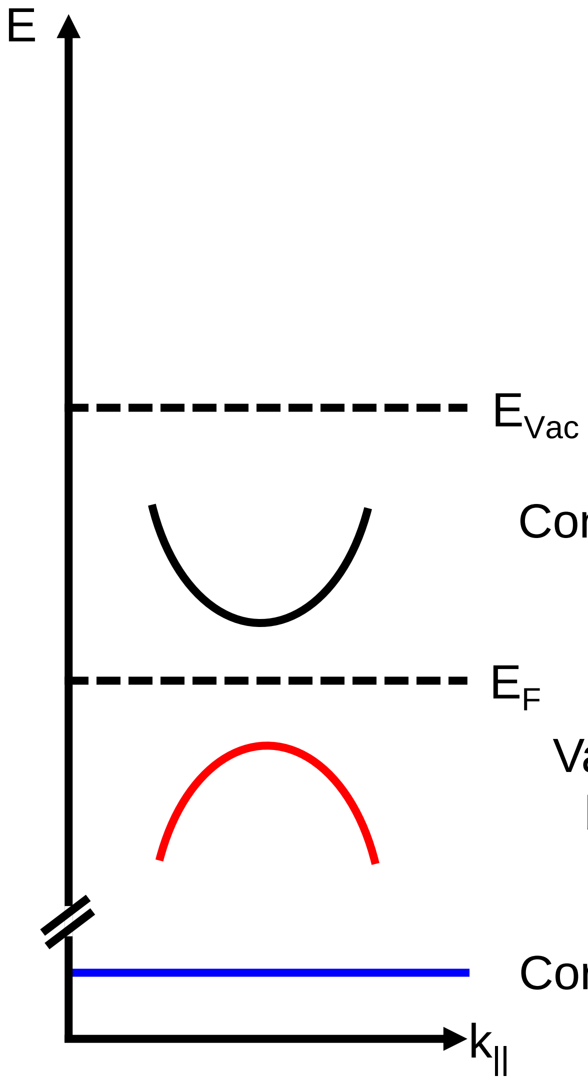


Electronic band structure is experimental observable of electronic wave function in energy and momentum space

Photoemission and the Band Structure Approach

Projection of band structure into the vacuum





The 2021 Ultrafast Spectroscopic Probes of Condensed Matter Roadmap – Chapter 8
J. Phys.: Condens. Matter 33 (2021) 353001

Photoemission and the Band Structure Approach

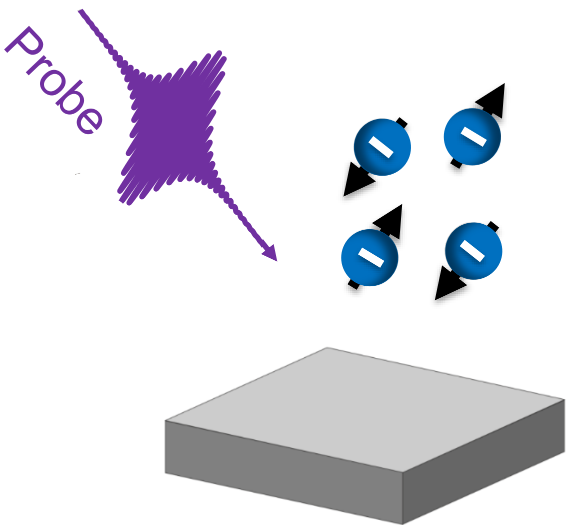
Projection of band structure into the vacuum

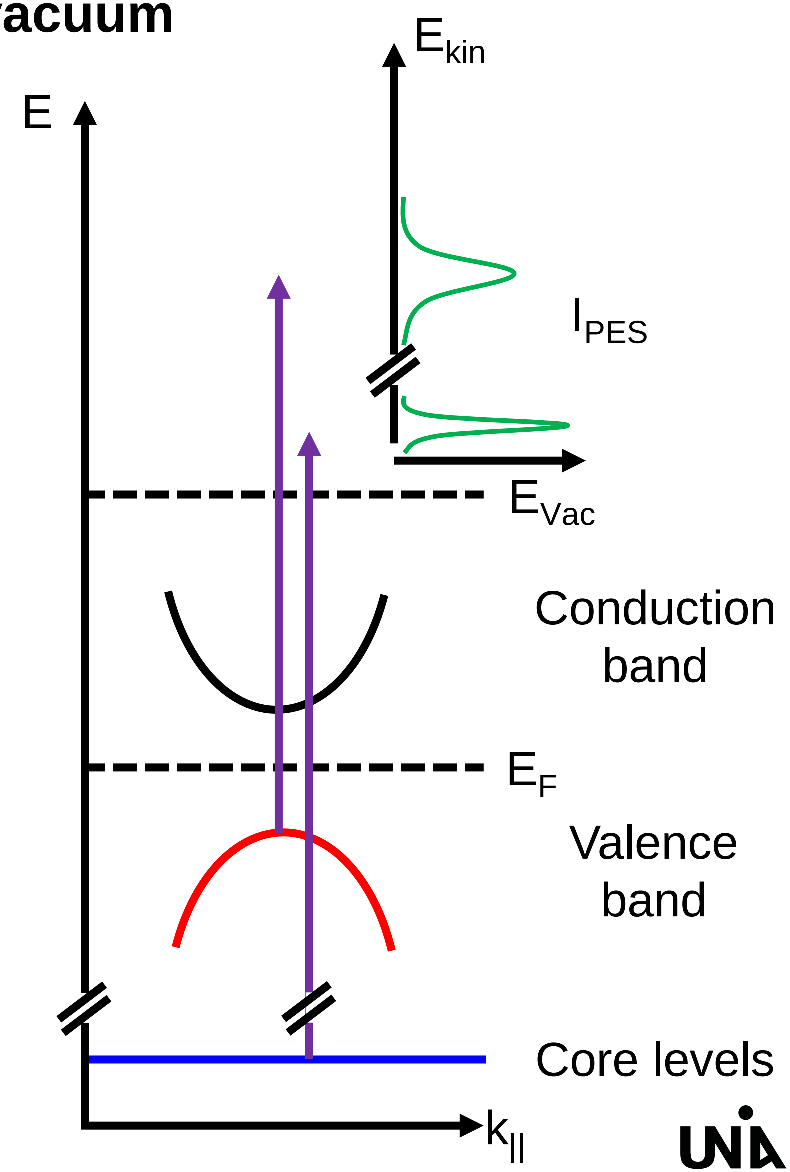
energy conservation

$$E_{kin}(\vec{k}_f) = h\nu - E_B(\vec{k}_i)$$

momentum conservation

$$\vec{k}_f = \vec{k}_i + \vec{k}_{h\nu} + \vec{G}$$





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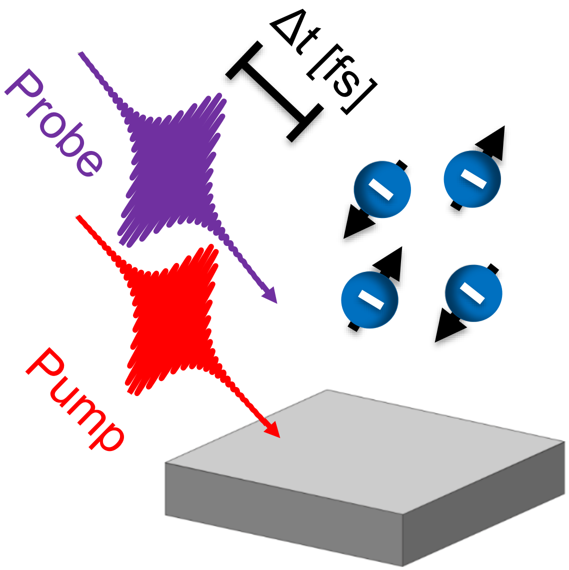

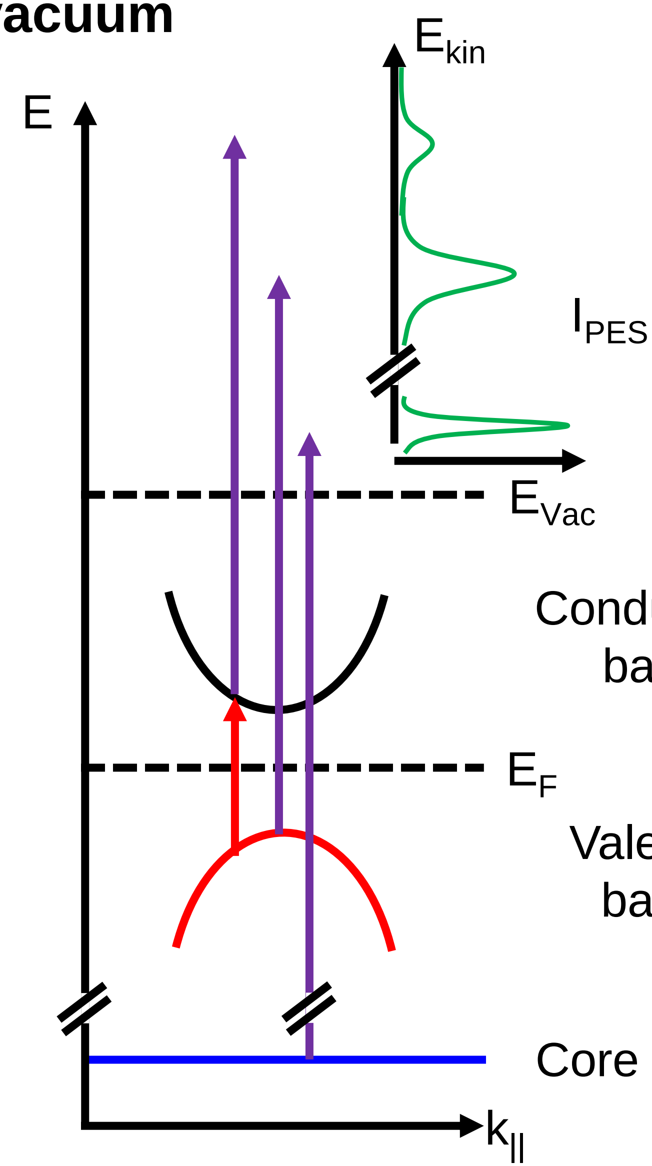

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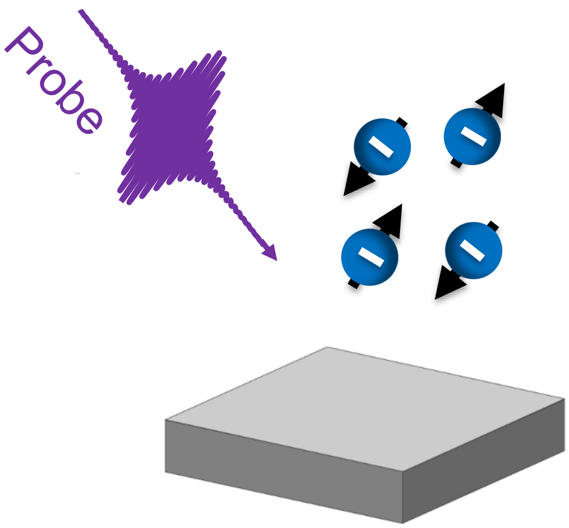
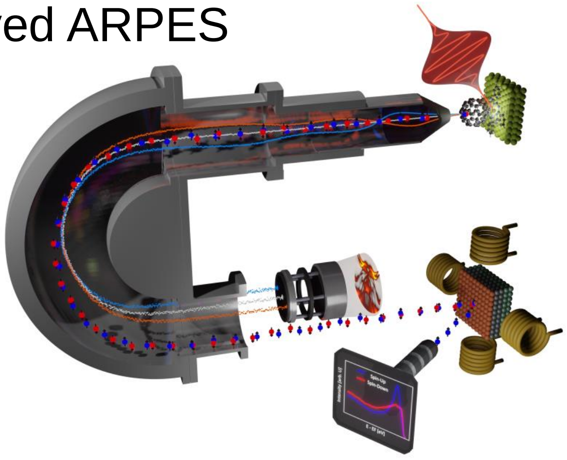
Photoemission and the Band Structure Approach

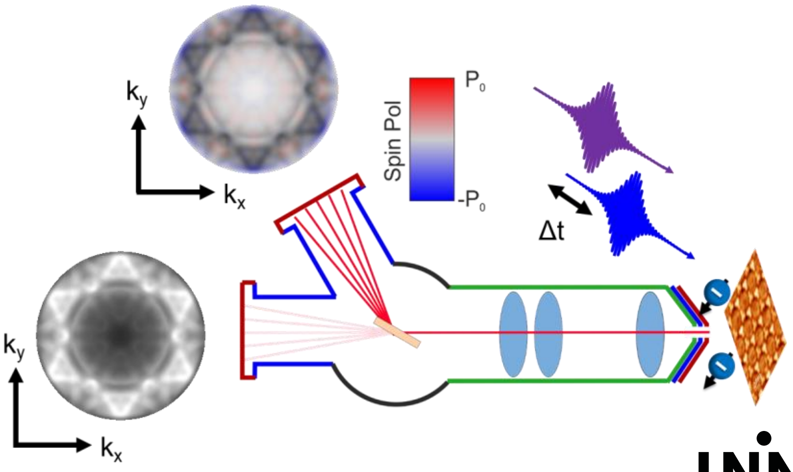
Multidimensional (time, spin, momentum, energy) photoemission spectroscopy fs XUV light pulses



Spin-resolved ARPES



Spin-resolved Momentum Microscopy



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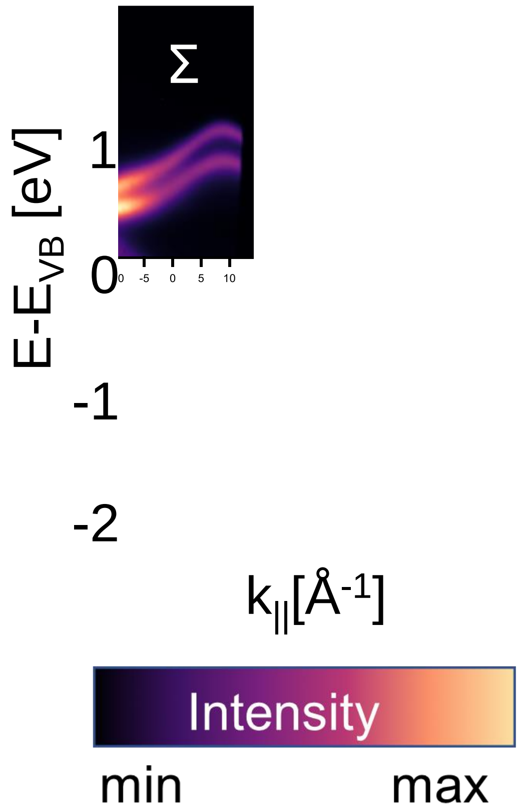
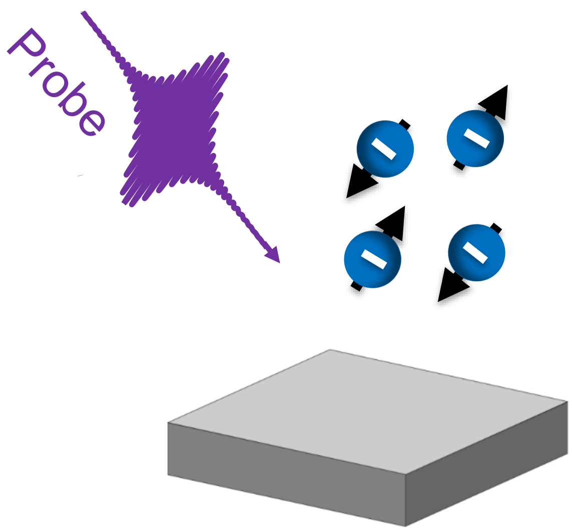
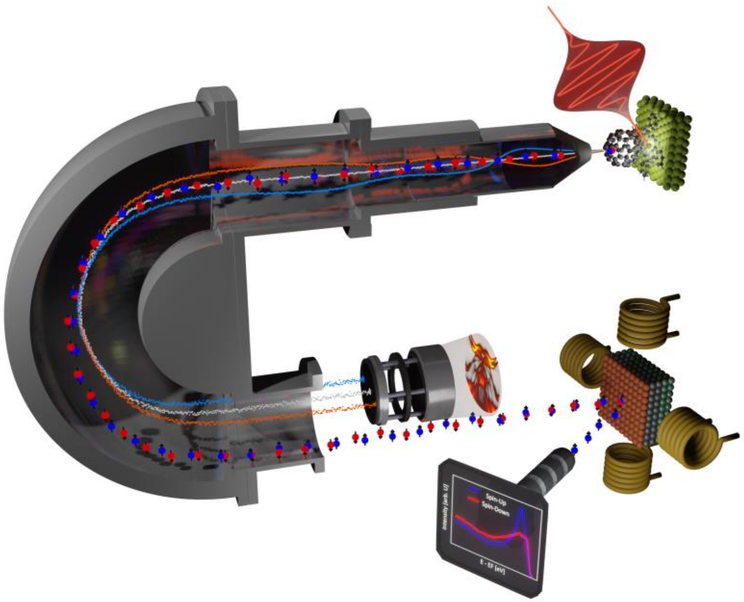
Photoemission and the Band Structure Approach

(Spin-) ARPES technique



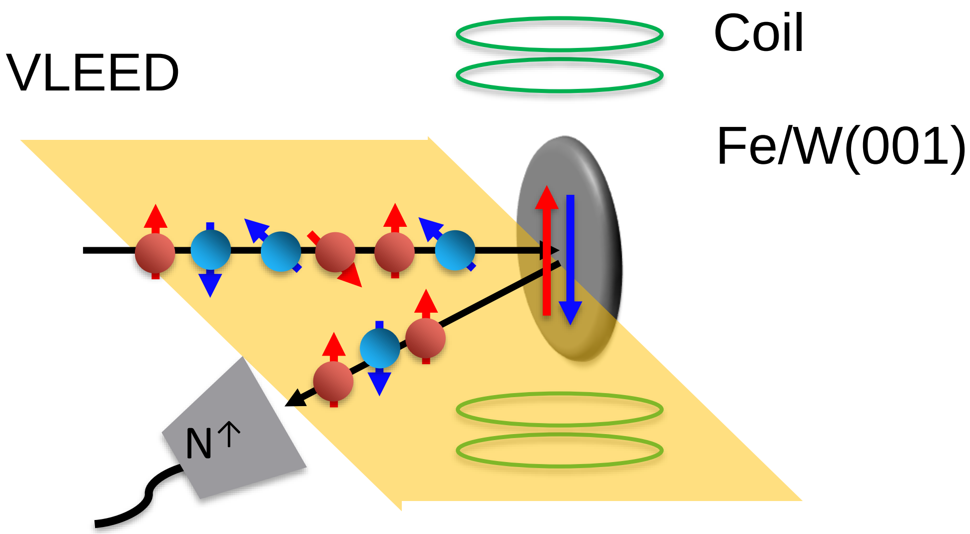


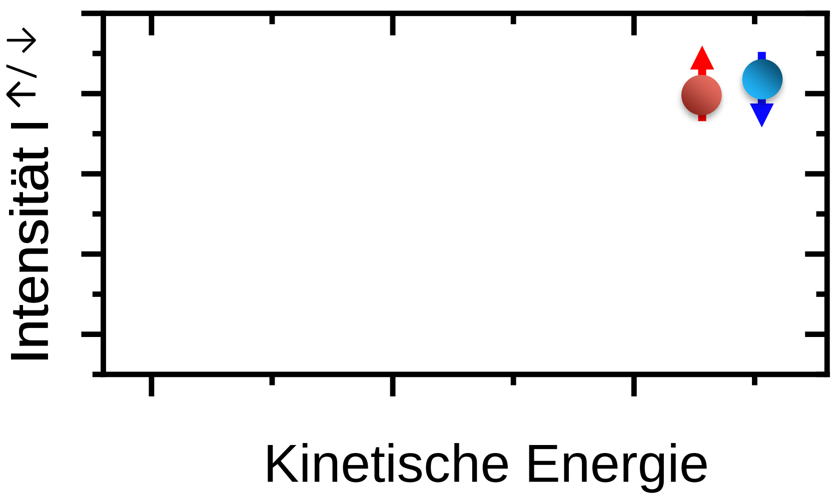


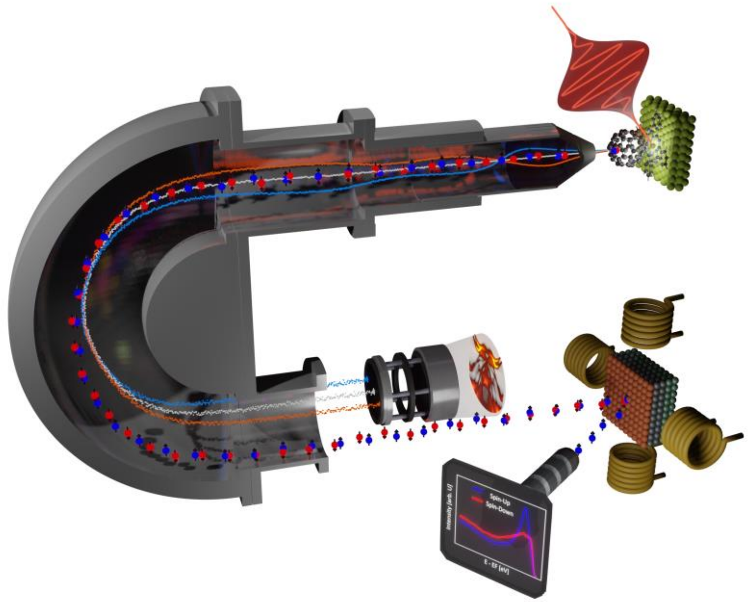
Hemispherical analyzer
with 3D spin filter
(*Focus Ferrum*)

Photoemission and the Band Structure Approach

(Spin-) ARPES technique







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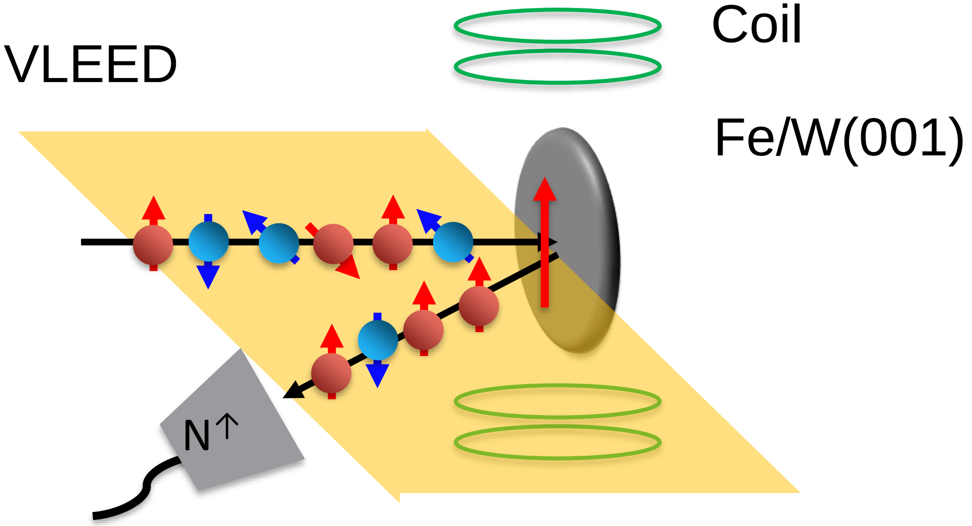

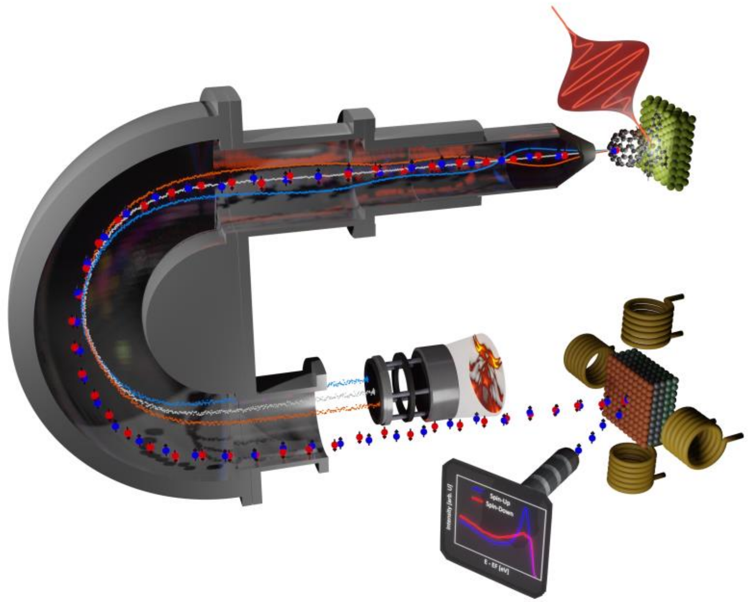

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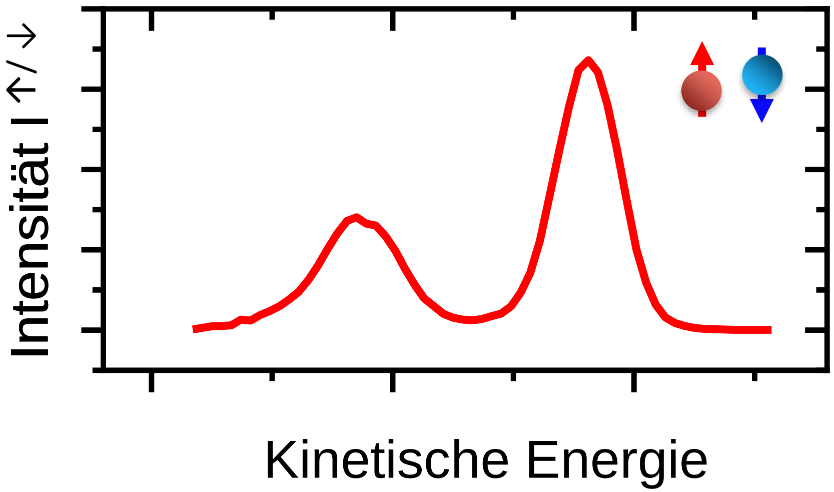
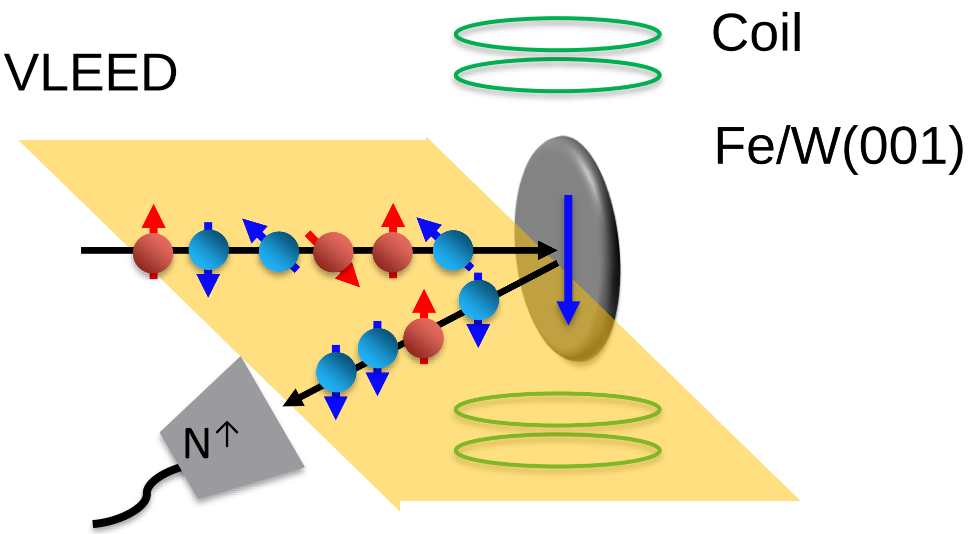
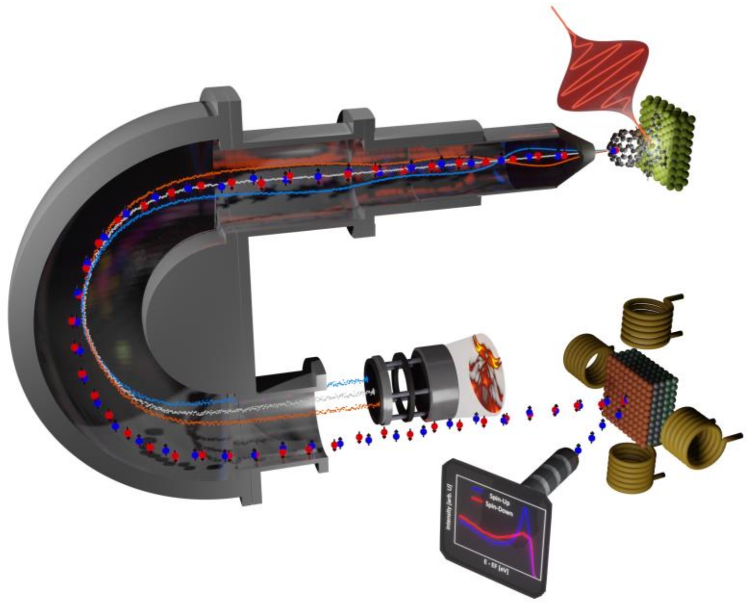
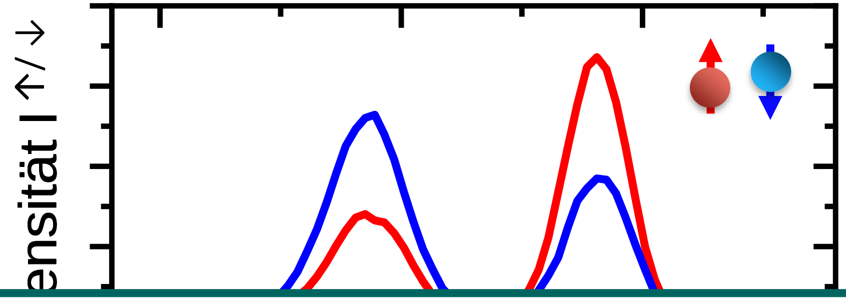


Photoemission and the Band Structure Approach

(Spin-) ARPES technique







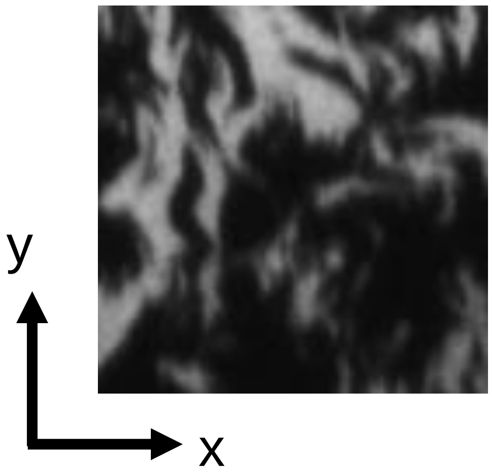
Hemispherical analyzer
with 3D spin filter
(*Focus Ferrum*)

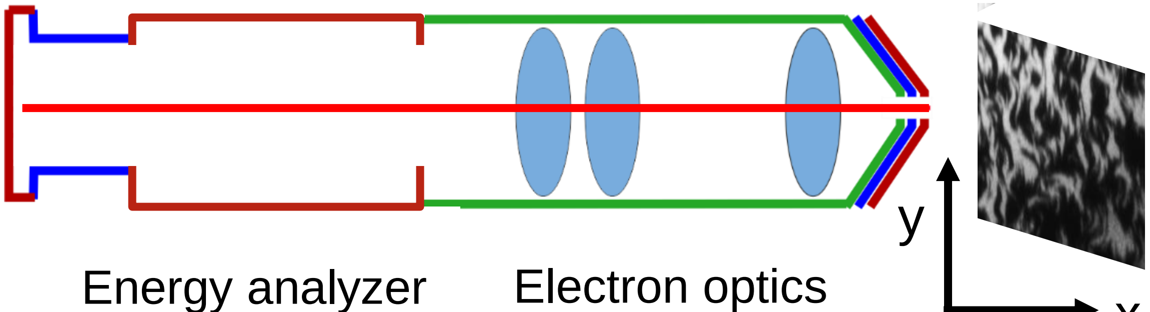
Access to momentum and spin-resolved photoemission yield at selected regions of the band structure

Photoemission and the Band Structure Approach

Momentum Microcopy

Image of sample surface





Photoemission and the Band Structure Approach

Momentum Microcopy

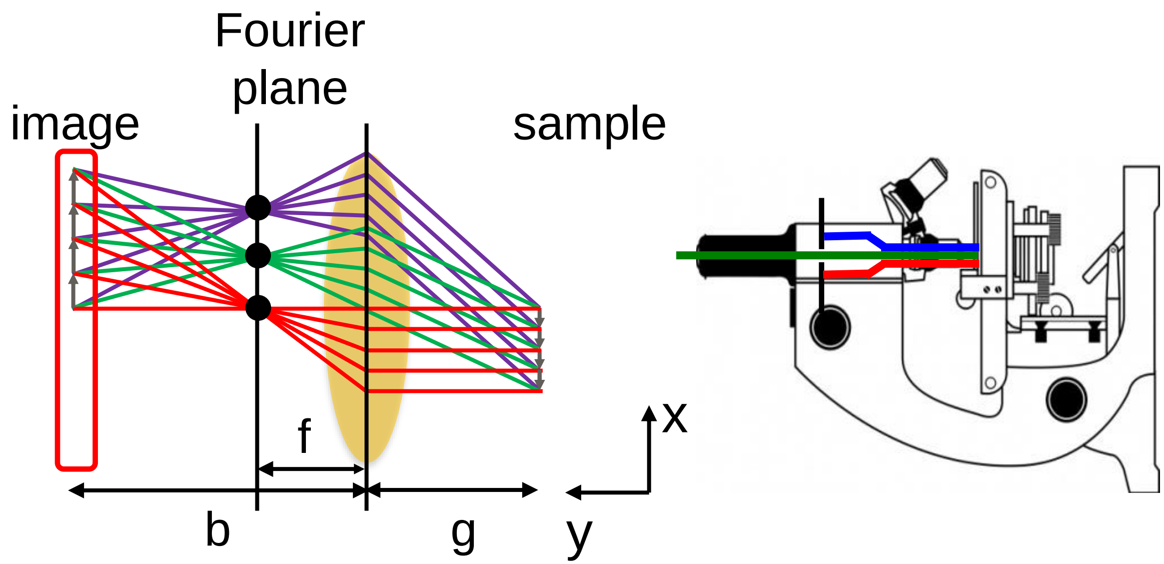
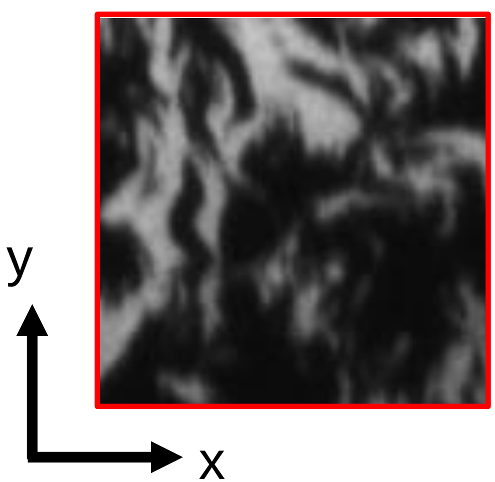
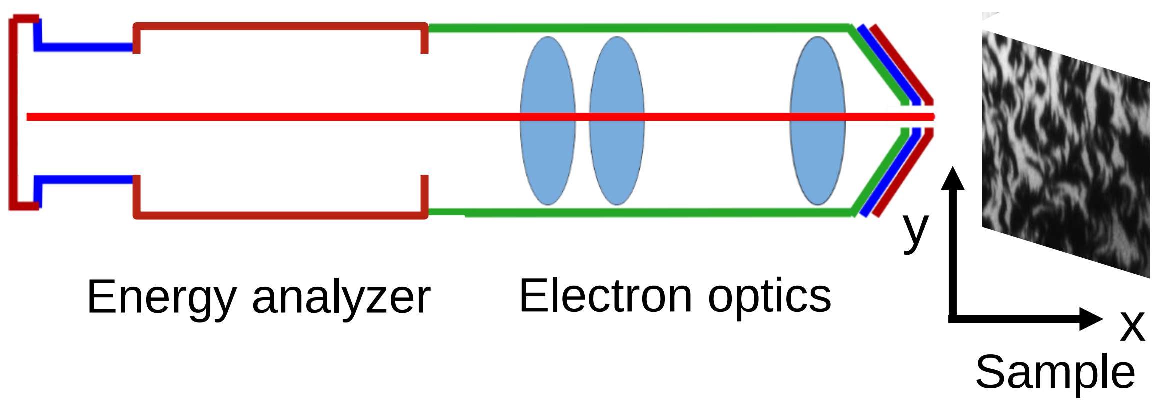


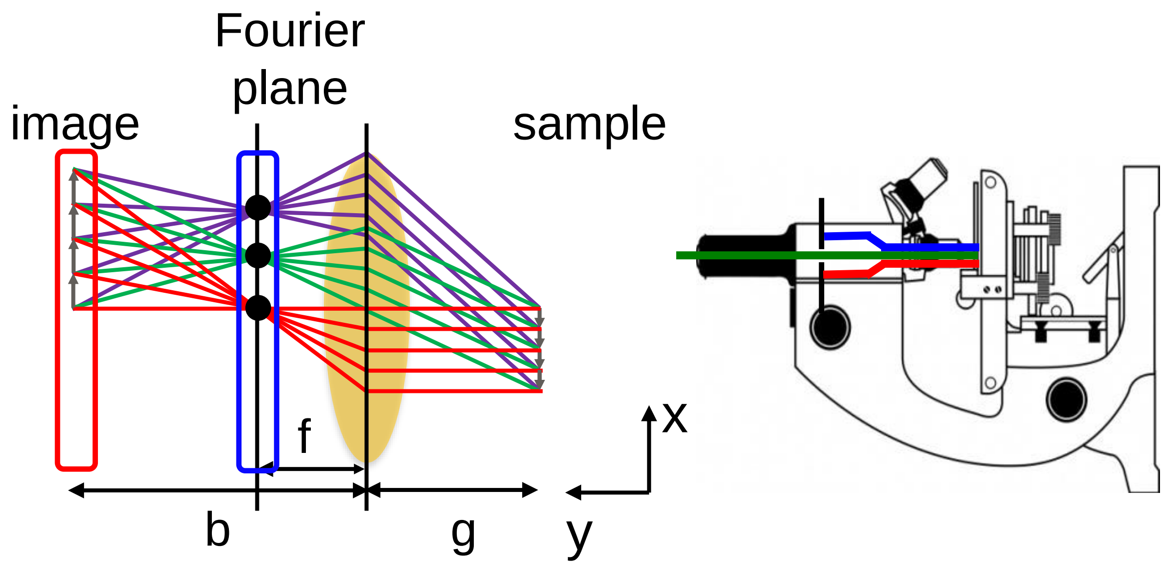
Image of sample surface





Photoemission and the Band Structure Approach

Momentum Microcopy



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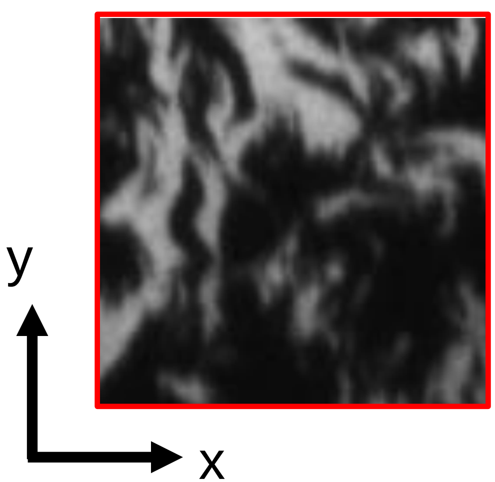
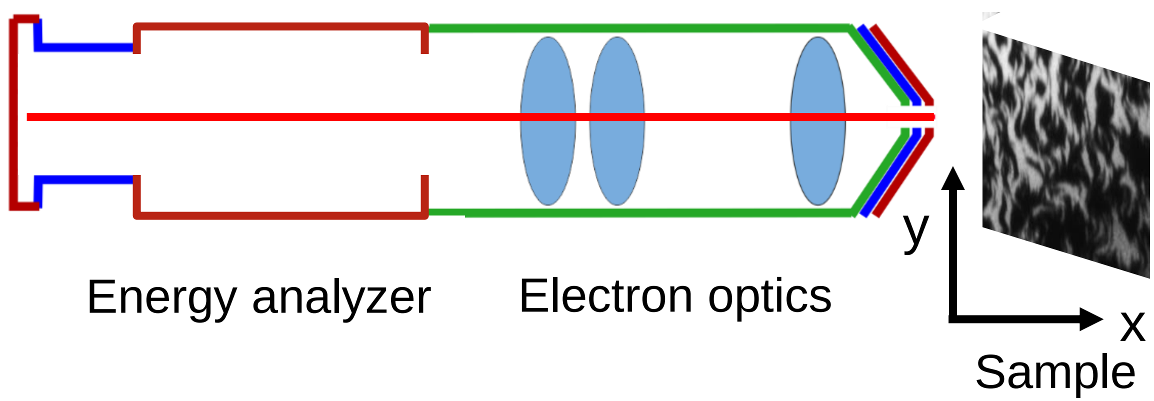
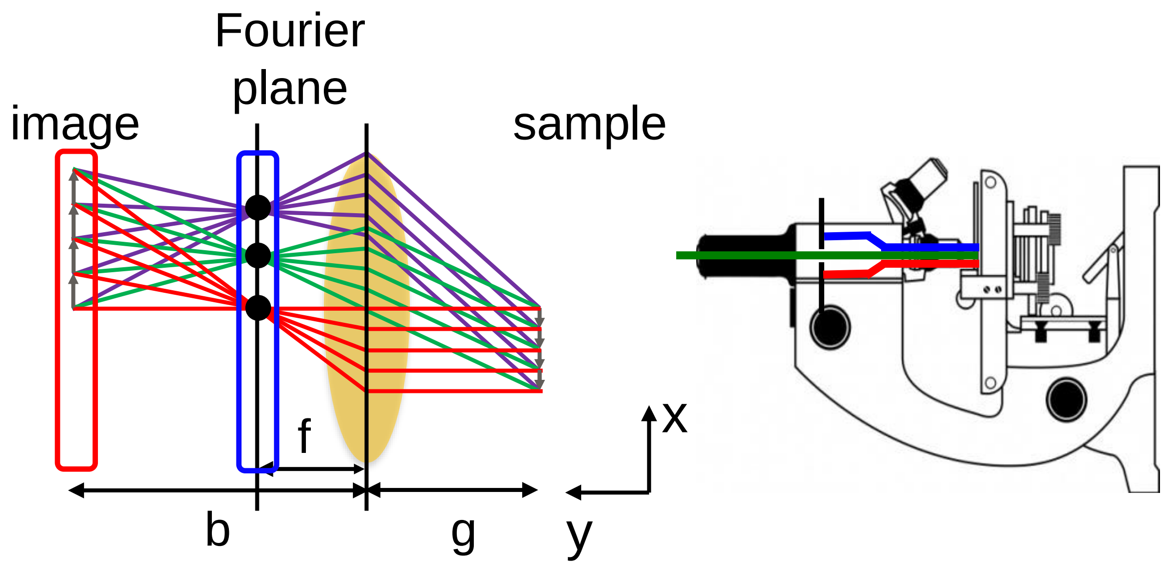
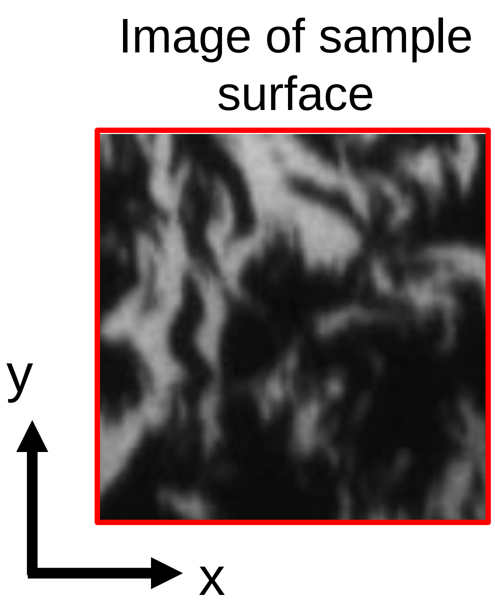


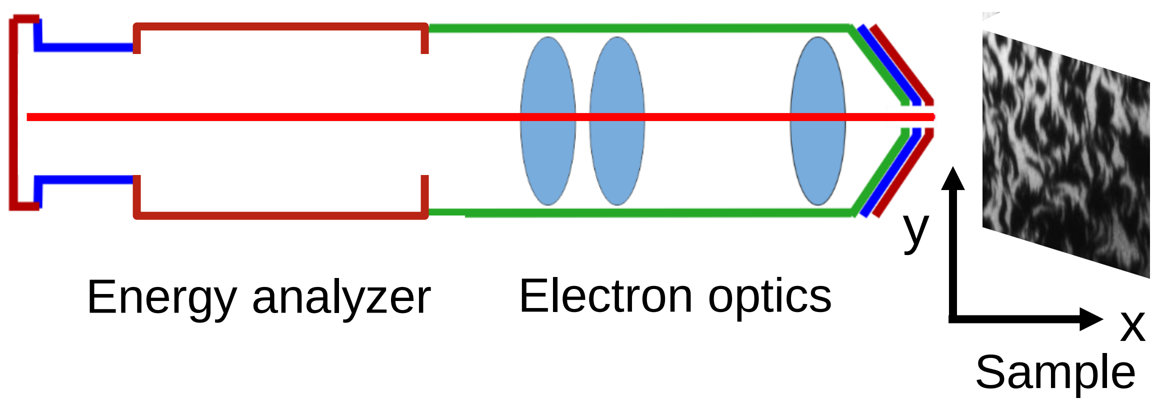


Photoemission and the Band Structure Approach

Momentum Microcopy

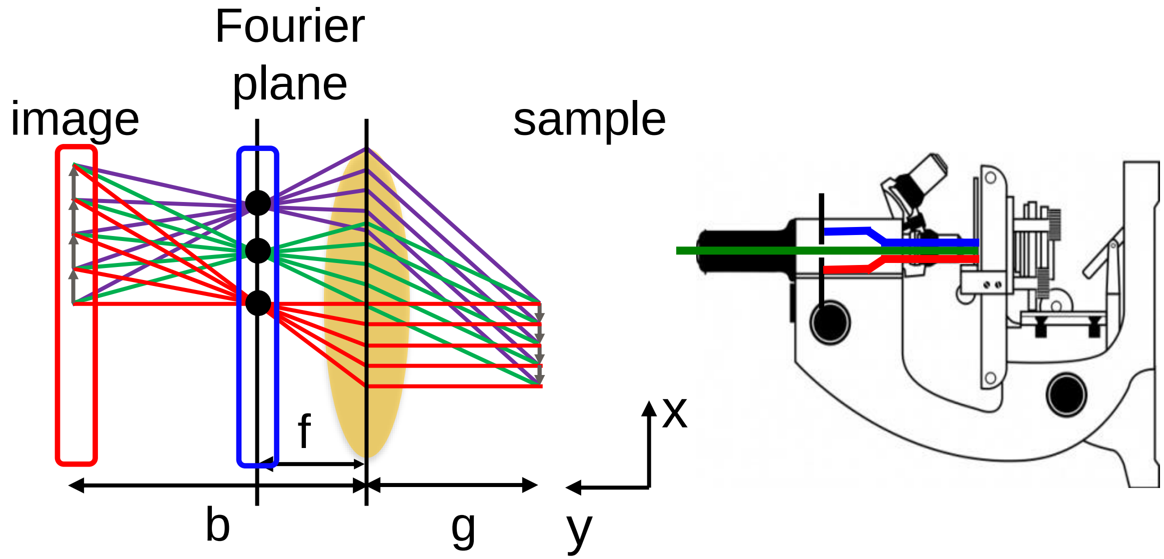




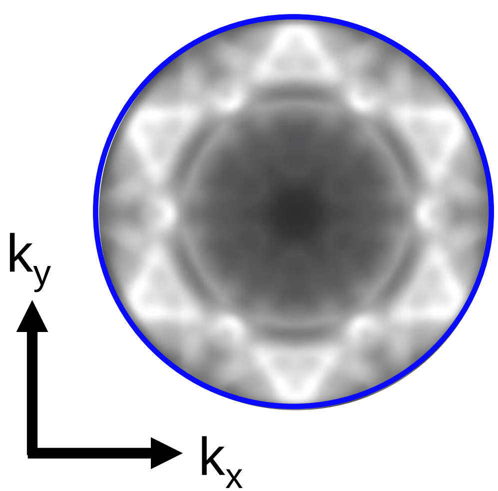


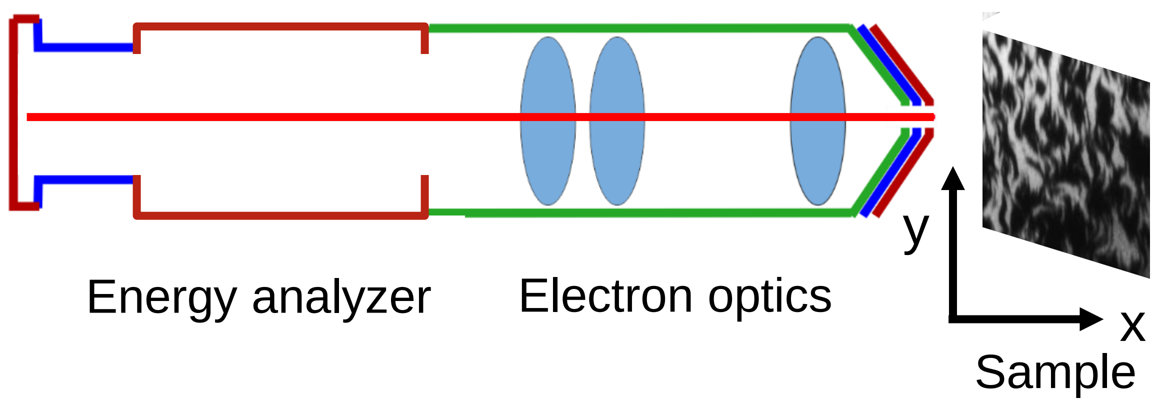
Photoemission and the Band Structure Approach

Momentum Microcopy



Momentum distribution of electrons

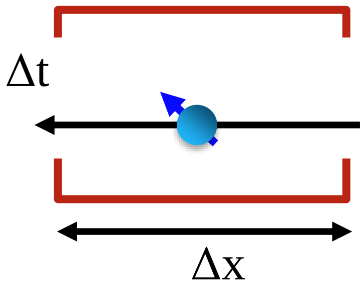


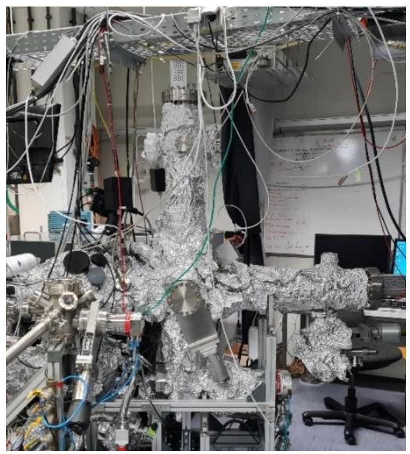


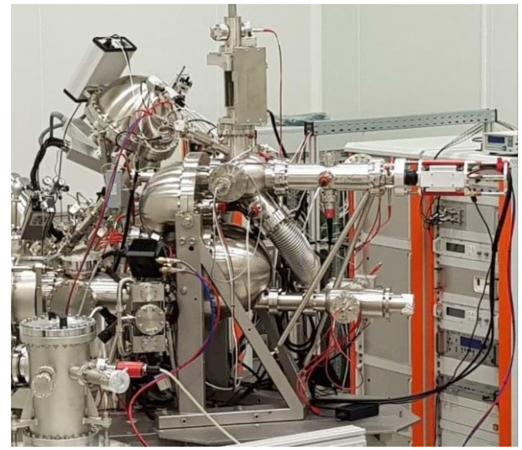
Photoemission and the Band Structure Approach

Momentum Microcopy

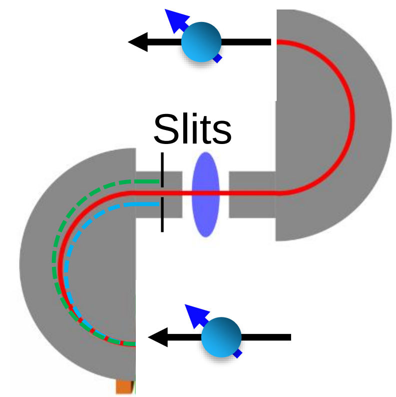
Time of flight energy analyzer



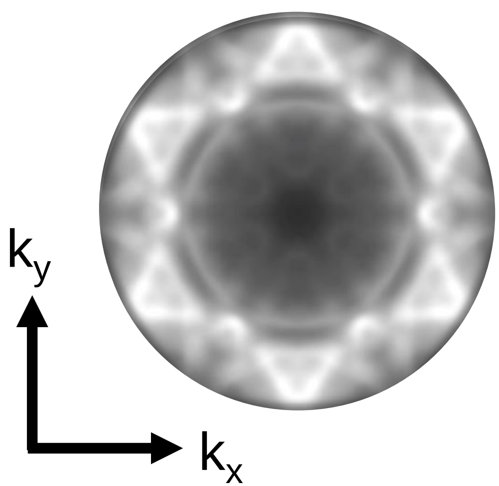


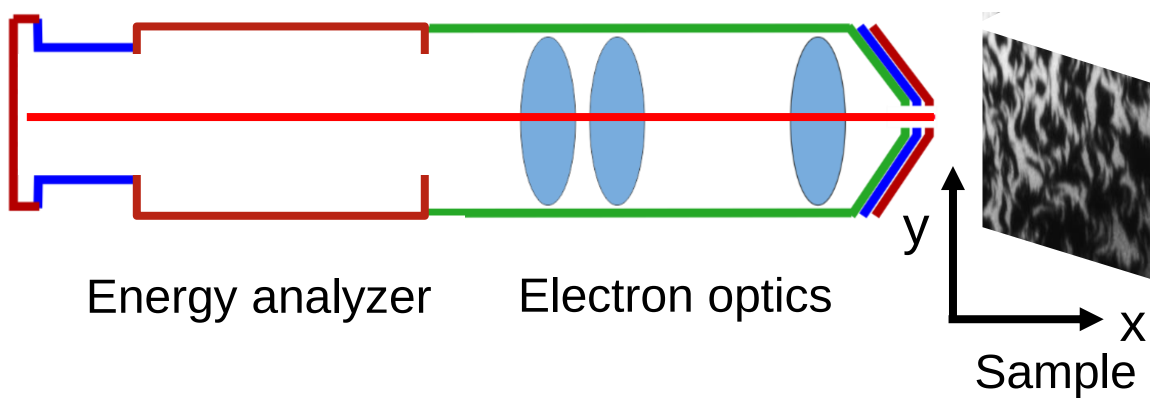


Dispersive energy analyzer



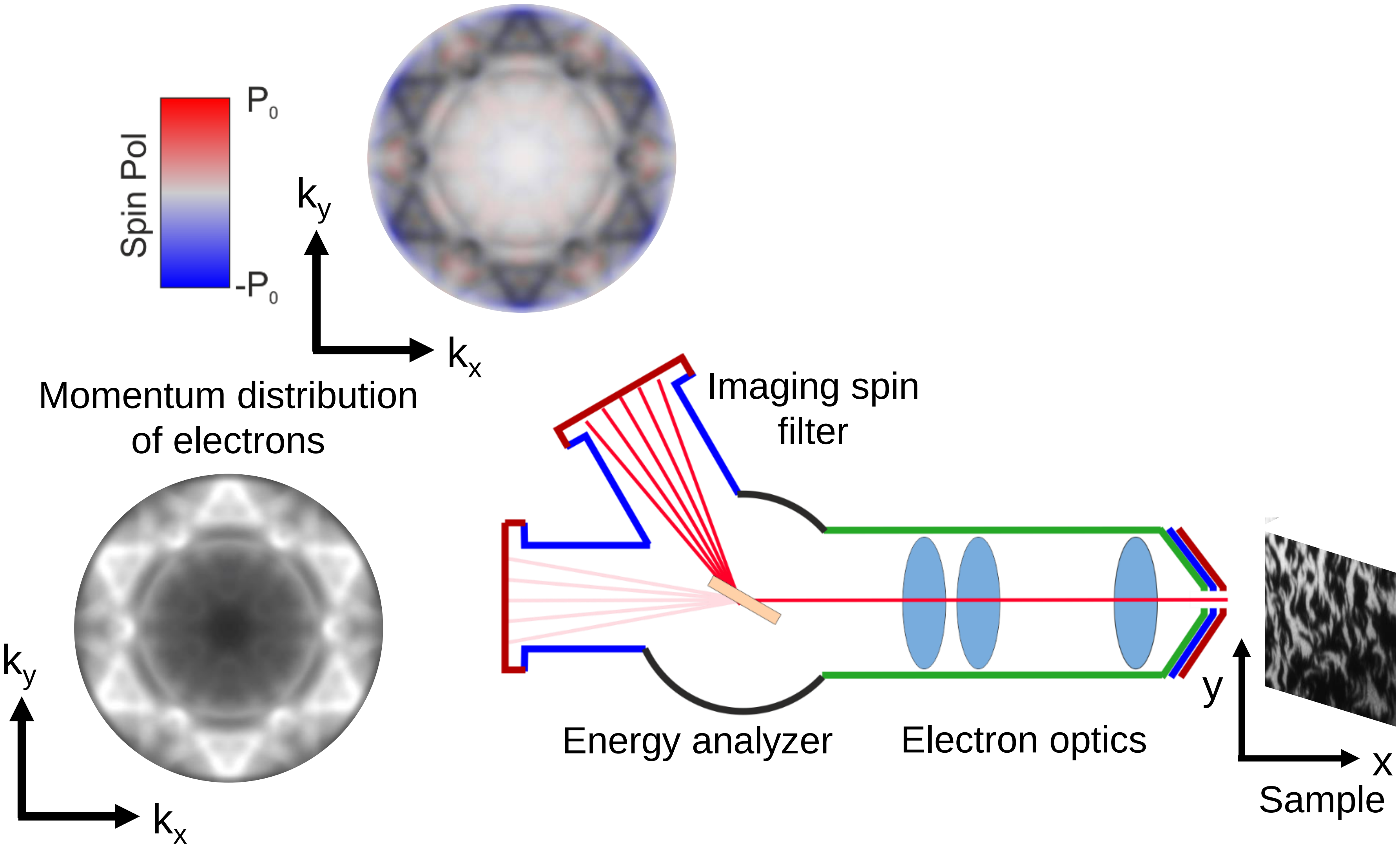
Momentum distribution of electrons





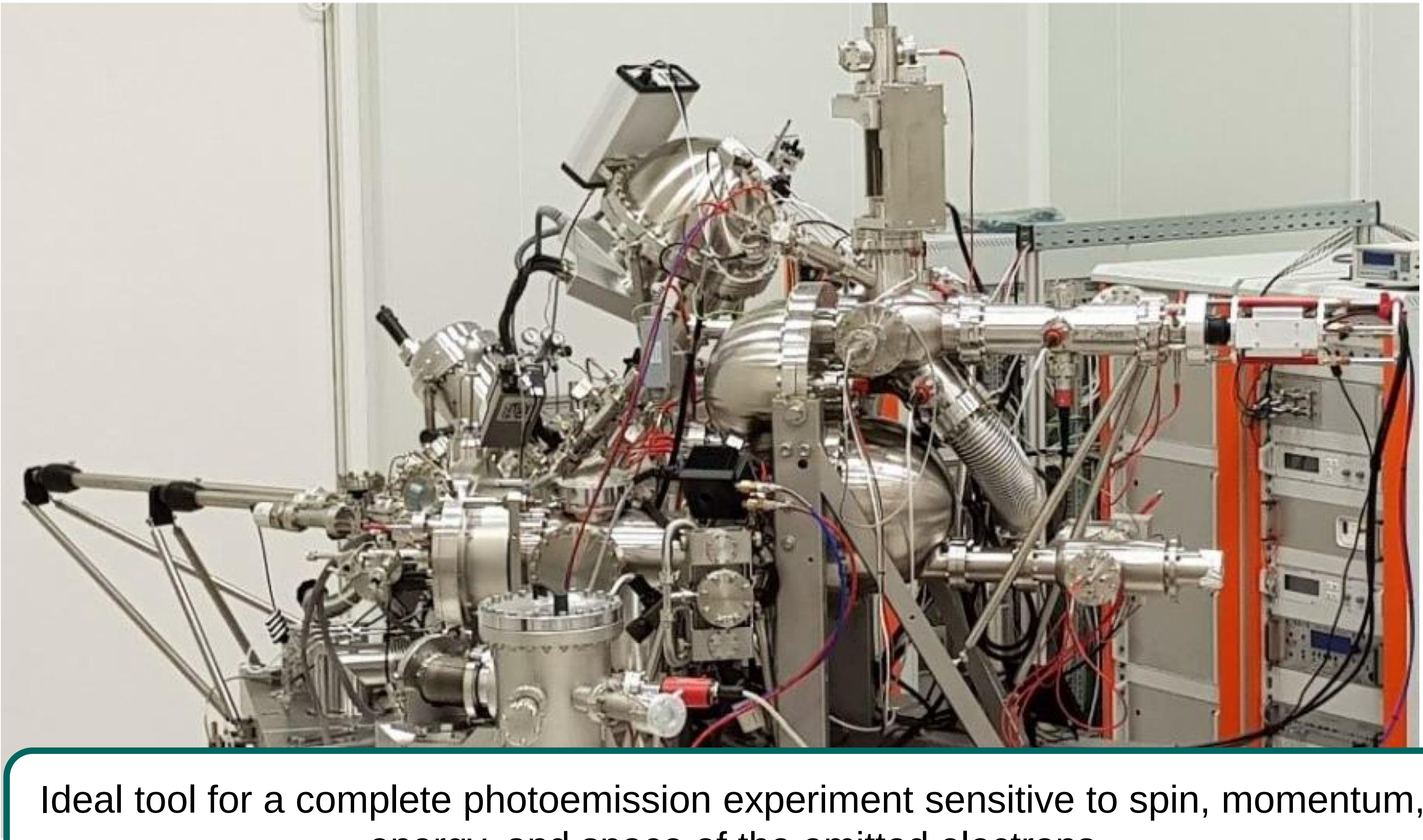
Photoemission and the Band Structure Approach

Momentum Microcopy



Photoemission and the Band Structure Approach

Spin-resolved momentum microscope “NanoEsca” @ ELI-Alps



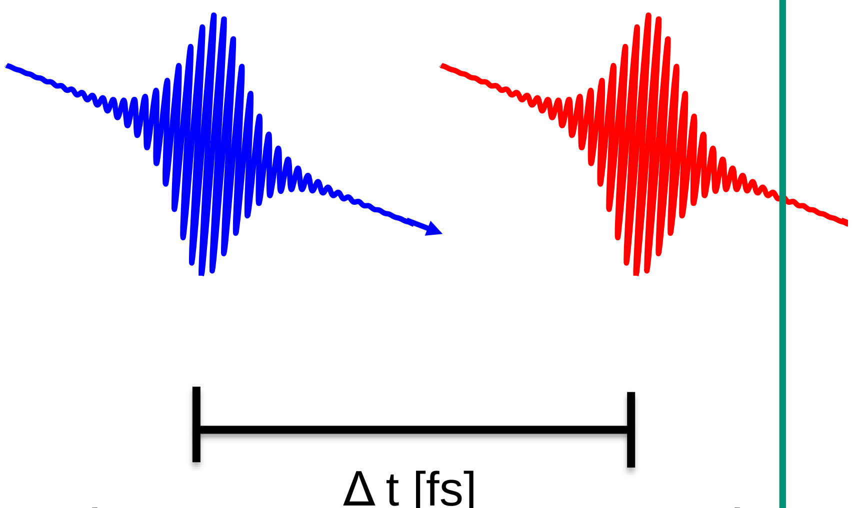
Ideal tool for a complete photoemission experiment sensitive to spin, momentum, energy, and space of the emitted electrons

Ultrafast Photoelectron Spectroscopy

Multidimensional (time, spin, momentum, energy) photoemission spectroscopy fs XUV light pulses

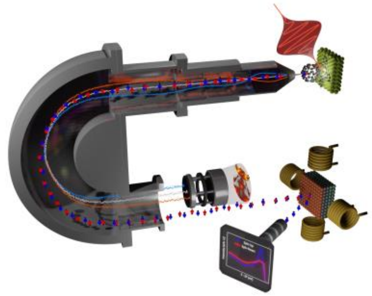
Probe Pulse
“Sensor”

Pump Pulse
“Trigger”

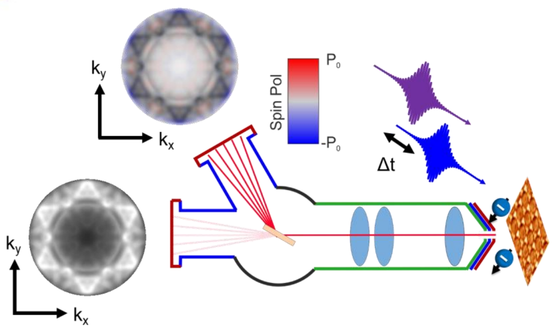


ultrafast clock

Spin-resolved ARPES



Momentum Microscopy



sensor for electron and spin system

The 2021 Ultrafast Spectroscopic Probes of Condensed Matter Roadmap – Chapter 8
J. Phys.: Condens. Matter 33 (2021) 353001

Ultrafast Photoelectron Spectroscopy

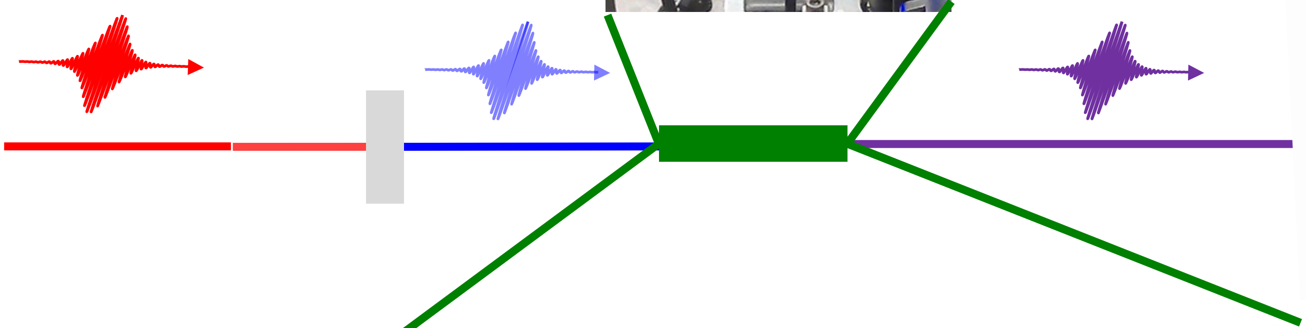
Multidimensional (time, spin, momentum, energy) photoemission spectroscopy fs XUV light pulses

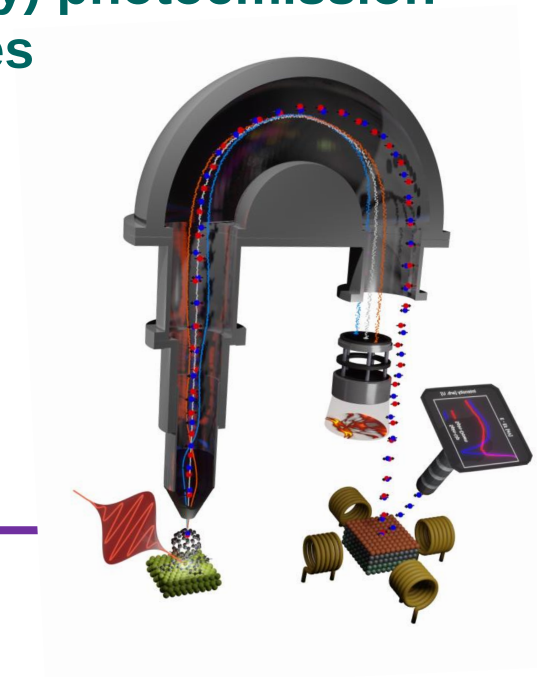
fs laser amplifier

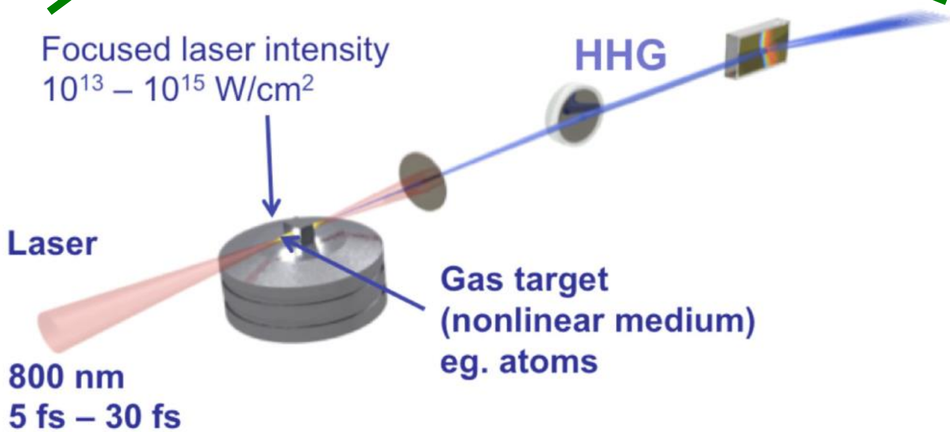
30 fs, 10kHz – 1MHz

Higher Harmonic Generation

30 fs, 20 eV – 200 eV

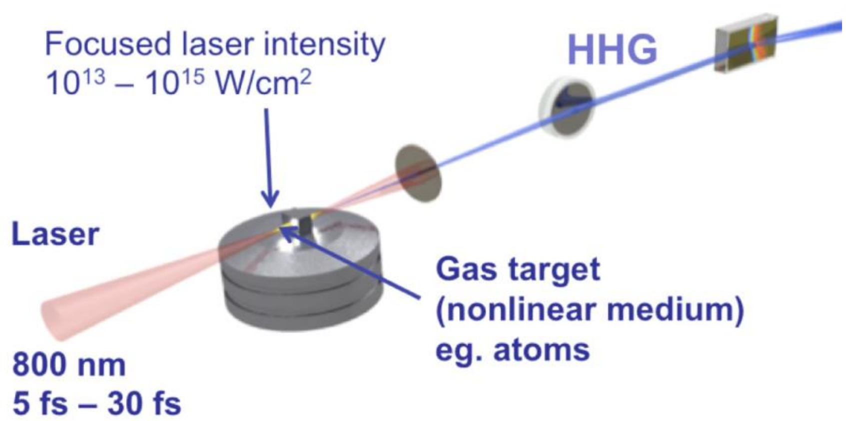




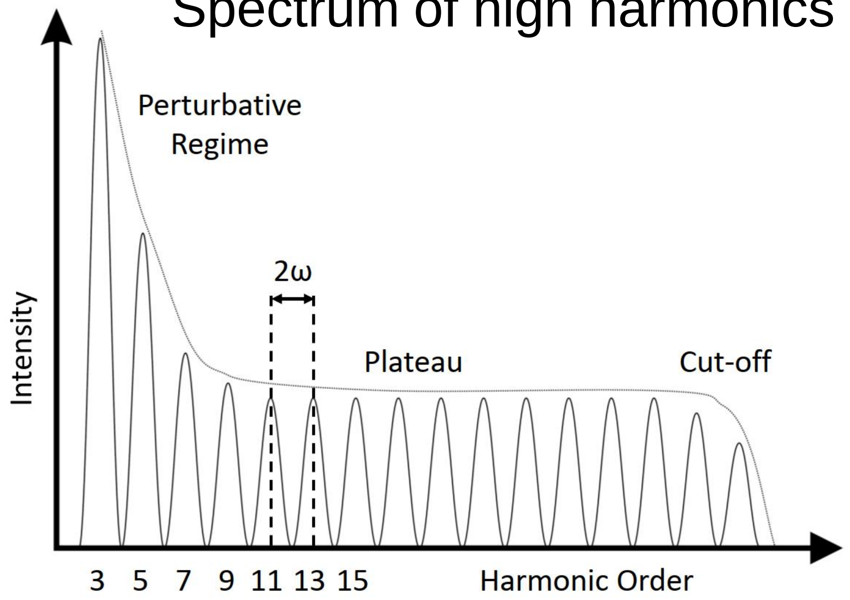


Ultrafast light source for extreme UV and soft X-rays

High harmonics generation



Spectrum of high harmonics



Photon energies:

$$\hbar\omega_{HHG} = (2n+1)\cdot\hbar\omega_0$$

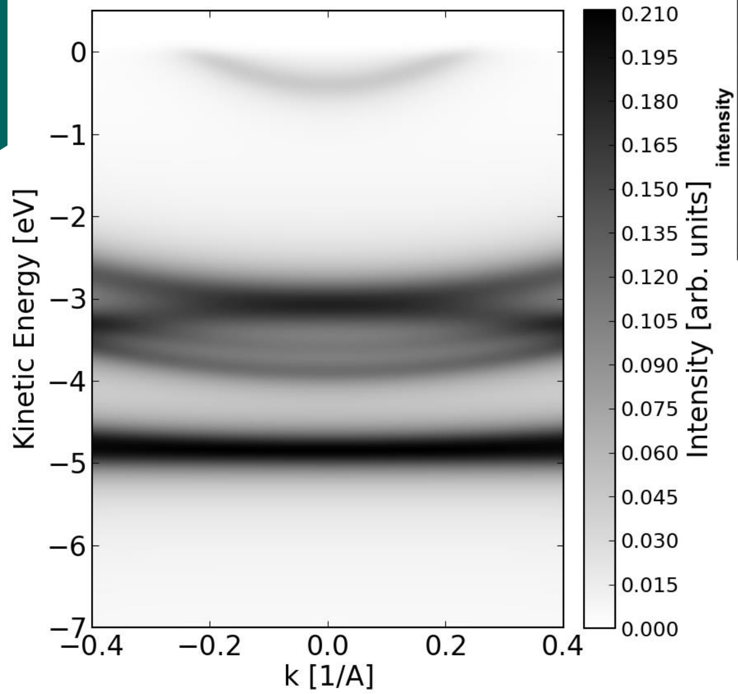
Cutoff energy

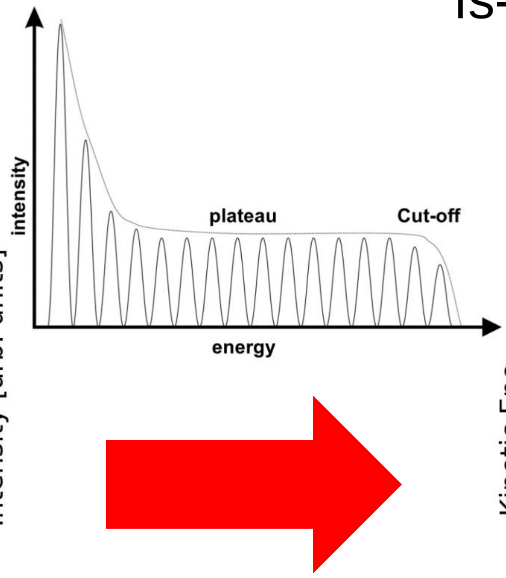
$$E_{photon,max} \propto I_{Laser}\cdot\lambda^2$$

Ultrafast light source for extreme UV and soft X-rays

High harmonics generation

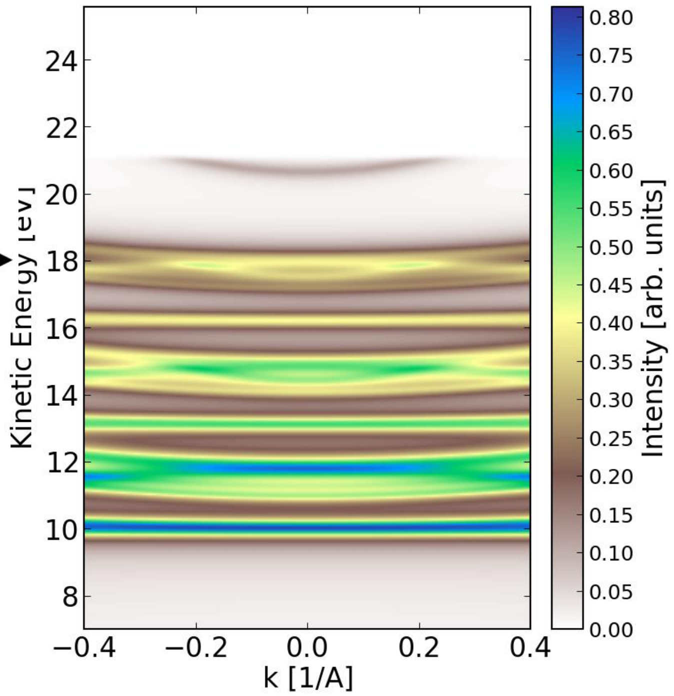
Monochromatic source





With all HHG lines

fs-XUV source based on HHG

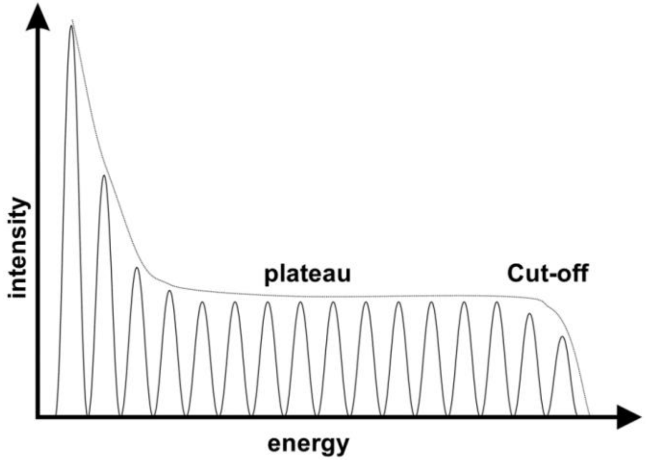


Superposition of several PES spectra generated by different photon energies

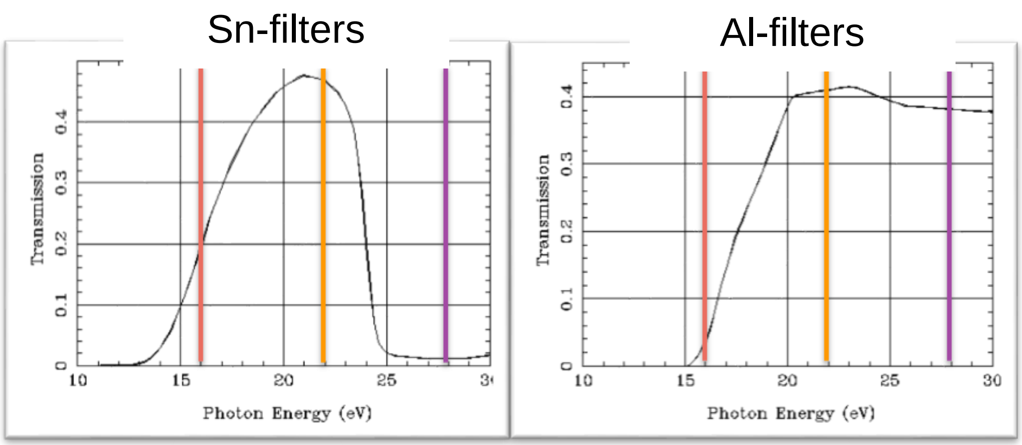
Ultrafast light source for extreme UV and soft X-rays

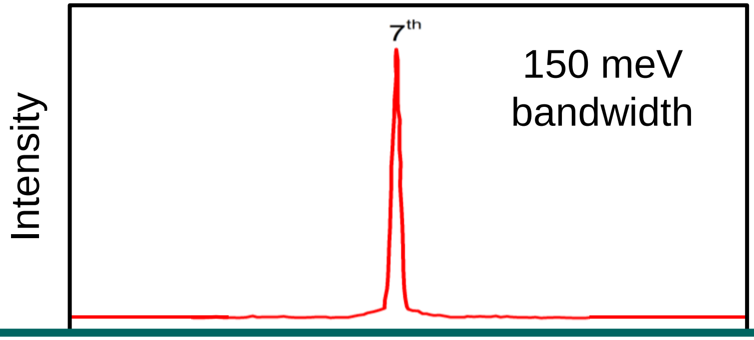
High harmonics generation

Polychromatic spectrum



Monochromatizing with pass filters

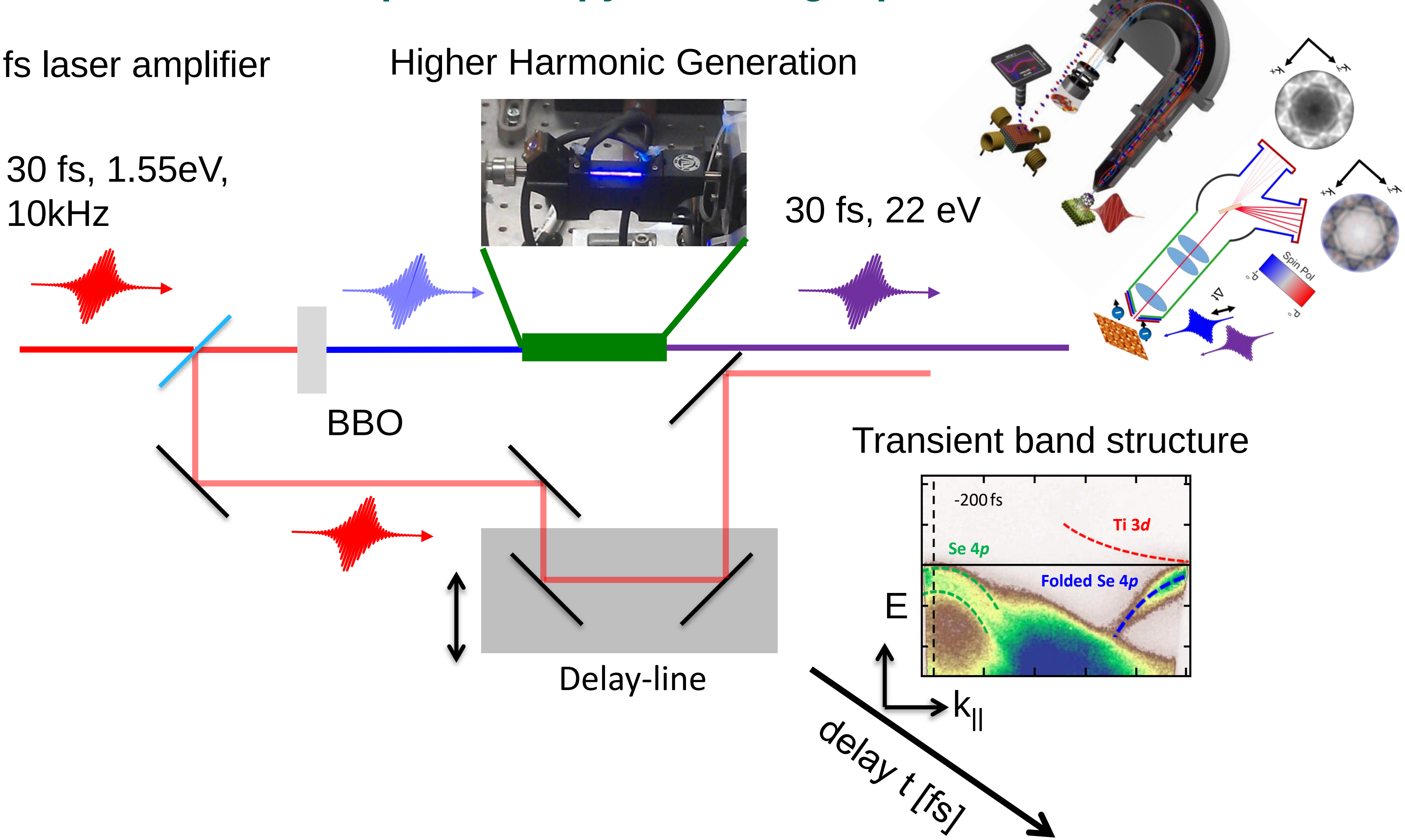




Quasi-monochromatic fs-XUV light source by monochromatization

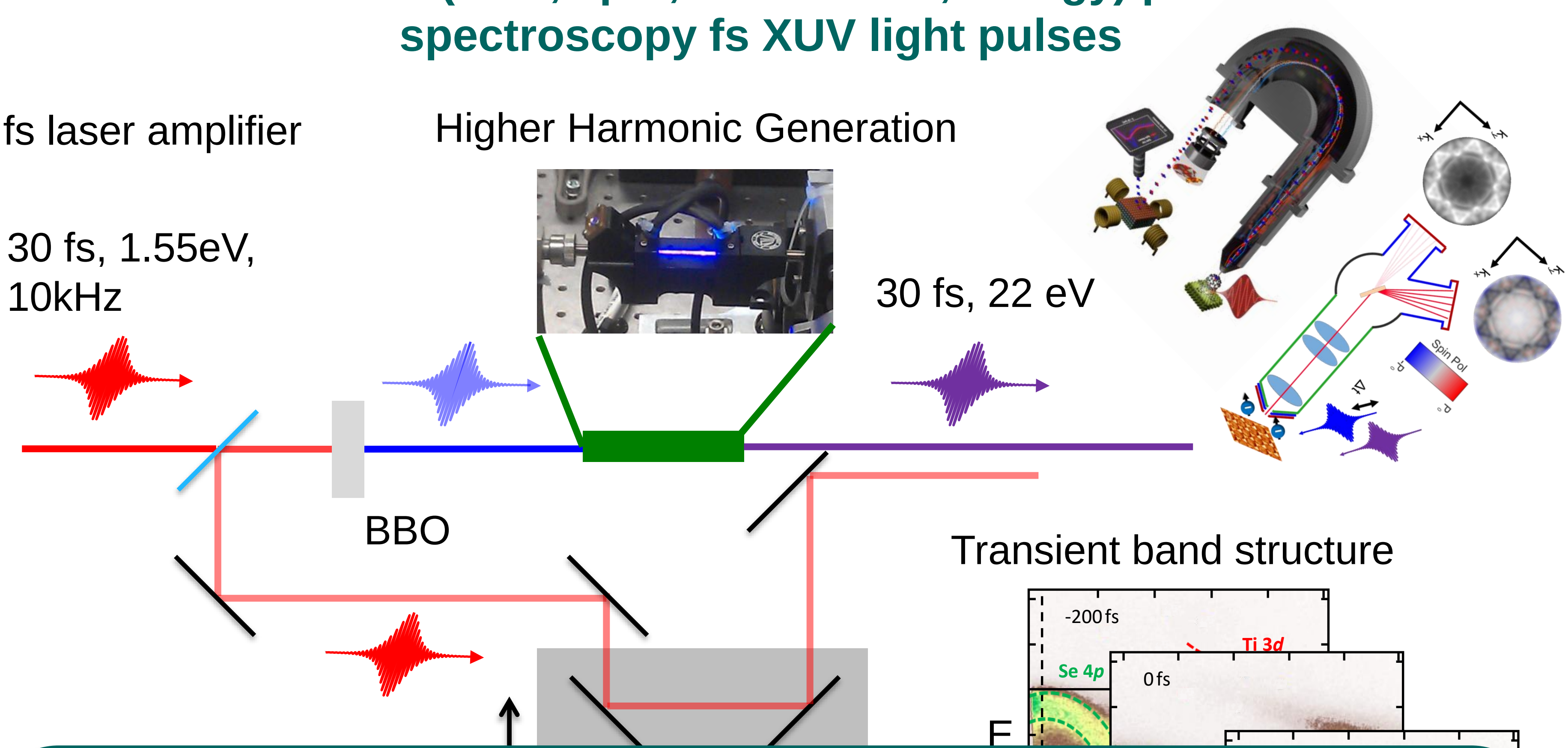
Ultrafast Photoelectron Spectroscopy

Multidimensional (time, spin, momentum, energy) photoemission spectroscopy fs XUV light pulses



Ultrafast Photoelectron Spectroscopy

Multidimensional (time, spin, momentum, energy) photoemission spectroscopy fs XUV light pulses



Complete photoemission experiment for ultrafast surface science:

1. Dynamics of charge and spin carrier in energy and momentum space
2. Transient renormalization of spin-dependent band structure

Challenges of Ultrafast Photoelectron Spectroscopy

Challenges of Ultrafast Photoelectron Spectroscopy

Photoelectron spectroscopy

Photon flux

Photons/s

Energy resolution

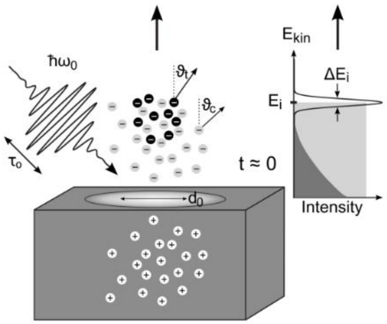
- Monochromatized photon source
- Small spectral bandwidth of source
- High photon flux

Challenges of Ultrafast Photoelectron Spectroscopy

Photoelectron spectroscopy | **Ultrafast Science**

Photon flux
Photons/s

Space charge effects



Energy resolution

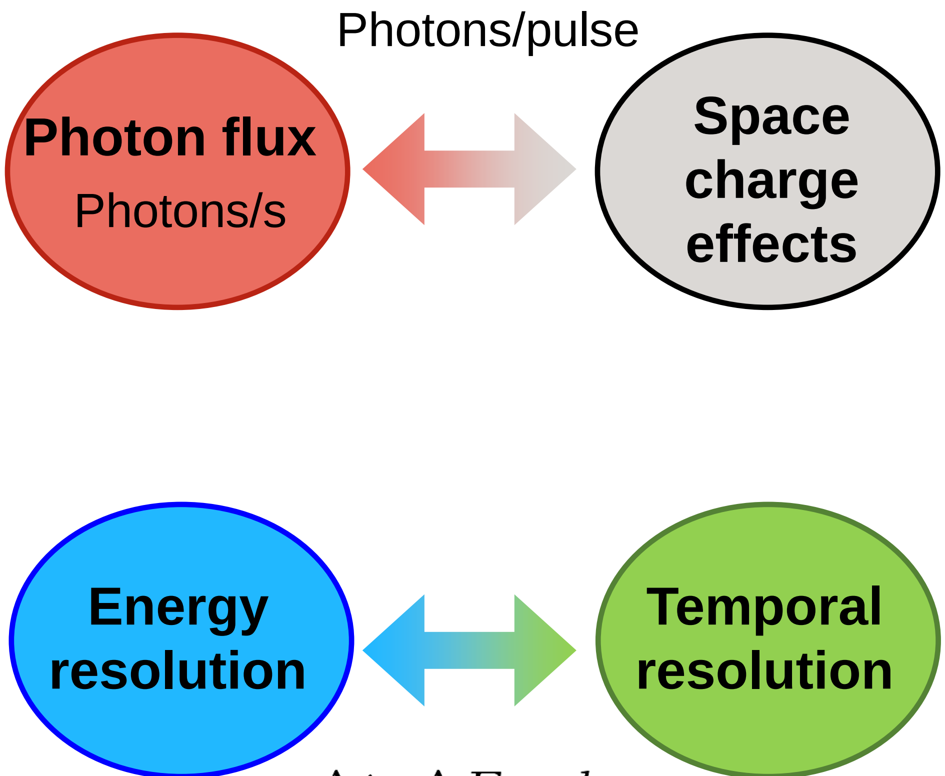
Temporal resolution

- Monochromatized photon source
- Small spectral bandwidth of source
- High photon flux

- High photon energies
- Attosecond pulse trains
- High Intensity per pulse

Challenges of Ultrafast Photoelectron Spectroscopy

Photoelectron spectroscopy | **Ultrafast Science**



$$\Delta t \cdot \Delta E \approx h$$

- Monochromatized photon source
- Small spectral bandwidth of source
- High photon flux

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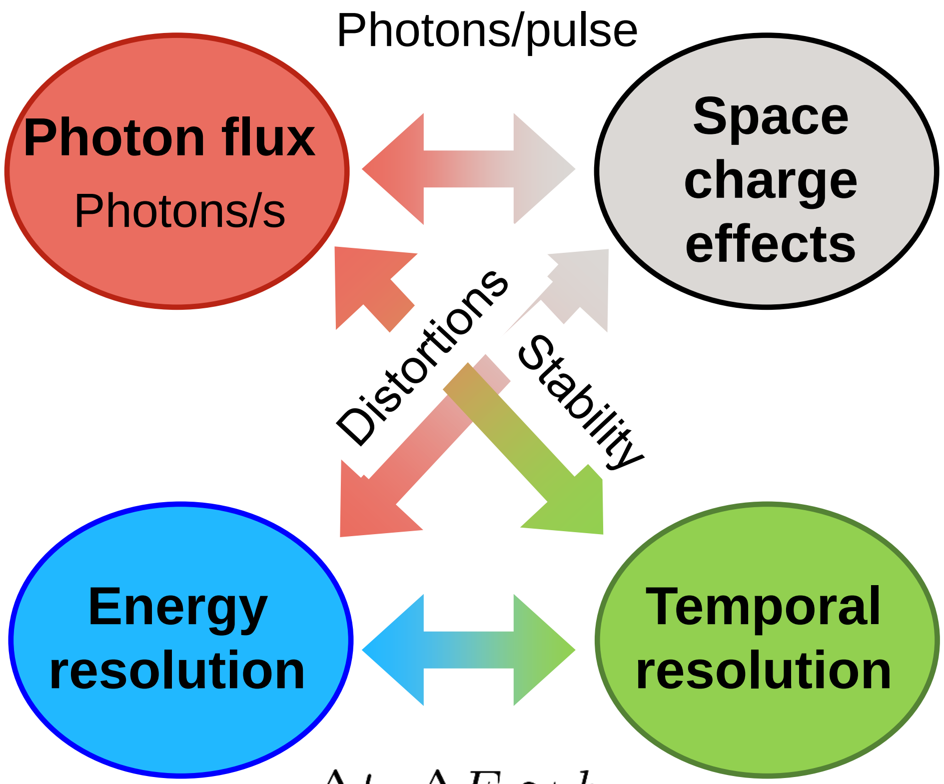
Challenges of Ultrafast Photoelectron Spectroscopy

Photoelectron spectroscopy | **Ultrafast Science**



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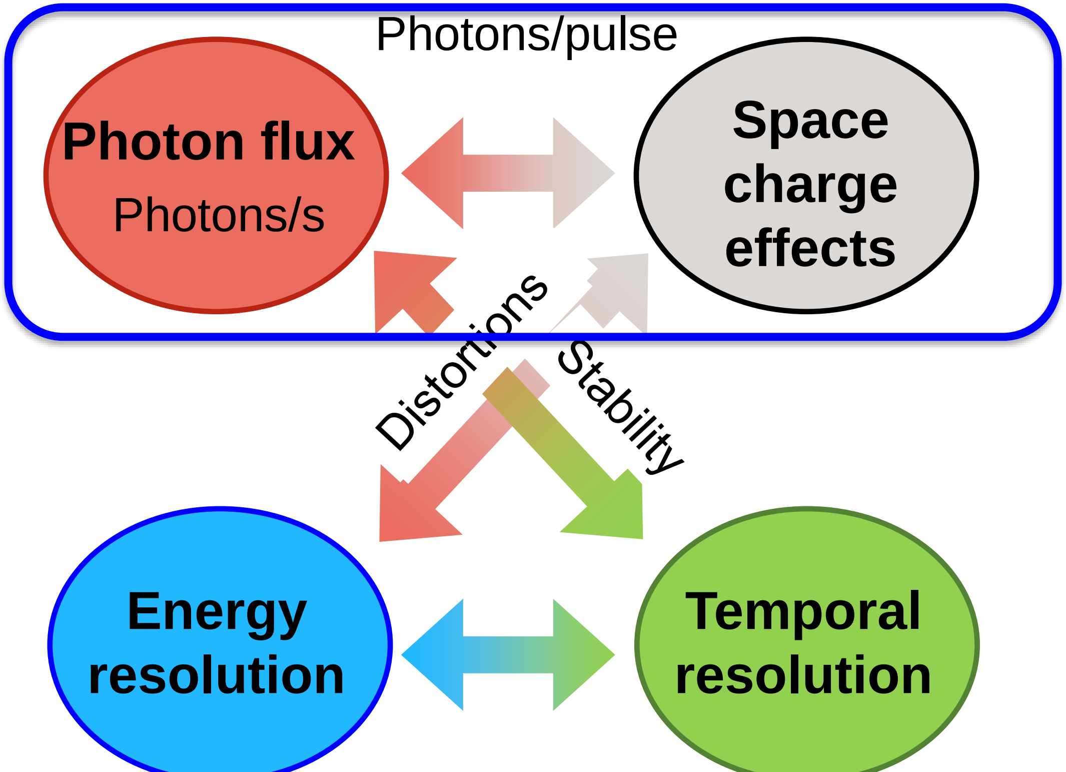
Challenges of Ultrafast Photoelectron Spectroscopy

Photoelectron spectroscopy | **Ultrafast Science**



$$\Delta t \cdot \Delta E \approx h$$

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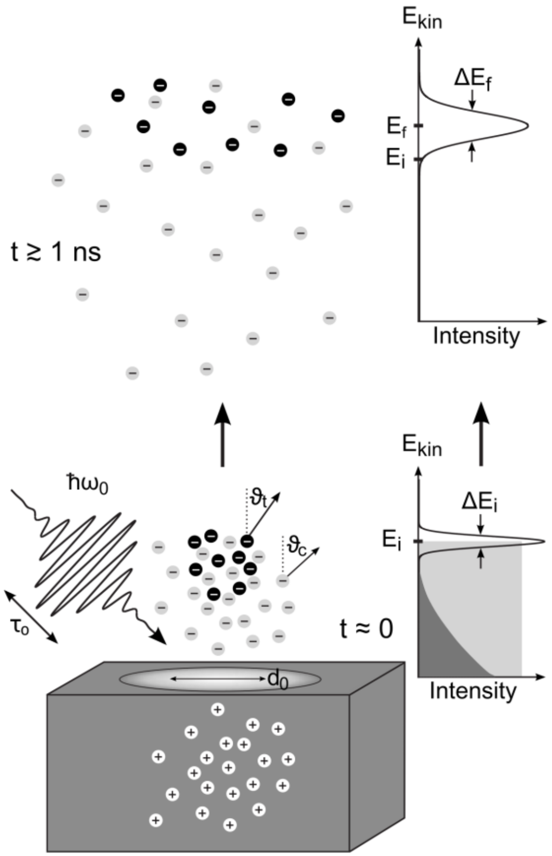
- Small spectral bandwidth of source
- High photon flux

- High photon energies
- Attosecond pulse trains
- High Intensity per pulse

Challenges of Ultrafast Photoelectron Spectroscopy

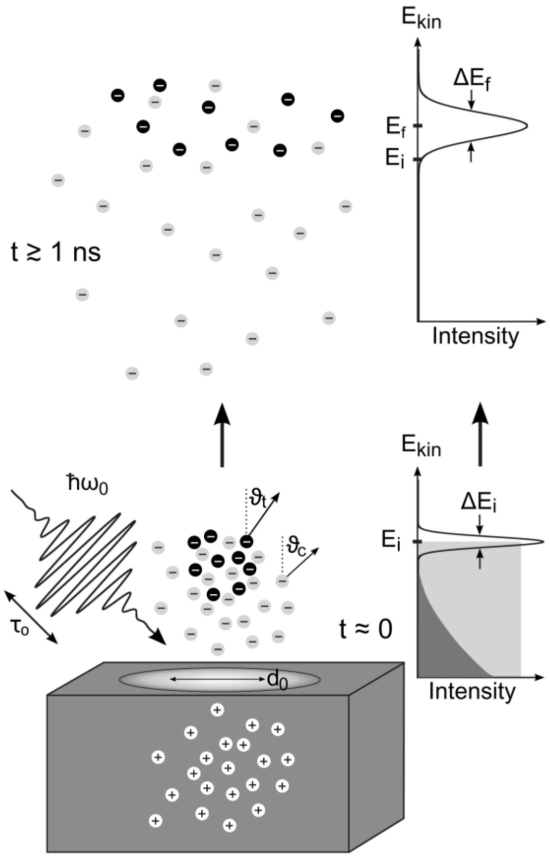
Space Charge Effects



Journ. Appl. Phys. **100**, 024912 (2006)

Challenges of Ultrafast Photoelectron Spectroscopy

Space Charge Effects

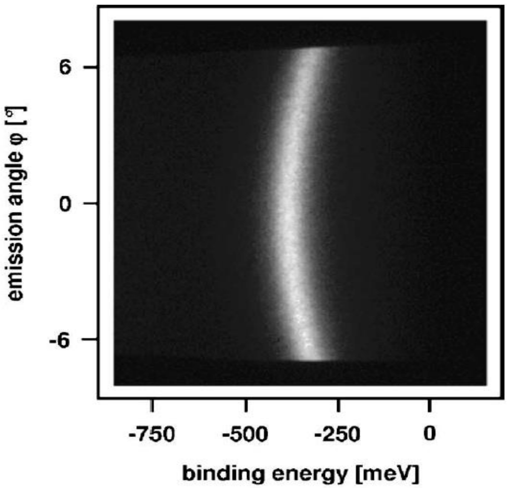


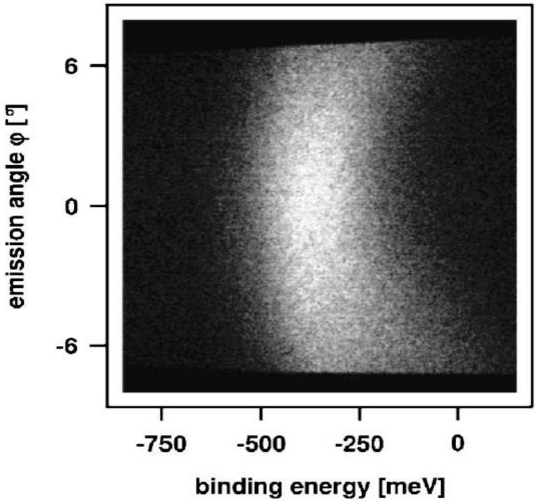
Journ. Appl. Phys. **100**, 024912 (2006)

Vacuum space charge in ARPES:
Example: Surface State of Cu(111) surface

1 electron/pulse

$>10^5$ electrons/pulse



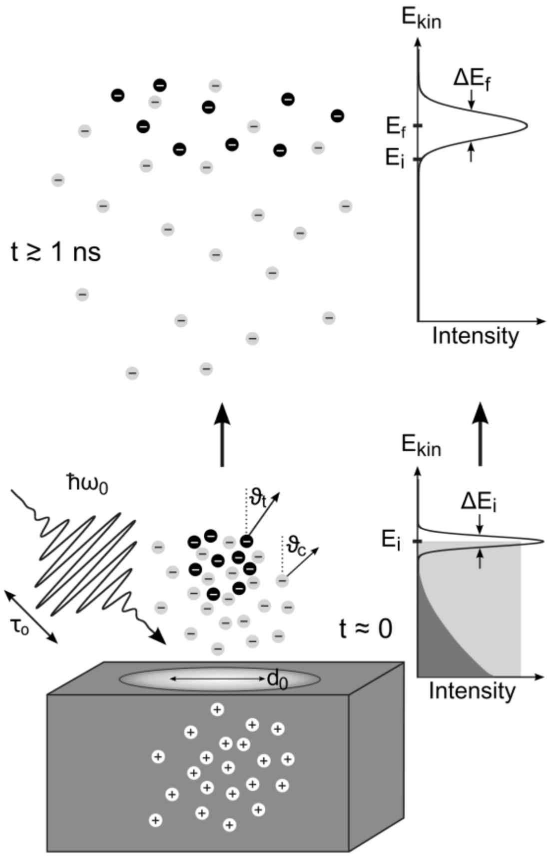


Space charge can lead to:

- Broadening in energy and momentum
- Energy shifts
- Total loss of information in ARPES data

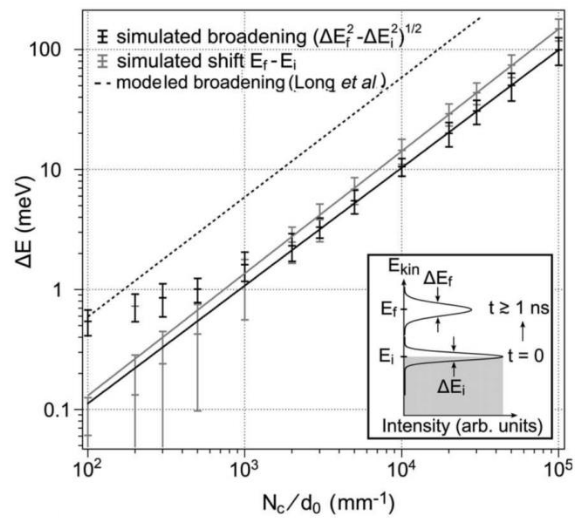
Challenges of Ultrafast Photoelectron Spectroscopy

Space Charge Effects

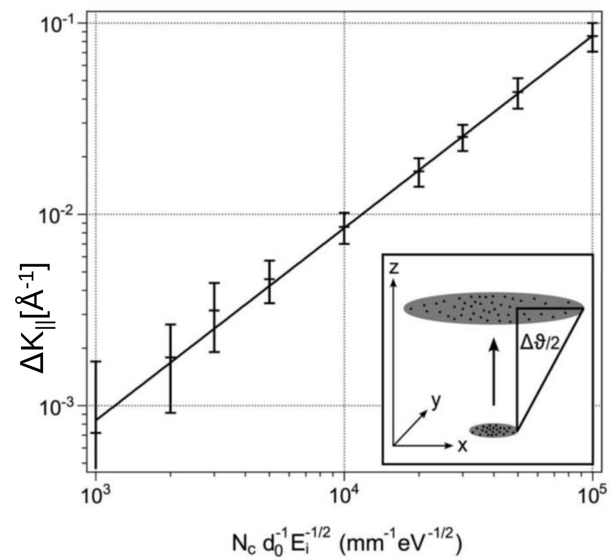


Vacuum space charge in ARPES:

Energy broadening



Momentum broadening

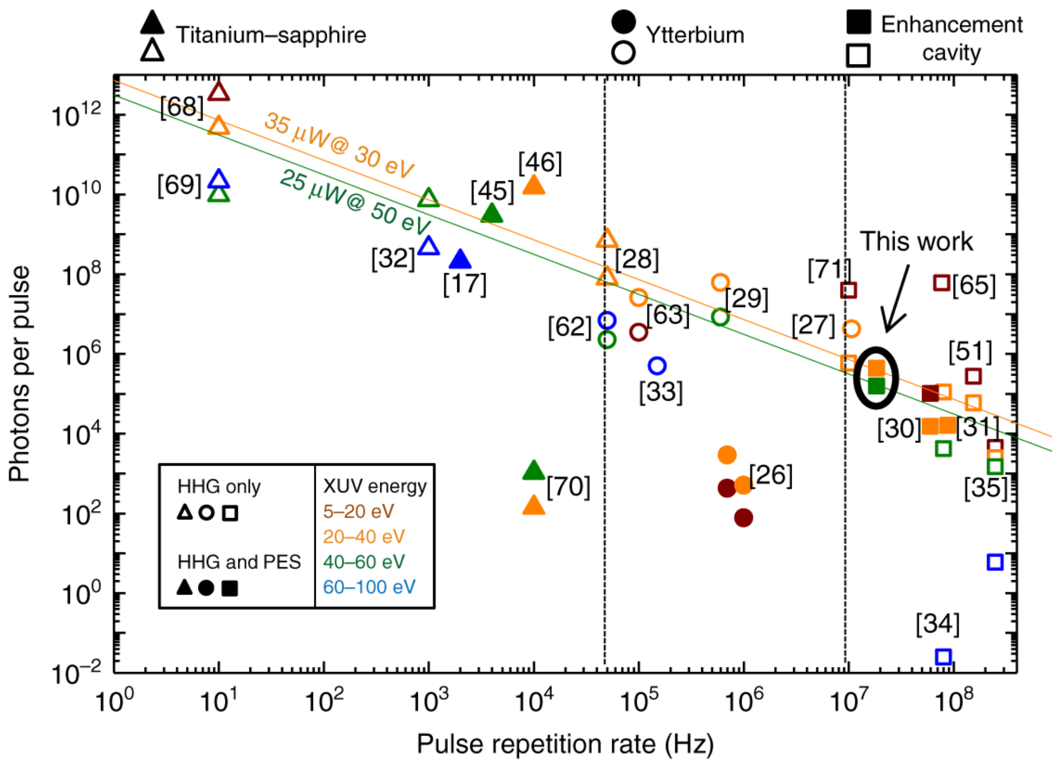


Journ. Appl. Phys. **100**, 024912 (2006)

Challenges of Ultrafast Photoelectron Spectroscopy

Repetition rate of fs-XUV sources

Photon flux and photons/pulse

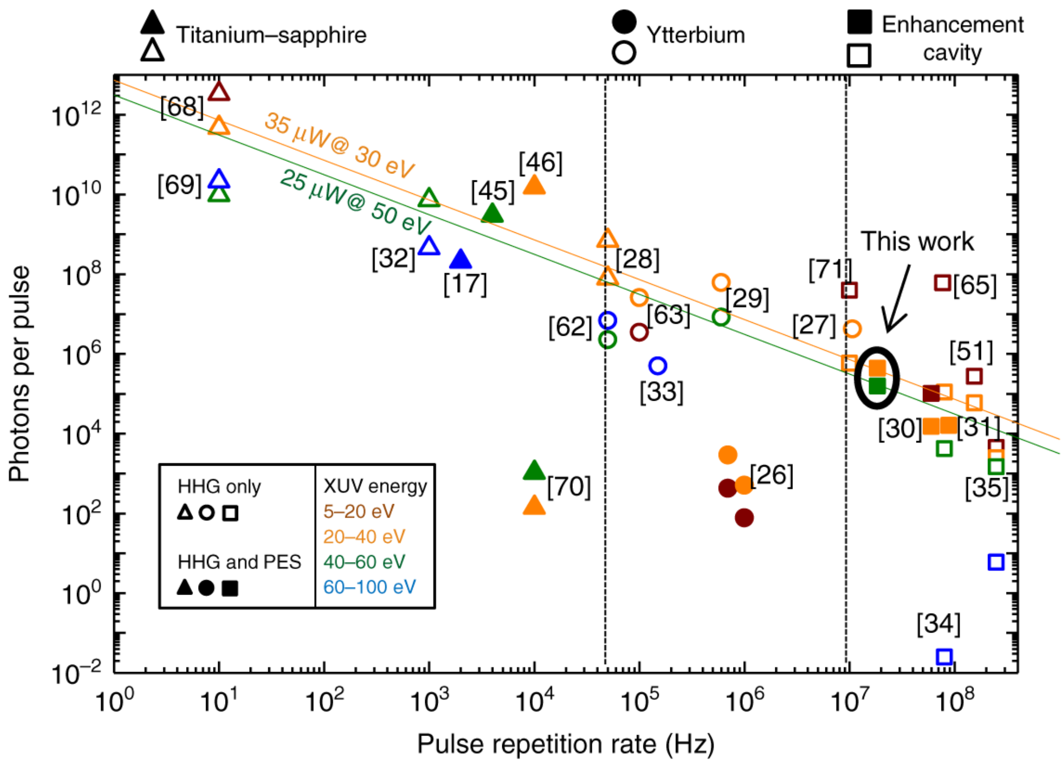


T. Saule et al. *Nat Commun* **10**, 458 (2019)

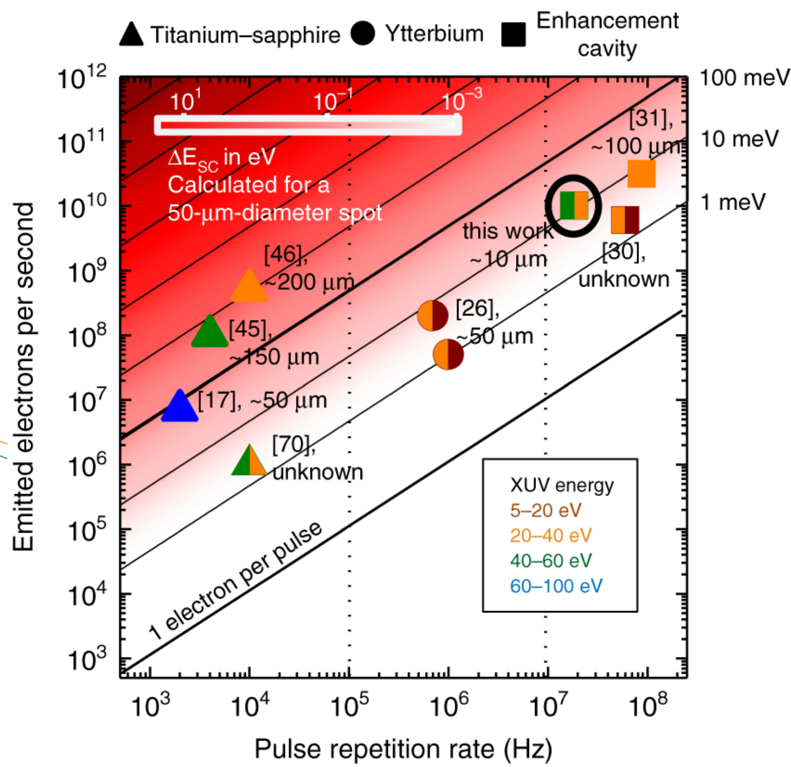
Challenges of Ultrafast Photoelectron Spectroscopy

Repetition rate of fs-XUV sources

Photon flux and photons/pulse



Photoelectrons and space charge



High repetition rate is essential for minor space charge effects and high data quality

T. Saule et al. *Nat Commun* **10**, 458 (2019)

Challenges of Ultrafast Photoelectron Spectroscopy

User facilities for ultrafast spectroscopy with fs/as time resolution

ELI-ALPS
Szeged, Hungary:
EU research facility

Surface science end station

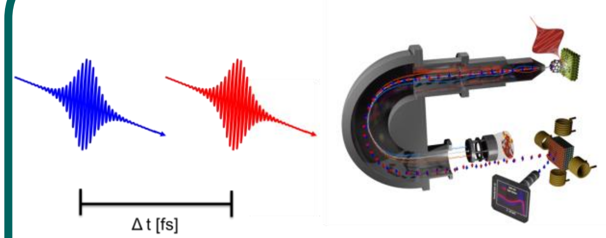
100kHz IR fiber-based, 3fs, 5mJ

ARTEMIS
Rutherford Appleton Lab, UK

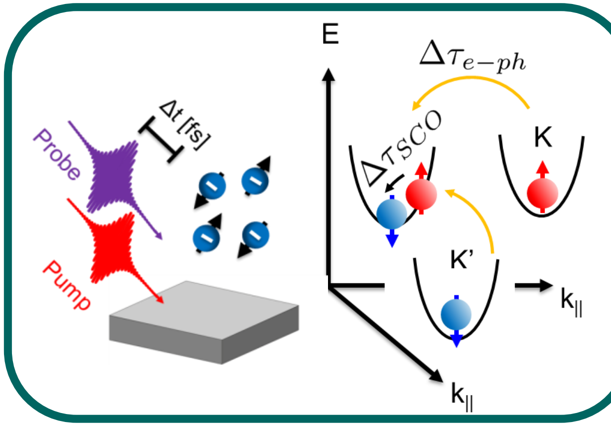
100 kHz IR OPCPA system

sub 10-fs XUV source (15 - 120 eV) for ultrafast time-resolved surface science studies

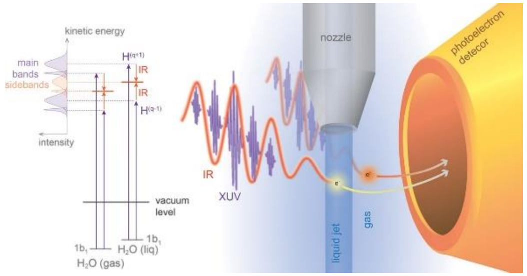
Outline



Ultrafast science in solids
… electrons in solids
… time-resolved photoemission spectroscopy



Monitoring….
…. the population dynamics of charge and spin carriers in momentum space
… the nature and spatial distribution of charge carriers in direct space
… interlayer charge separation across interfaces



Perspectives and challenges of attosecond surface science

The Team

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S. Mathias, M. Jansen,
W. Bennecke

GEORG-AUGUST-UNIVERSITÄT
GÖTTINGEN

eli

Elettra NanoEsca team
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University of Graz
P. Puschnig

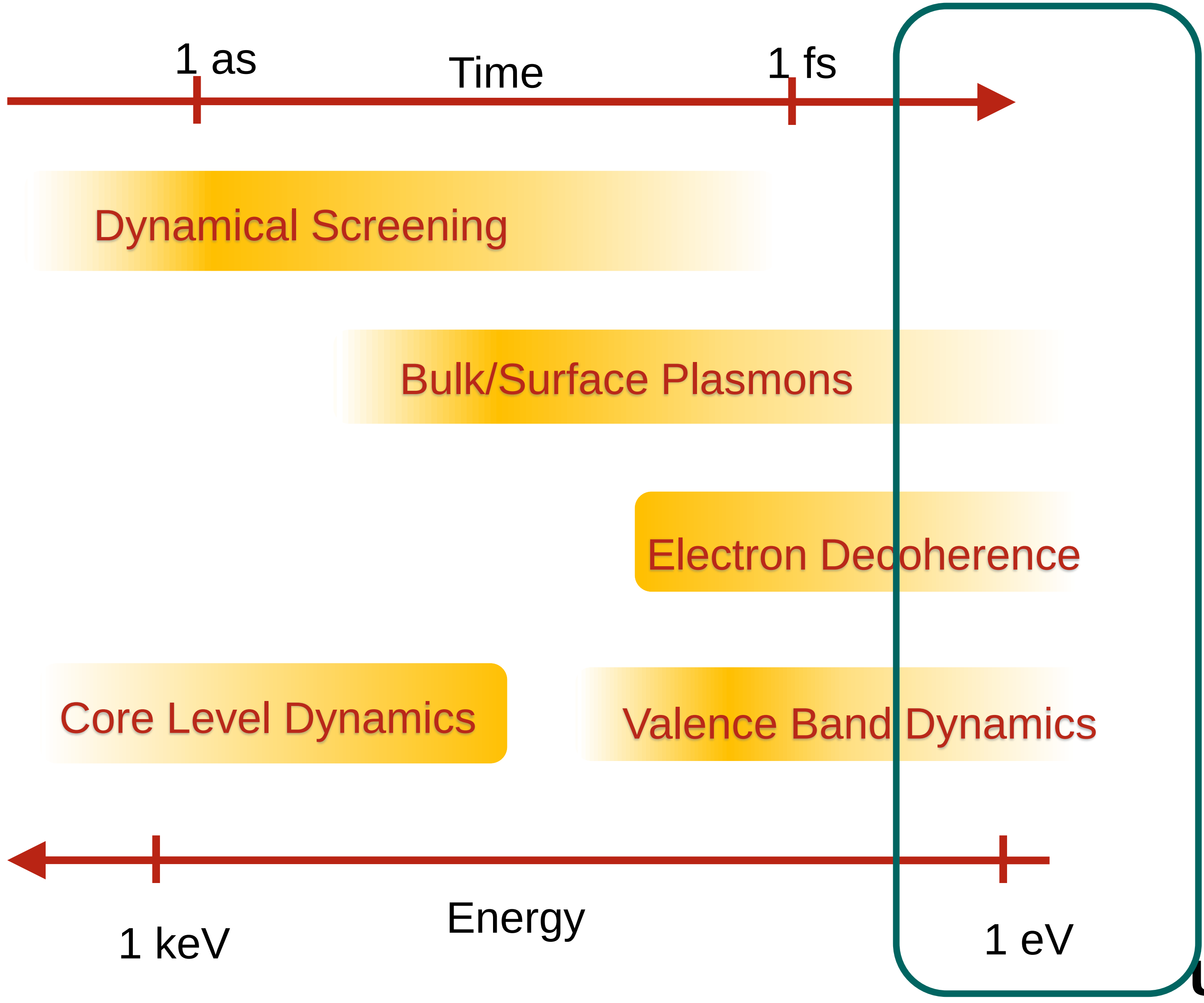
UNI GRAZ

LMU München
J. Braun, H. Ebert

LMU LUDWIG-MAXIMILIANS-UNIVERSITÄT MÜNCHEN

Elettra Sincrotrone Trieste

The characteristic timescales of (condensed) matter

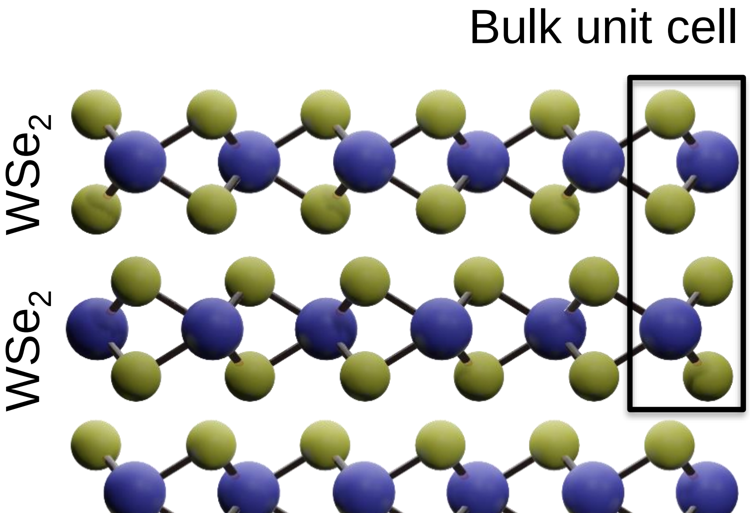


Population Dynamics in 2D Semiconductors

Spin-valley-layer locked valence band structure

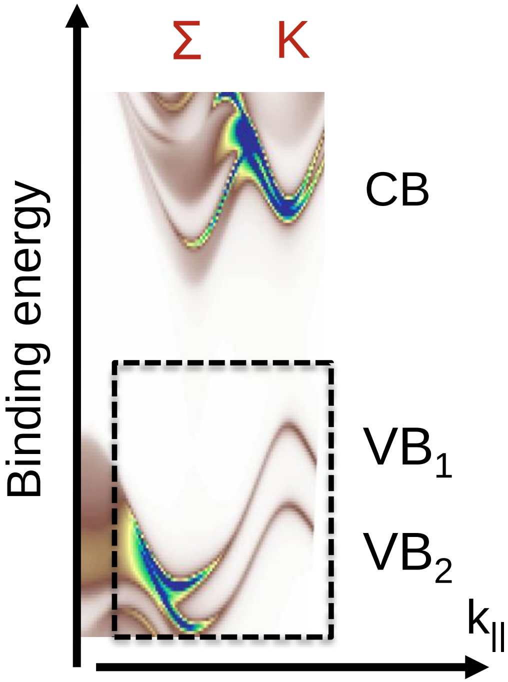
Hidden spin polarization

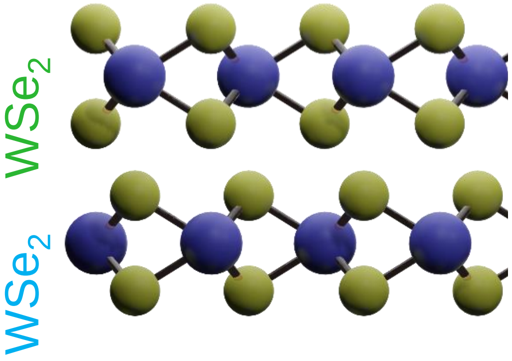
X. Zhang et al.
Nature Phys 10, 387–393 (2014)
Riley et al. Nat. Phys. 10 (2014) 835



Population Dynamics in 2D Semiconductors

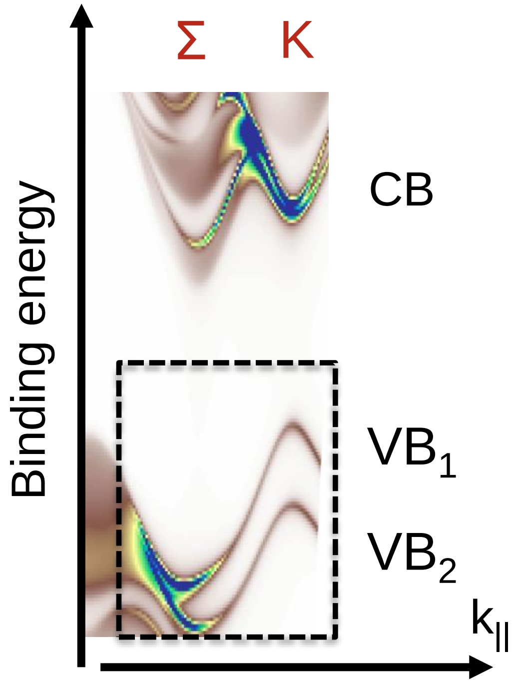
Spin-dependent **valence** band structure

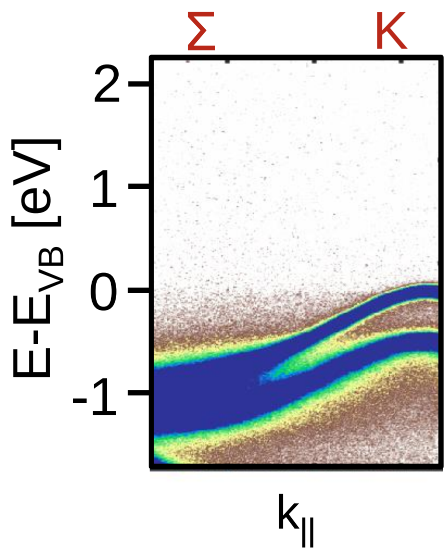


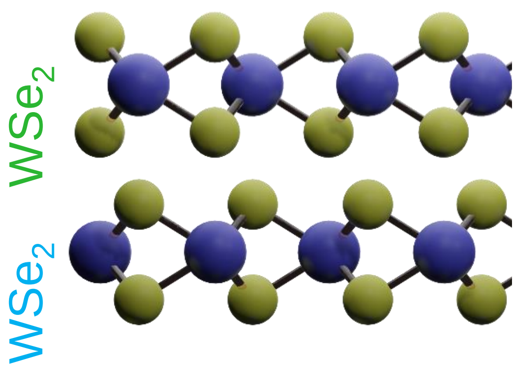


Population Dynamics in 2D Semiconductors

Spin-dependent **valence** band structure







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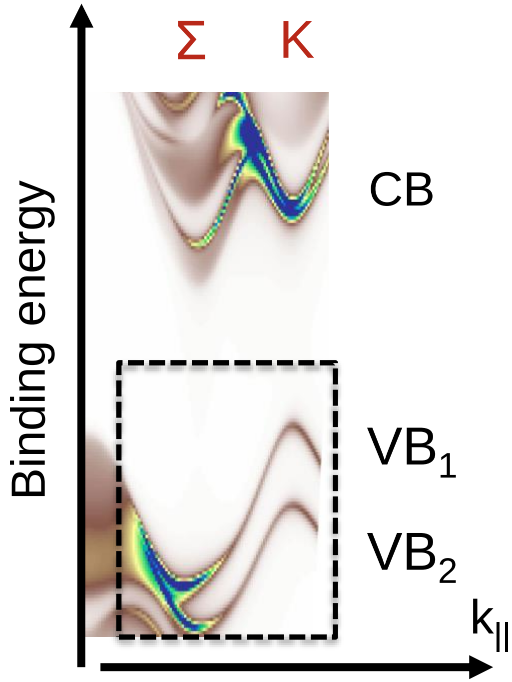
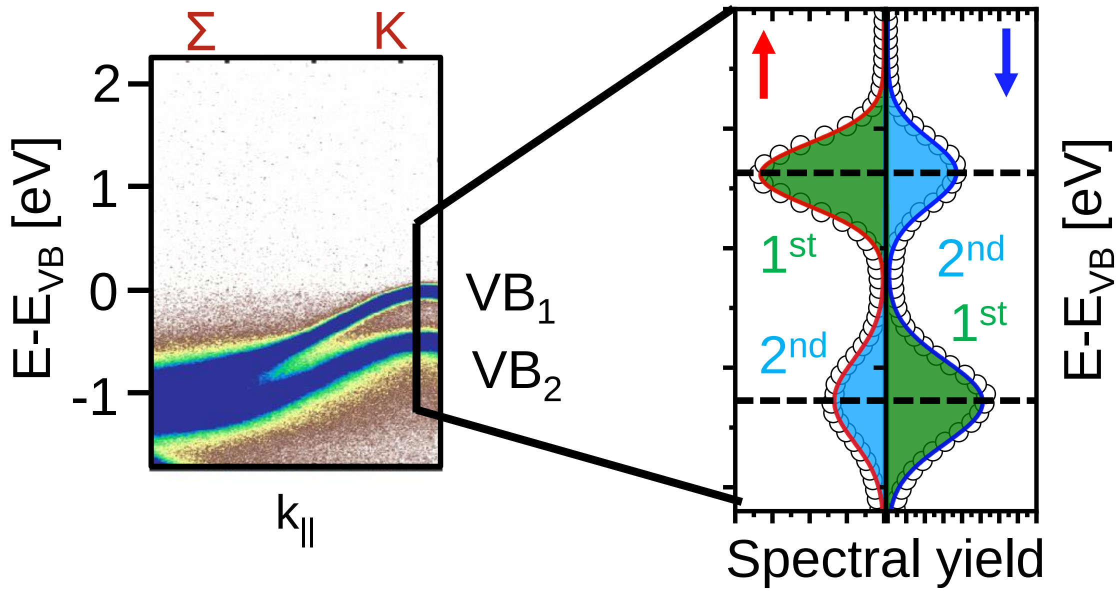
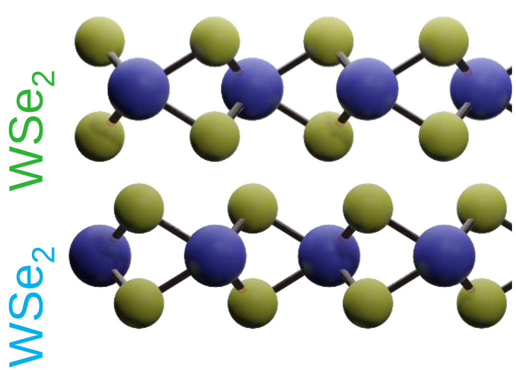
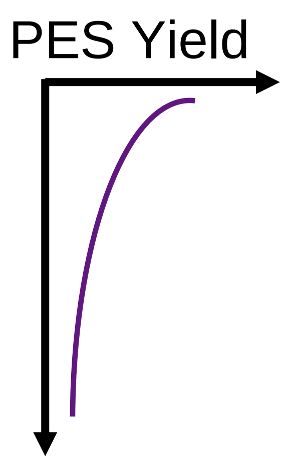
Spin-dependent **valence** band structure

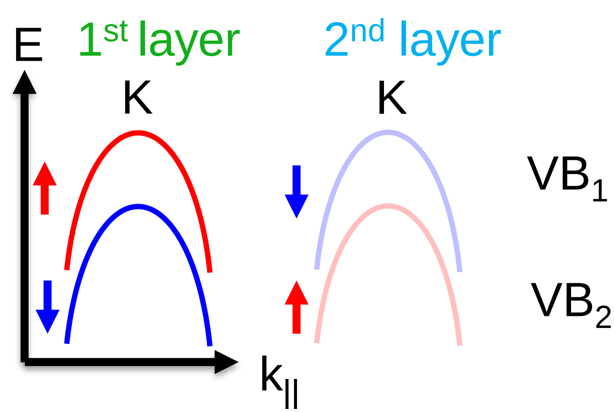






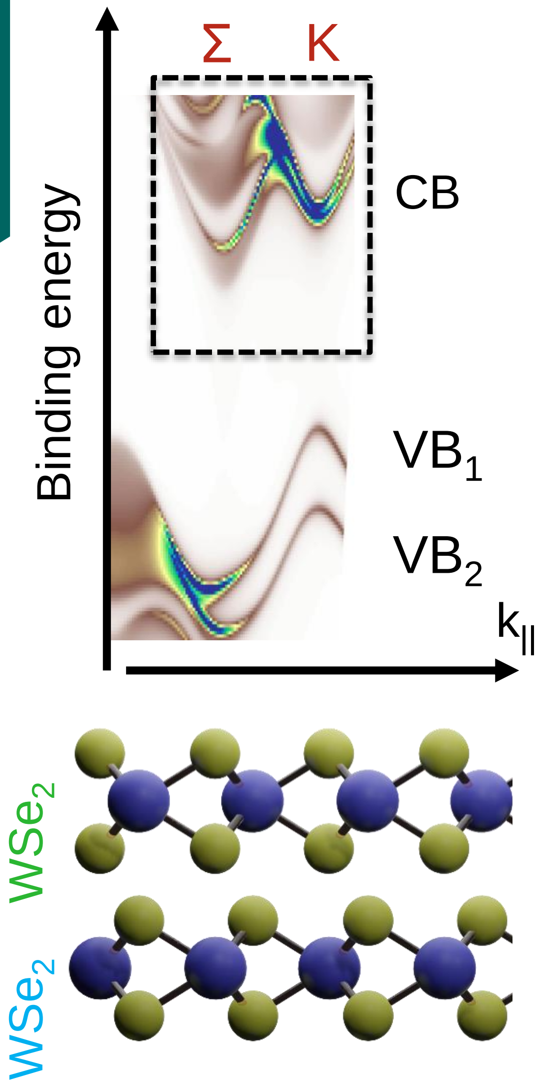
Nat. Phys. 10 (2014) 835





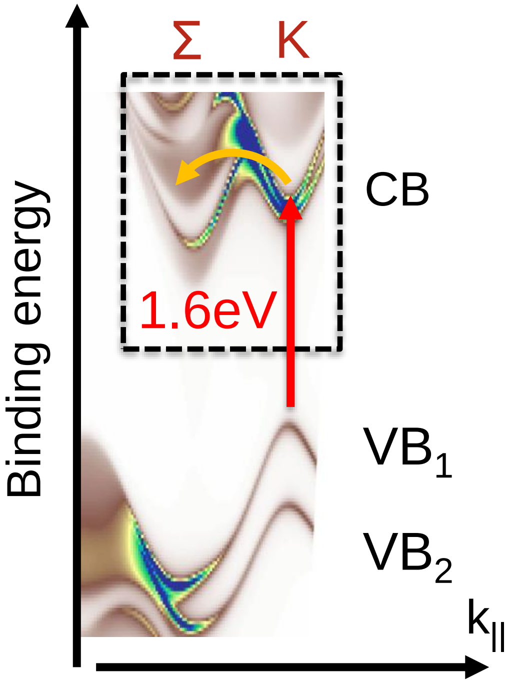
Population Dynamics in 2D Semiconductors

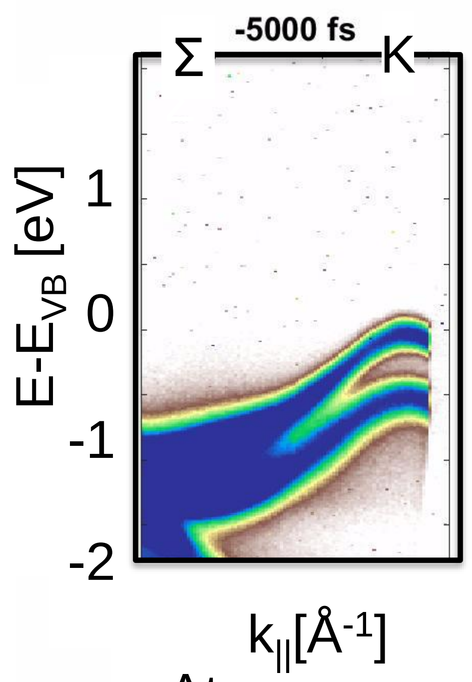
Spin-dependent **unoccupied** band structure



Population Dynamics in 2D Semiconductors

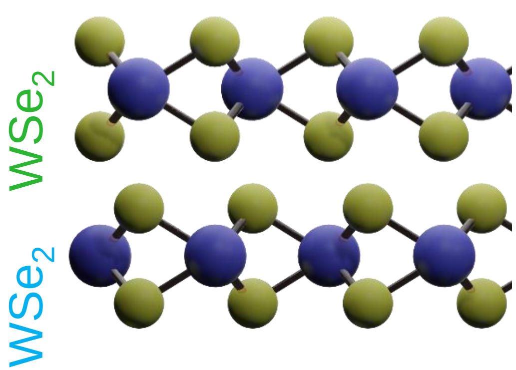
Spin-dependent **excited states** band structure

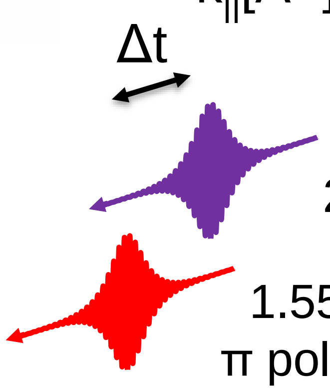




Conduction band

Valence band

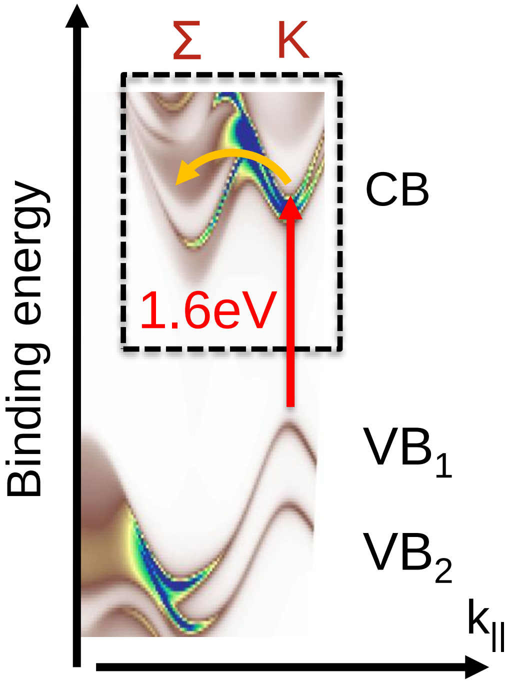


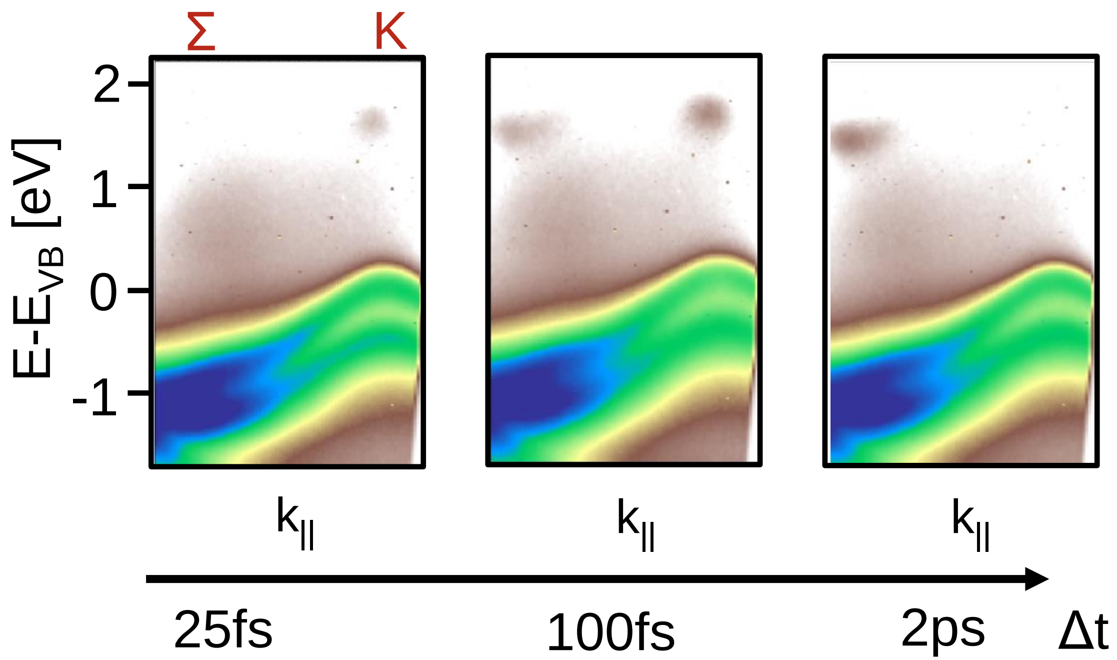


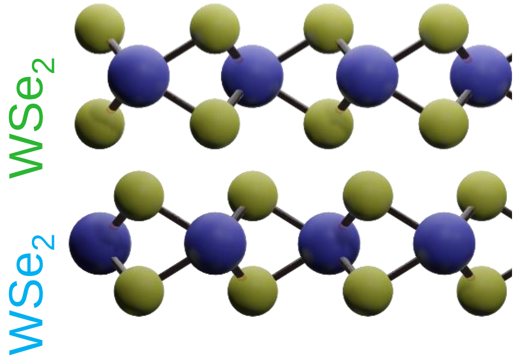
Bertoni *et al. Phys. Rev. Lett.* **117, 277201** (2016)

Ultrafast Spin Carrier Dynamics of WSe_2

Spin-dependent **excited states** band structure



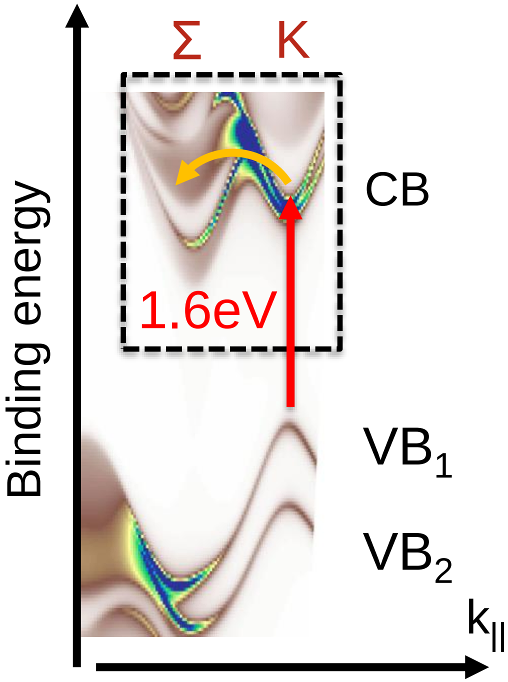


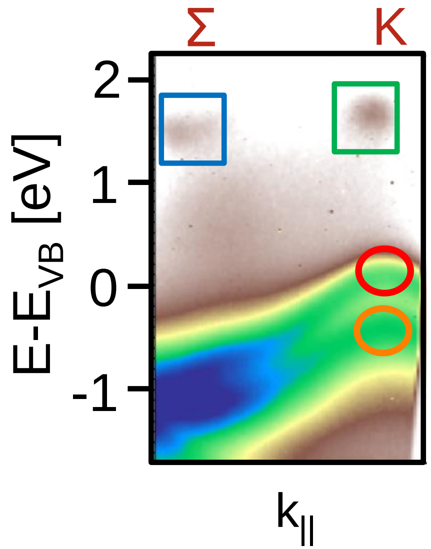


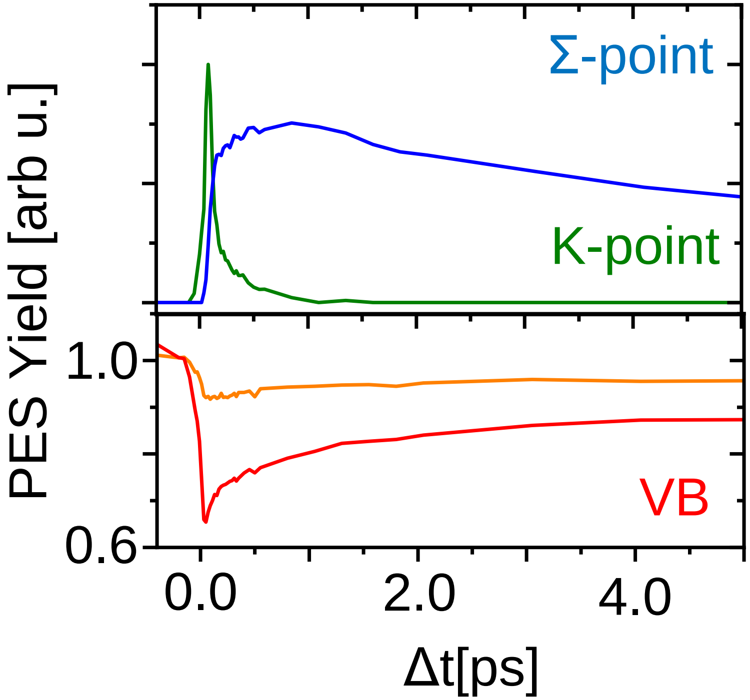
- Capture a movie of carrier dynamics in real time

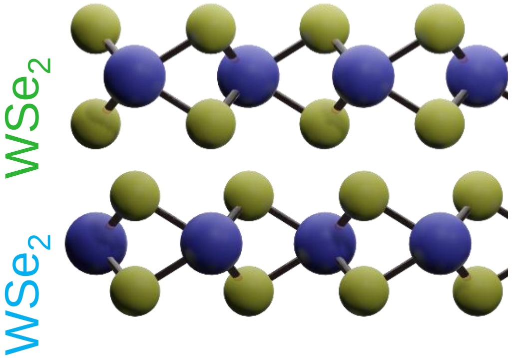
Ultrafast Spin Carrier Dynamics of WSe_2

Spin-dependent **excited states** band structure





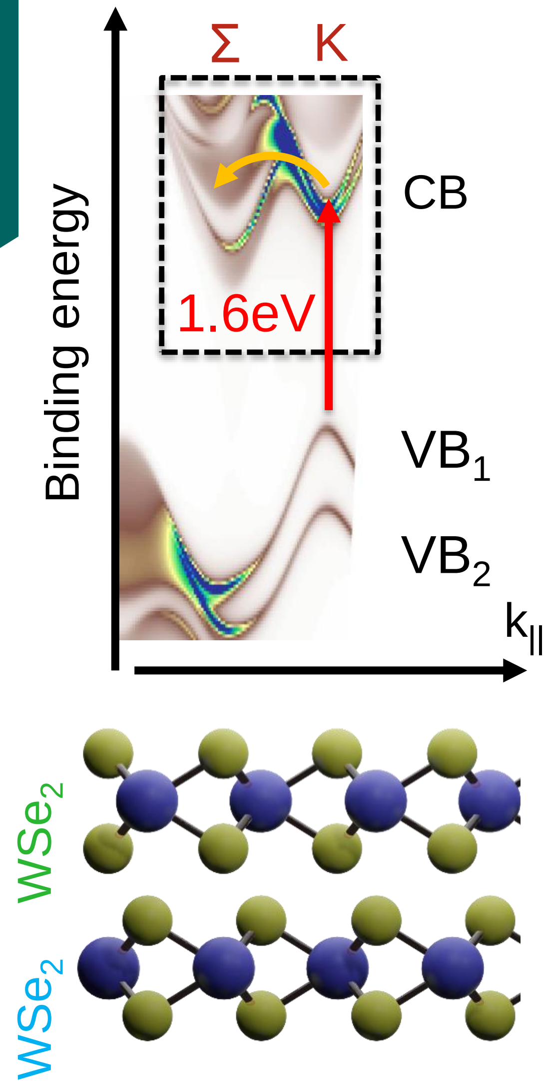


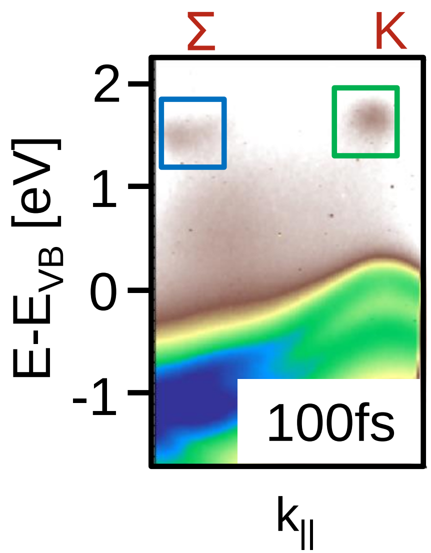


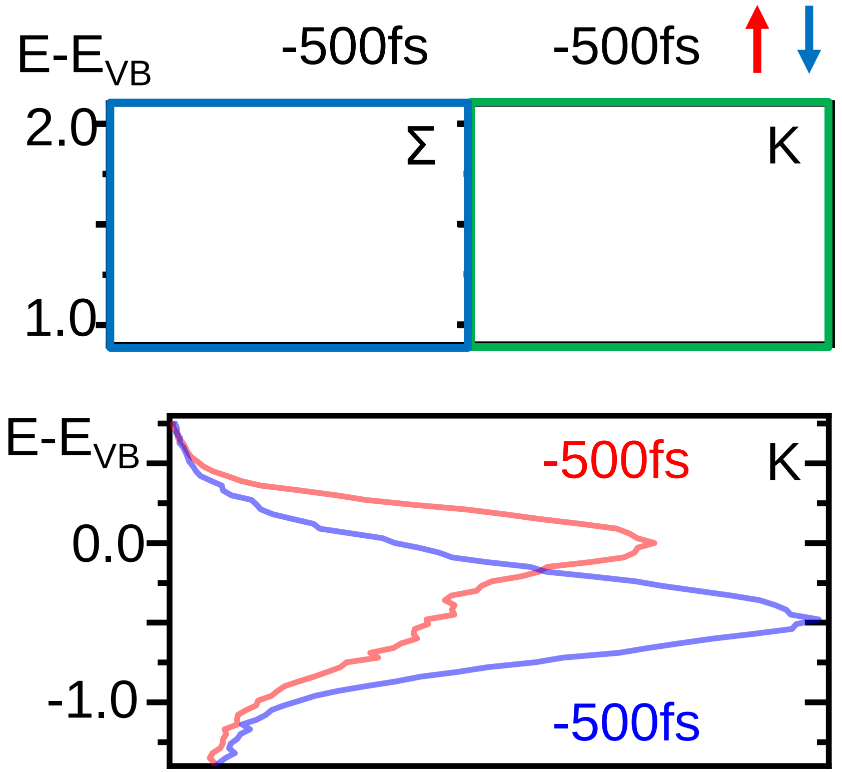
- Capture a movie of carrier dynamics in real time
- Quantify the population dynamics of electrons and holes in momentum space

Ultrafast Spin Carrier Dynamics of WSe_2

Spin-dependent **excited states** band structure





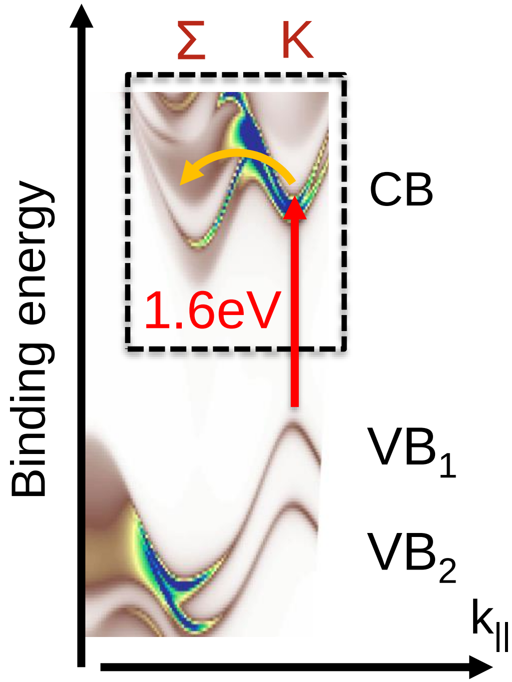


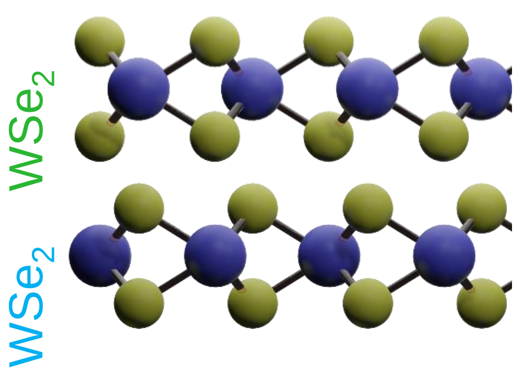
- Spin conserving intervalley scattering from K to Σ valley

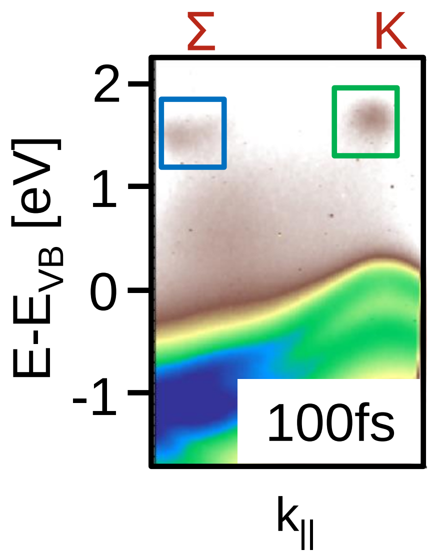
1.55eV, 30fs, π pol, 0.6mJ/cm²

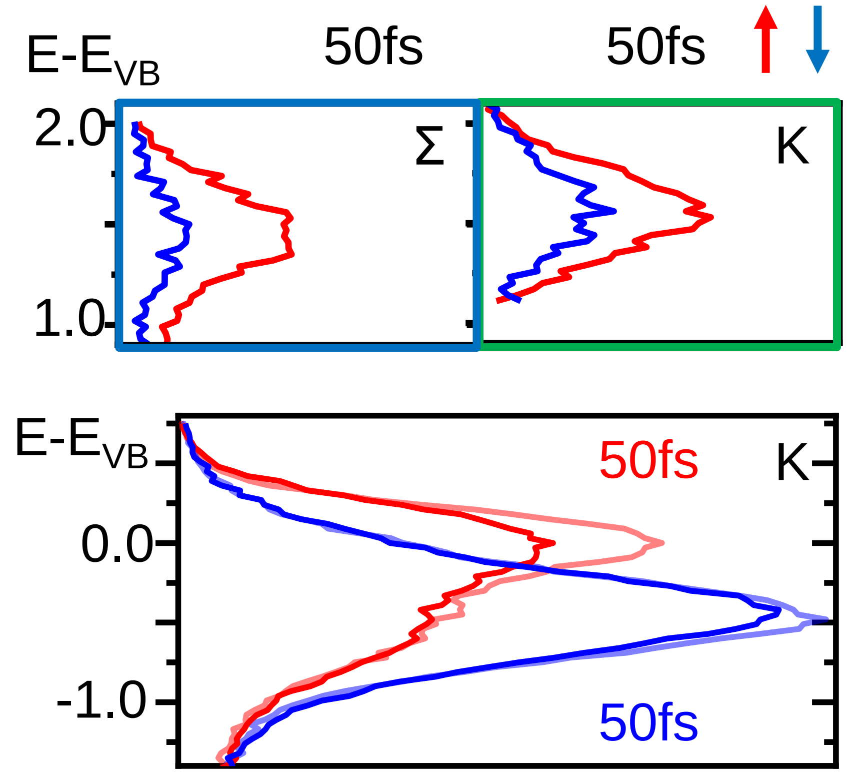
Ultrafast Spin Carrier Dynamics of WSe_2

Spin-dependent **excited states** band structure









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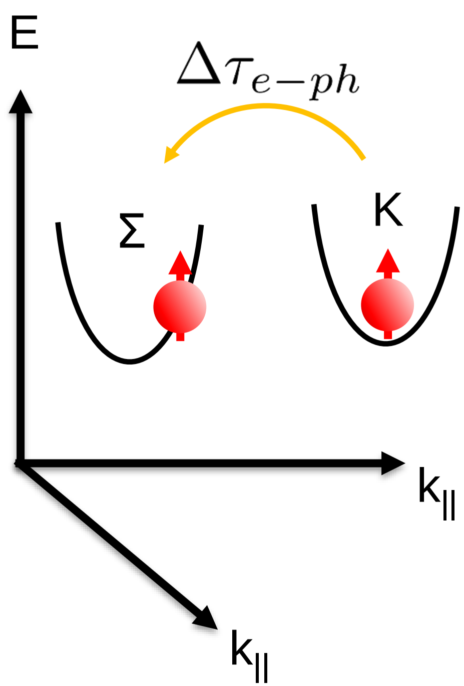
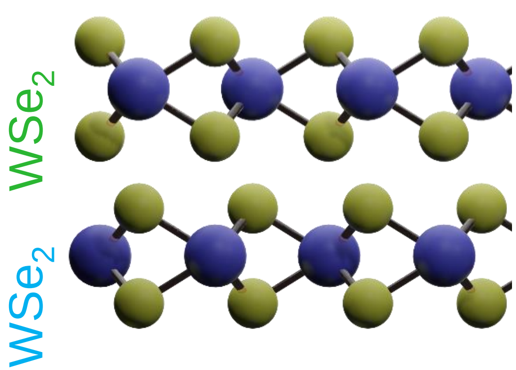
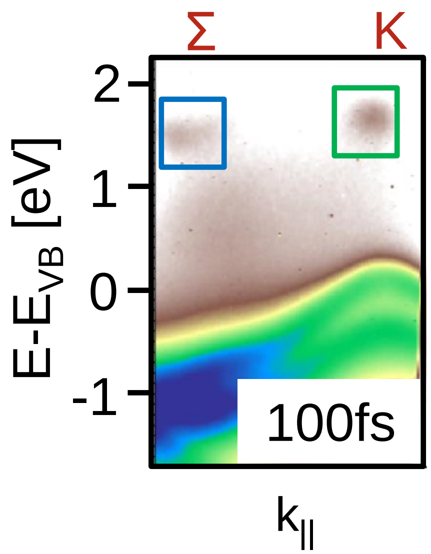
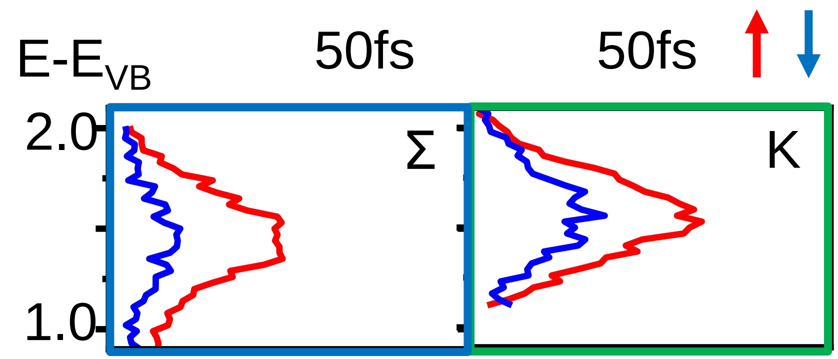
Ultrafast Spin Carrier Dynamics of WSe_2

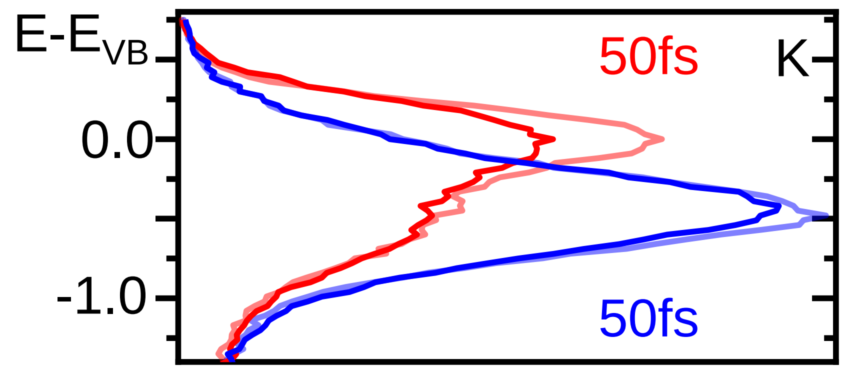
Spin-dependent **excited states** band structure











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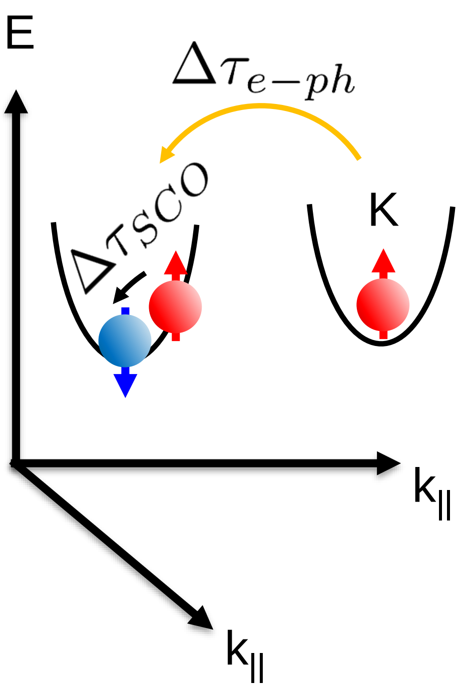
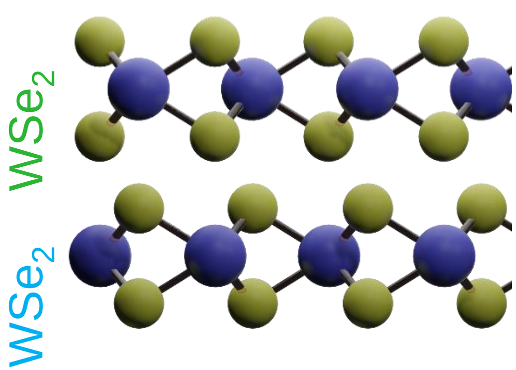
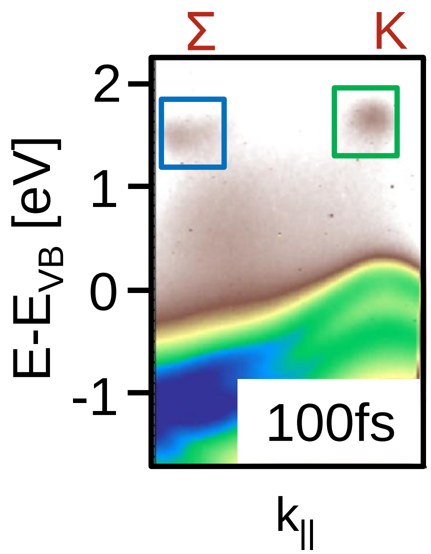
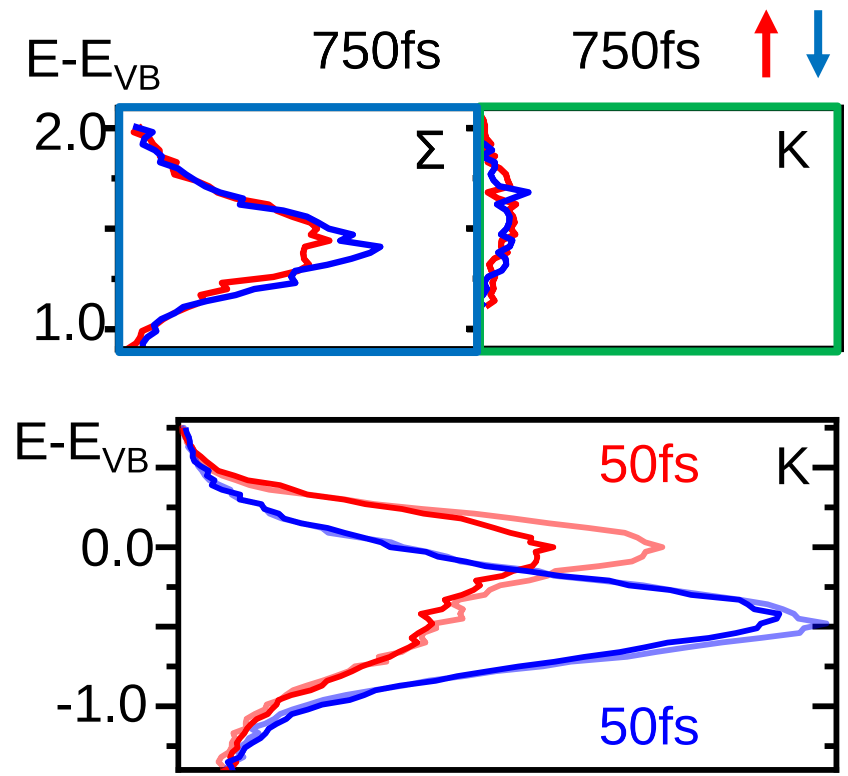
Ultrafast Spin Carrier Dynamics of WSe_2

Spin-dependent **excited states** band structure





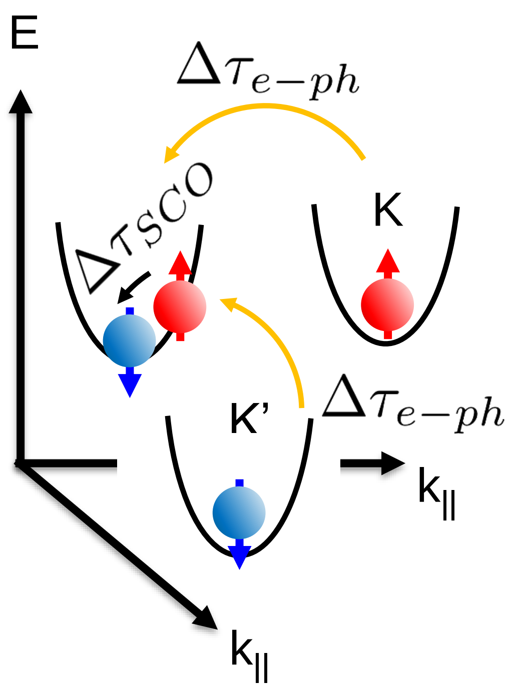


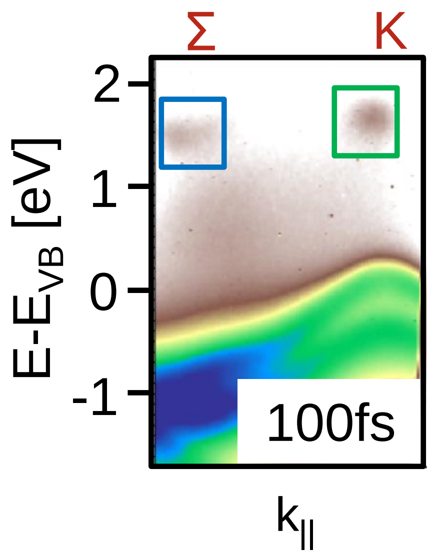


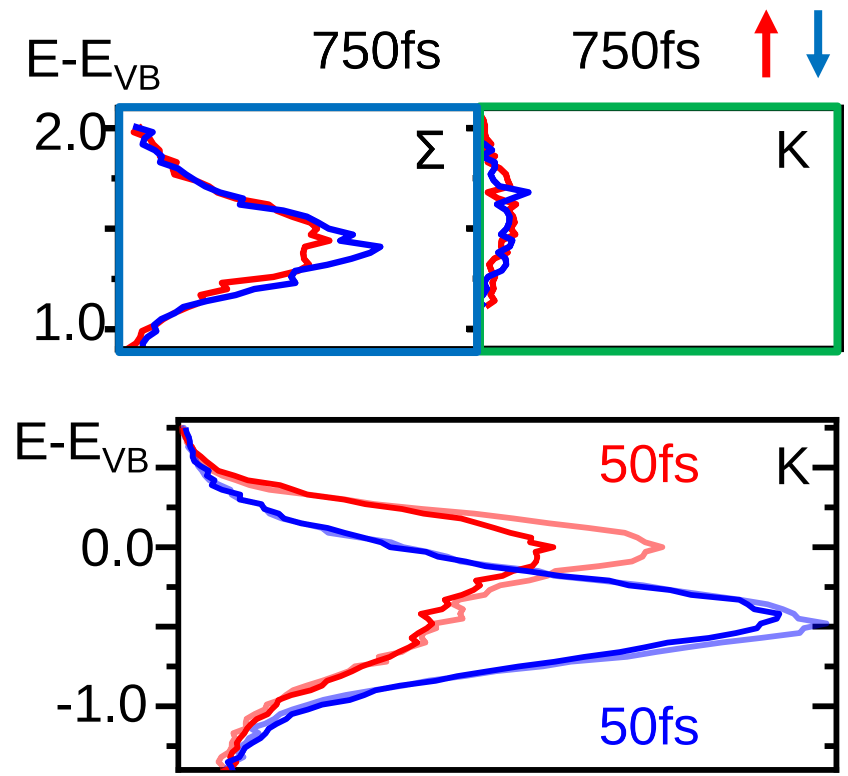
- Spin conserving intervalley scattering from K to Σ valley
- Reversal of spin polarization Σ valley

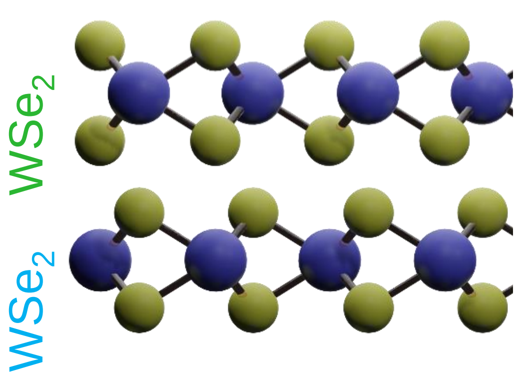
Ultrafast Spin Carrier Dynamics of WSe_2

Spin-dependent **excited states** band structure









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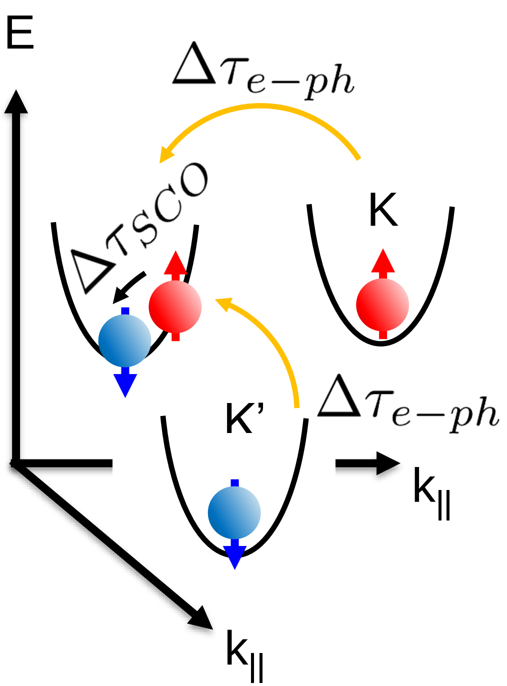
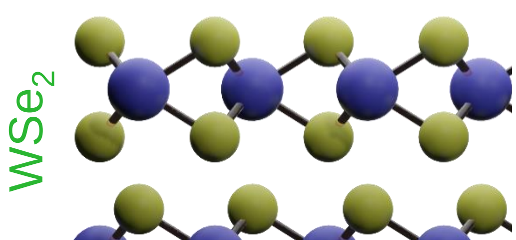
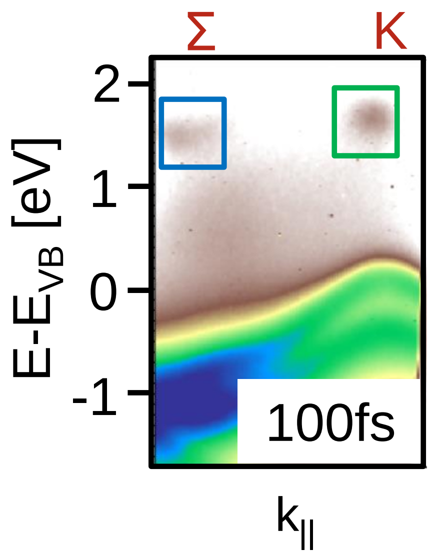
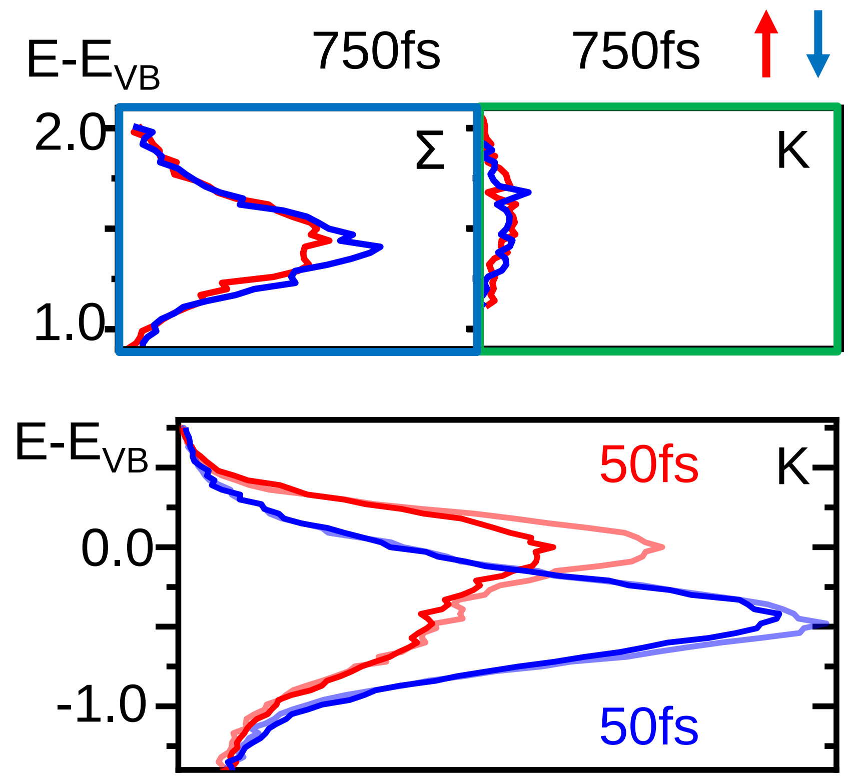
Ultrafast Spin Carrier Dynamics of WSe_2

Spin-dependent **excited states** band structure









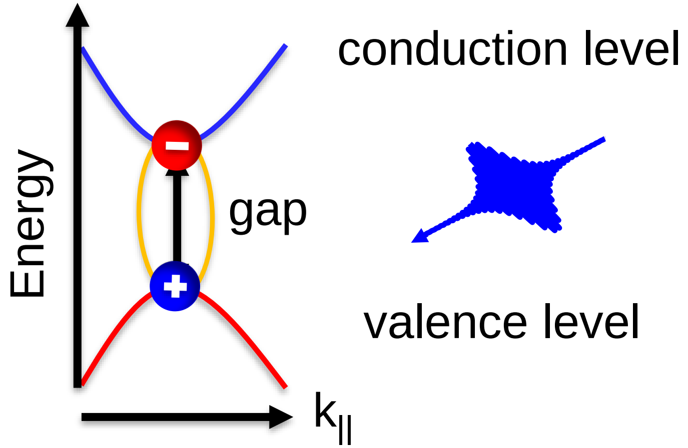
- Spin conserving intervalley scattering from K to Σ valley

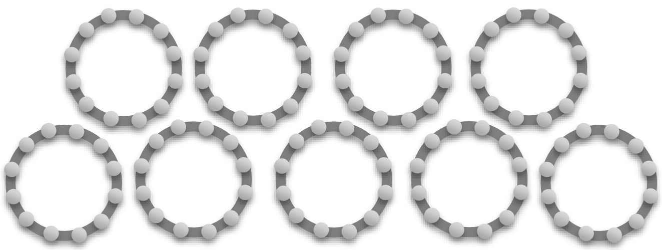
Direct access to spin-dependent inter- and intraband scattering of electrons and holes in real time

Excited Charge Carriers in Ultrathin Semiconductors

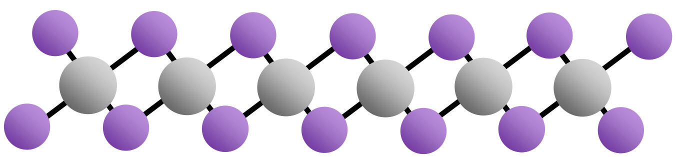
Spatial confinement of carriers

Energy level diagram





Molecular semiconductors



Inorganic semiconductors

Excited Charge Carriers in Ultrathin Semiconductors

Spatial confinement of carriers

Different types of excitons:

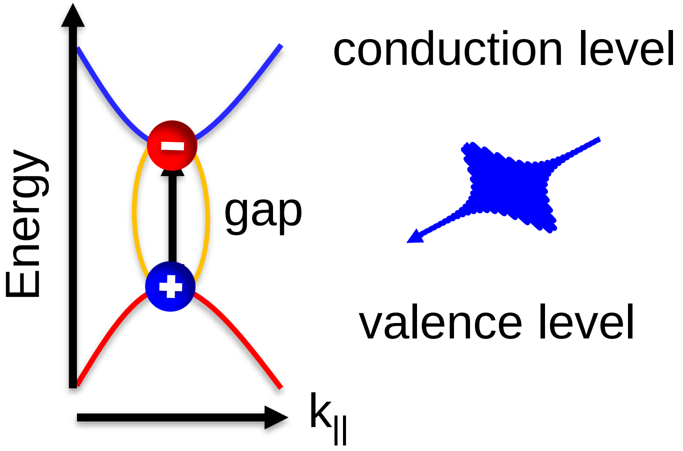
- Localized Frenkel excitons

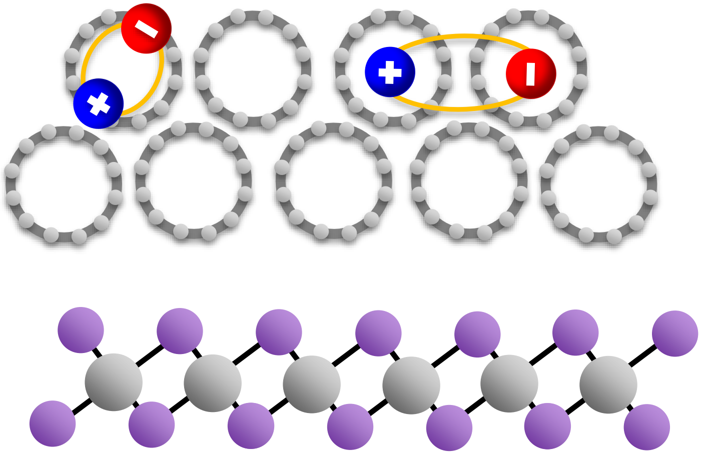
→ Charge neutral quasi-particles

- Charge transfer (CT) excitons

→ Charge-separated states

Energy level diagram





Molecular semiconductors

Inorganic semiconductors

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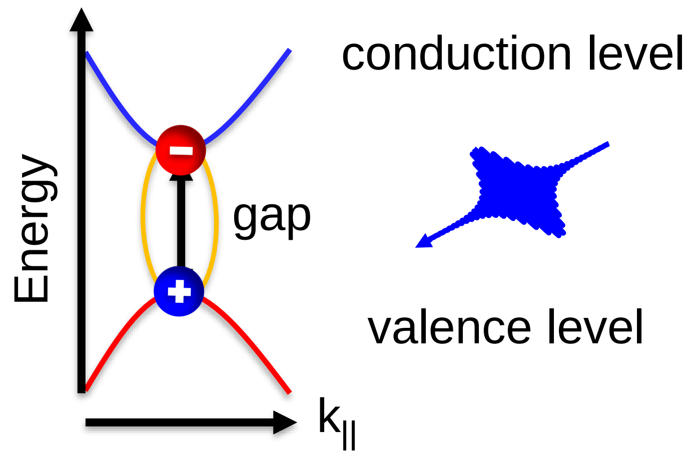
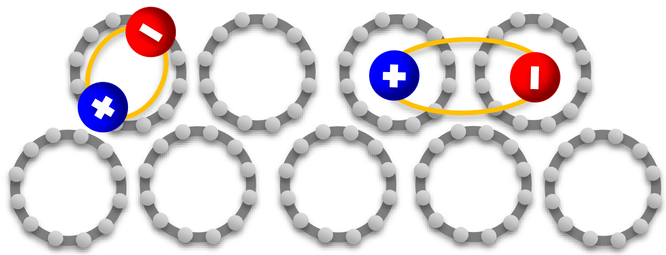
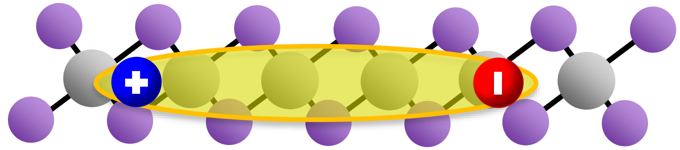
Energy level diagram





Molecular semiconductors



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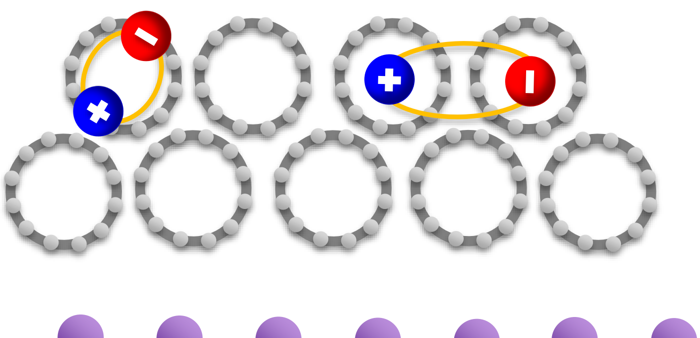
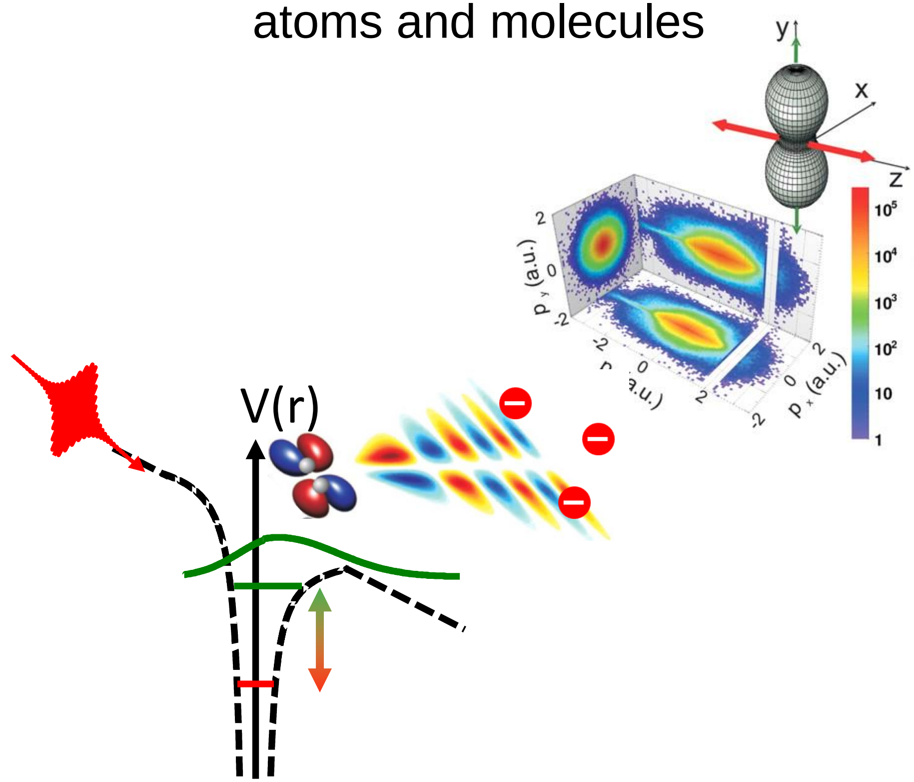


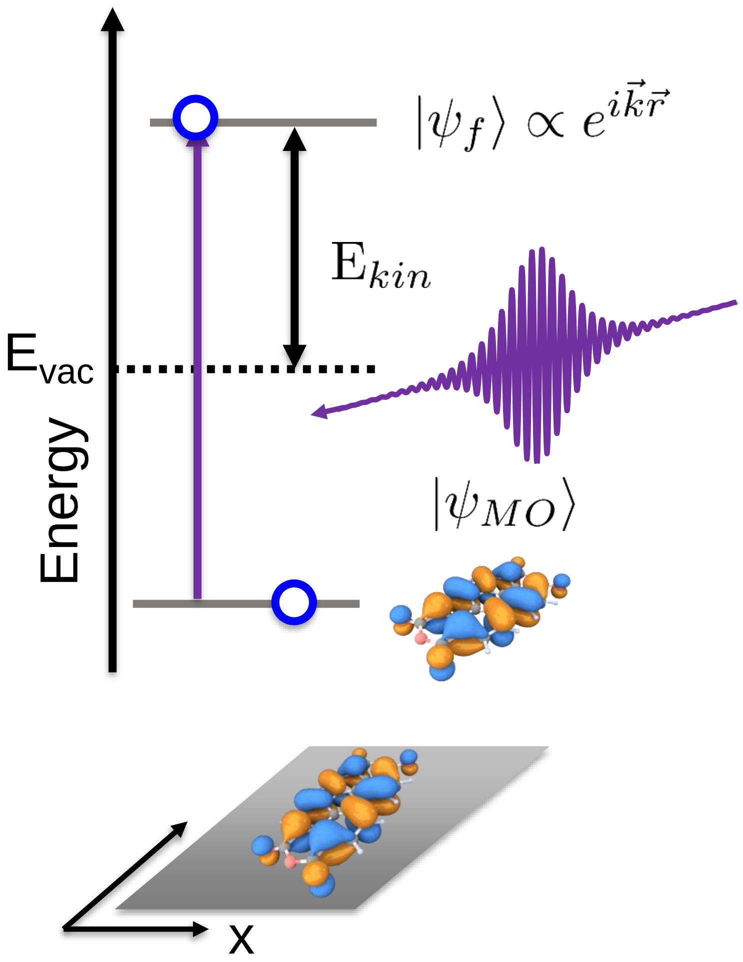
Orbital tomography for free atoms and molecules



Can we also image the spatial confinement of carriers in solids?

Spatial Confinement of Charge Carriers in Solids

Photoemission Orbital Tomography (POT) for Molecular Materials



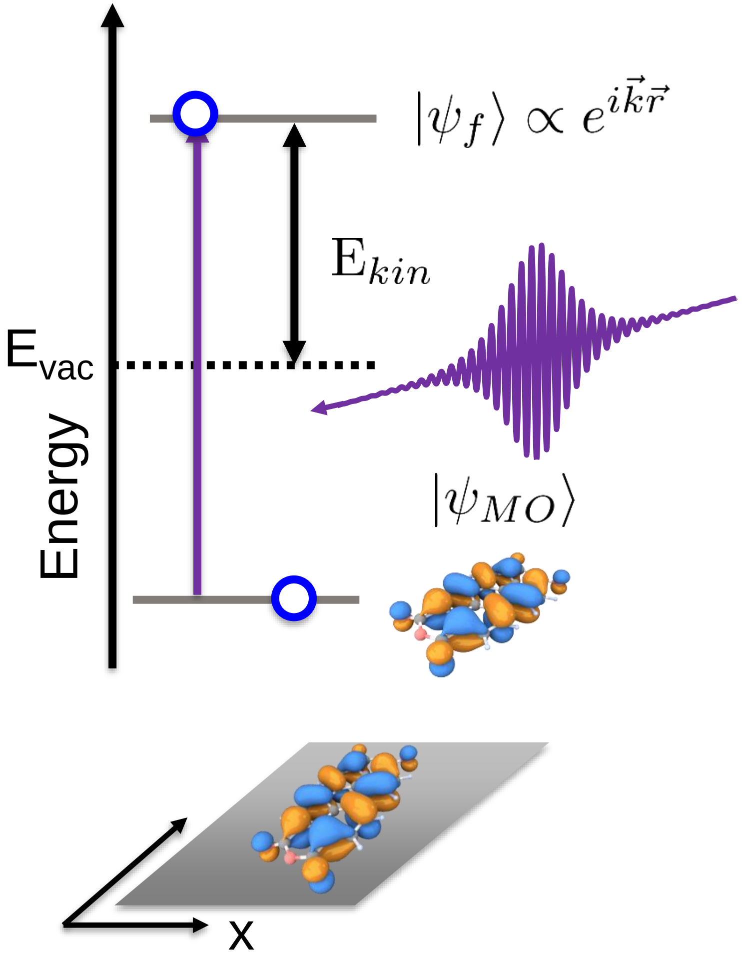
Photocurrent: Fermi's Golden Rule

$$I(E_{kin}, \theta, \varphi) \propto |FT[\psi_{MO}]|^2$$

P. Puschnig, M. Ramsey, Science **326**, 702 (2009)

Spatial Confinement of Charge Carriers in Solids

Photoemission Orbital Tomography (POT) for Molecular Materials



Photocurrent: Fermi's Golden Rule

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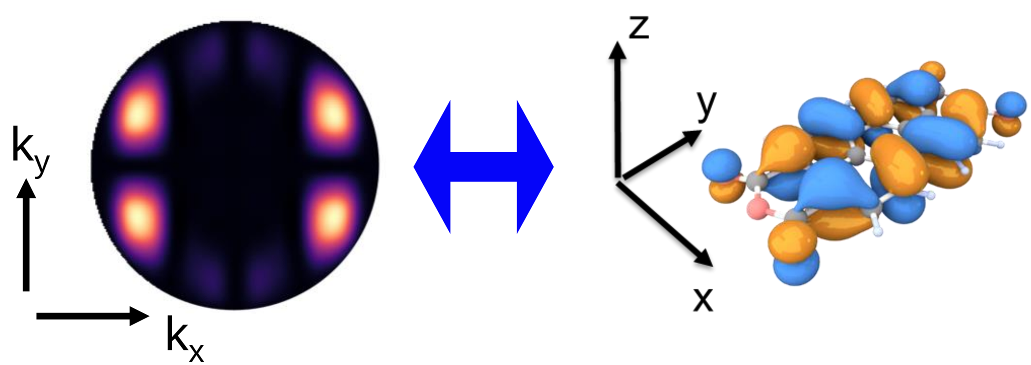
Momentum-resolved photoemission yield

Fourier Transform of Initial State



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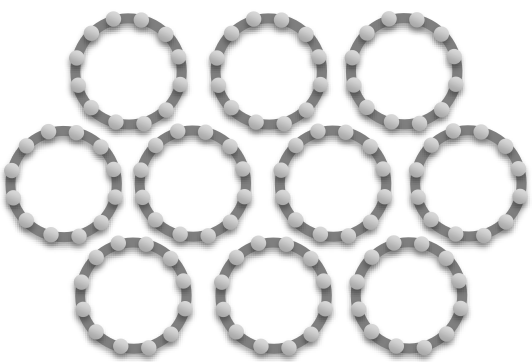
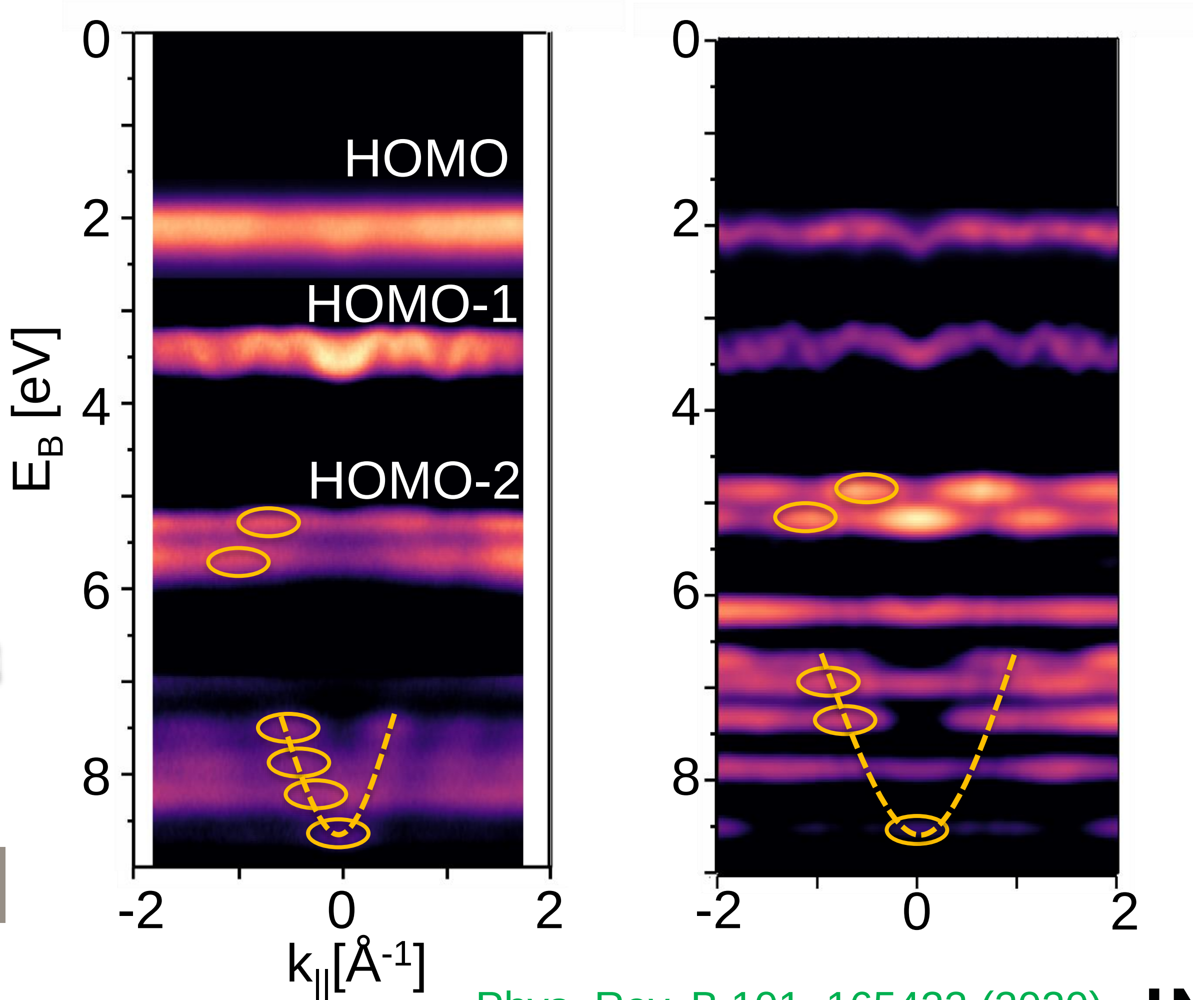
Spatial Confinement of Charge Carriers in Solids

Excited States POT for Molecular Materials



C_{60} on Ag(111)

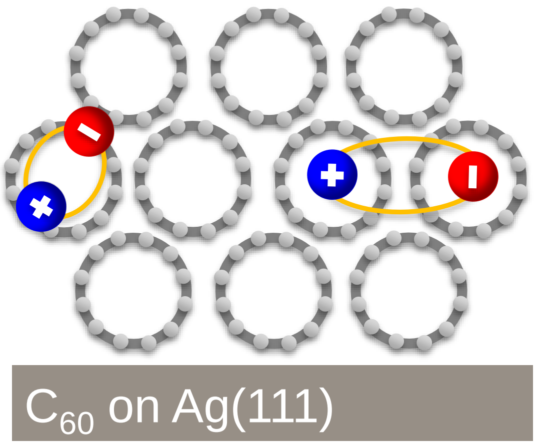
ARPES C_{60} thin film | DFT and POT simulation

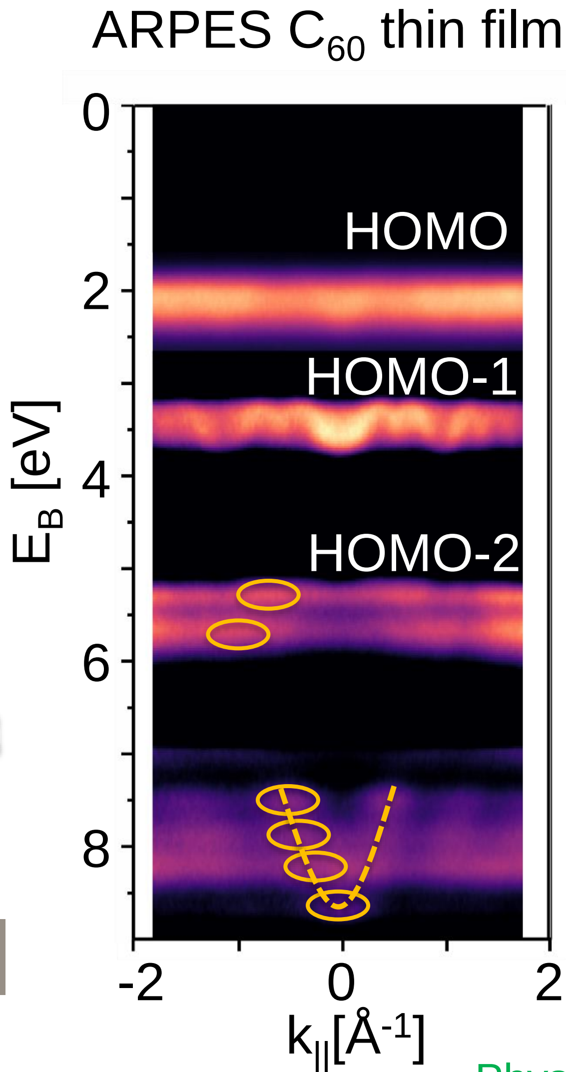


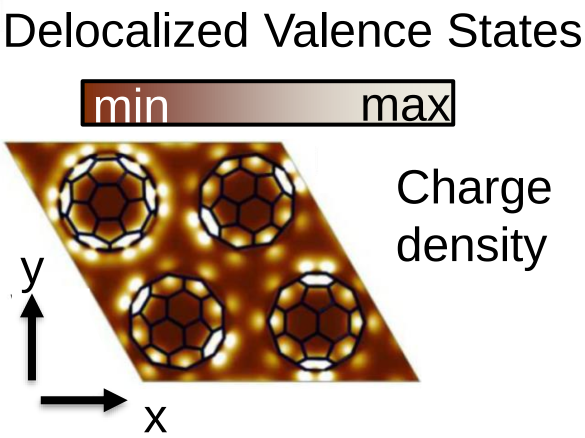
Phys. Rev. B 101, 165422 (2020)

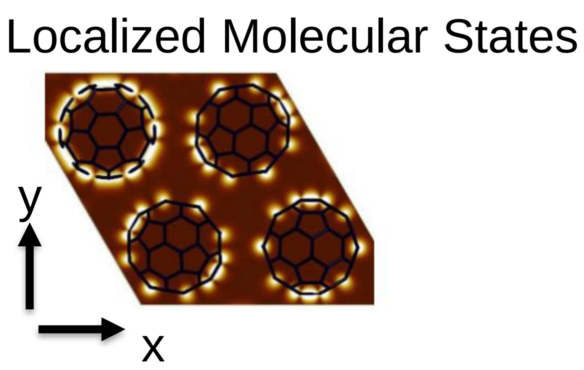
Spatial Confinement of Charge Carriers in Solids

Excited States POT for Molecular Materials





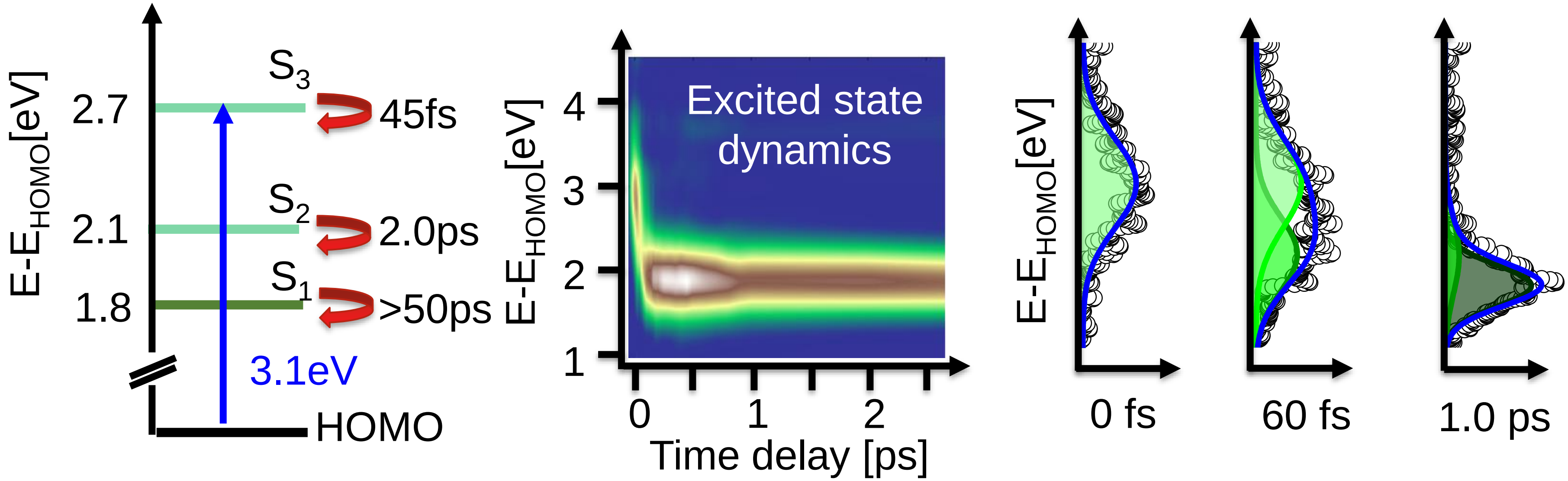


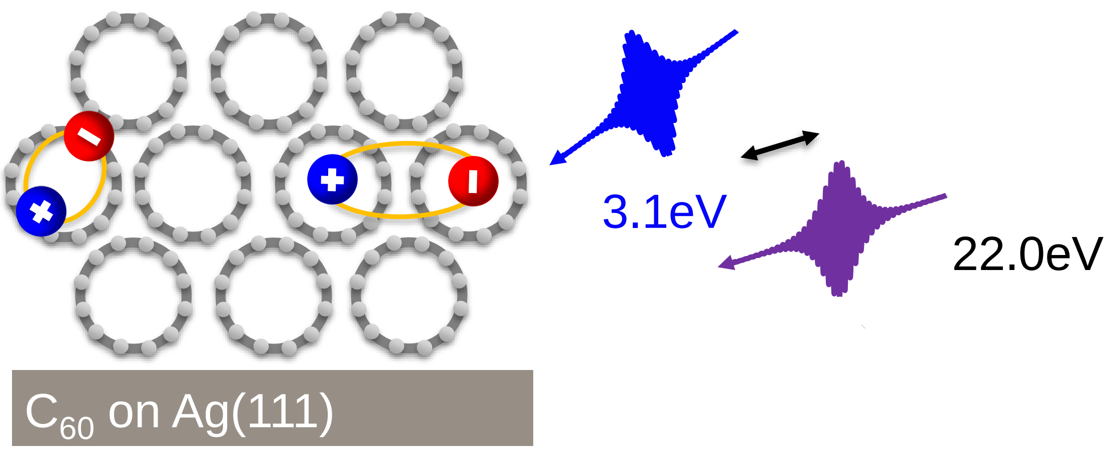


Phys. Rev. B 101, 165422 (2020)

Spatial Confinement of Charge Carriers in Solids

Excited States POT for Molecular Materials





Nat. Commun. 10, 1470 (2019)

Spatial Confinement of Charge Carriers in Solids

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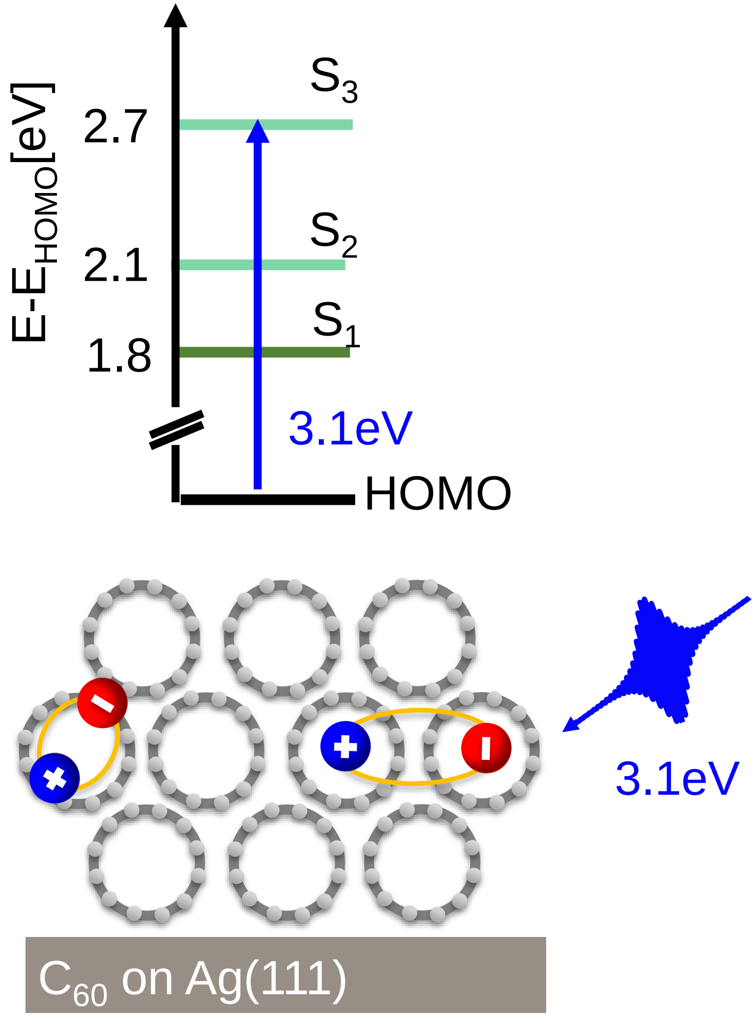

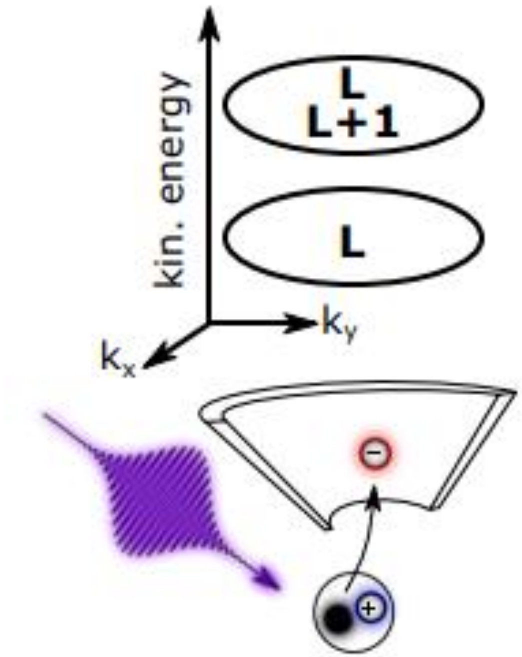

Nat. Commun. 10, 1470 (2019) ; Nat. Commun. 15, 1804 (2024)

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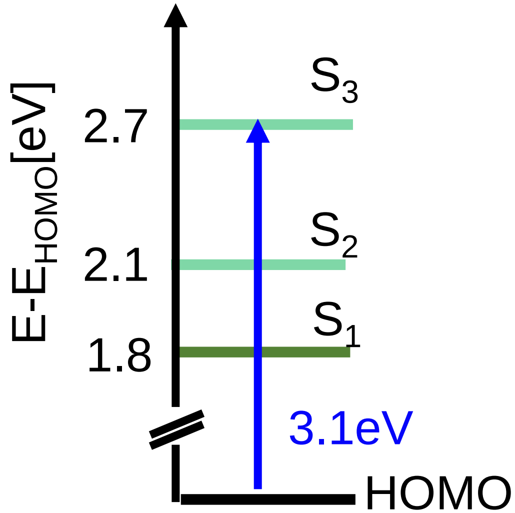
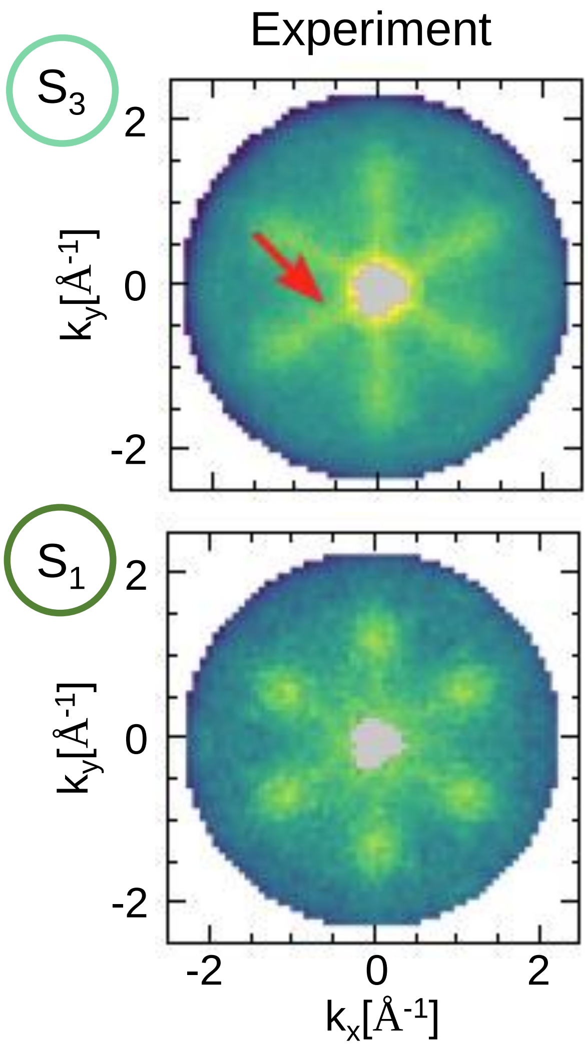
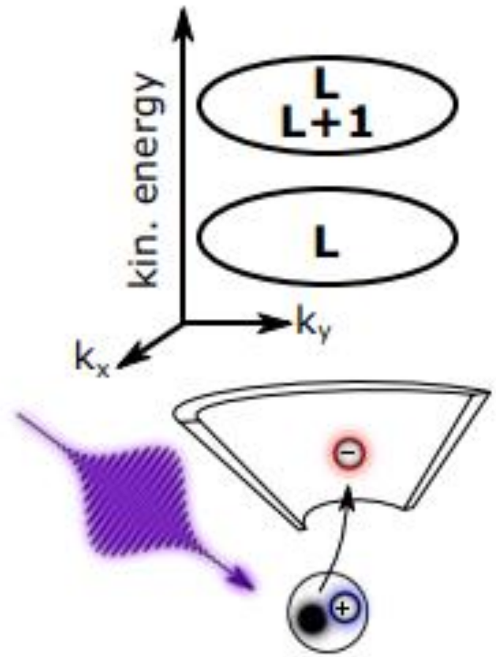
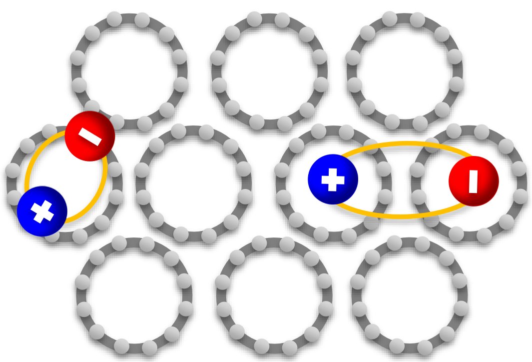






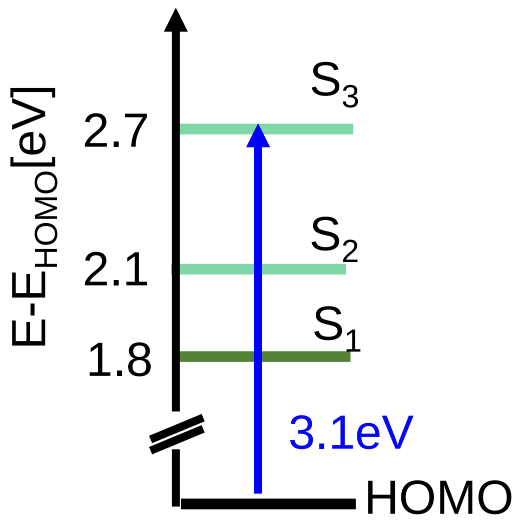


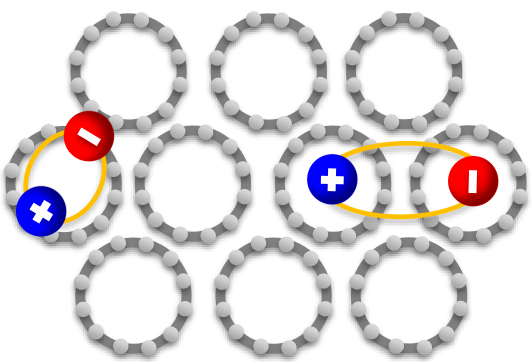
C_{60} on Ag(111)

Nat. Commun. 10, 1470 (2019) ; Nat. Commun. 15, 1804 (2024)

Spatial Confinement of Charge Carriers in Solids

Excited States POT for Molecular Materials





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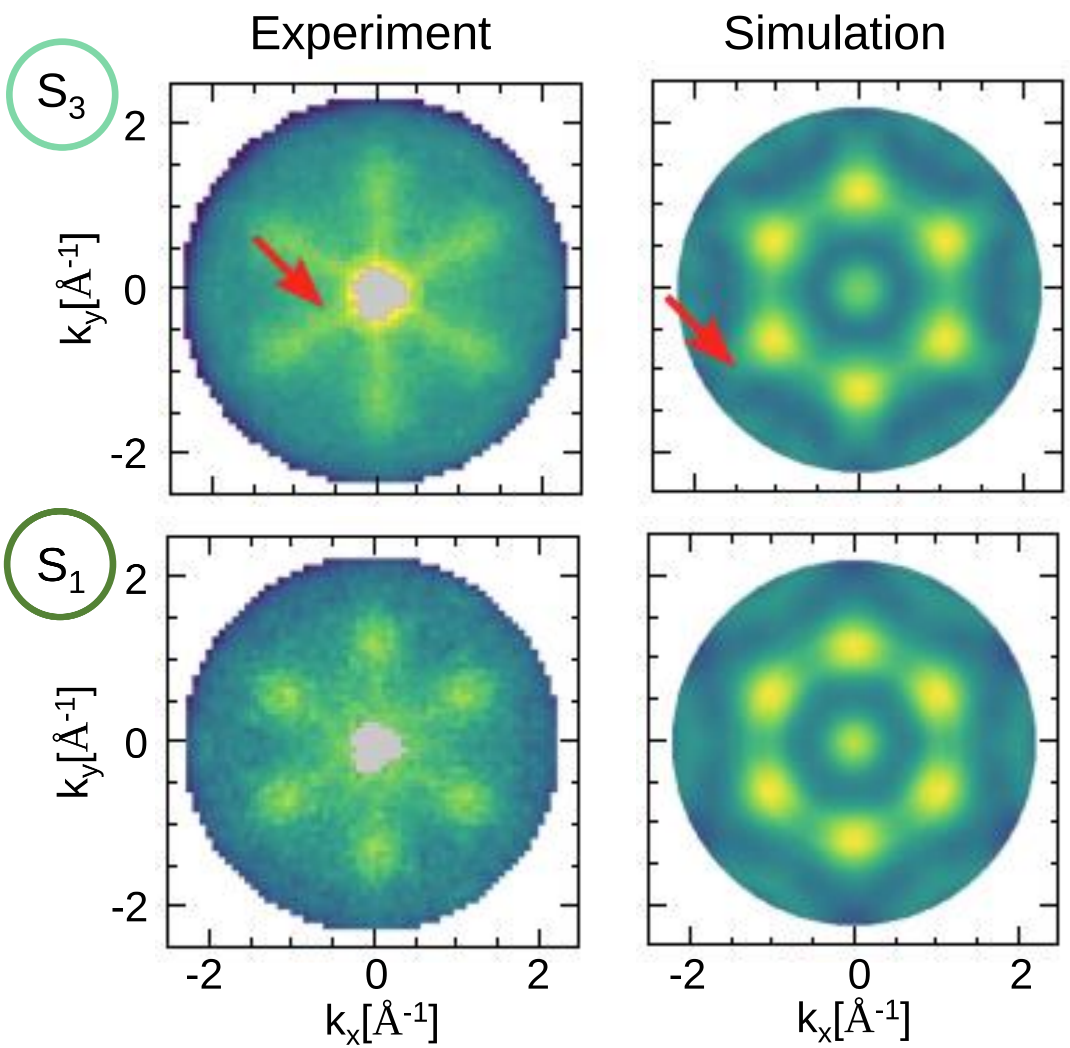

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Spatial Confinement of Charge Carriers in Solids

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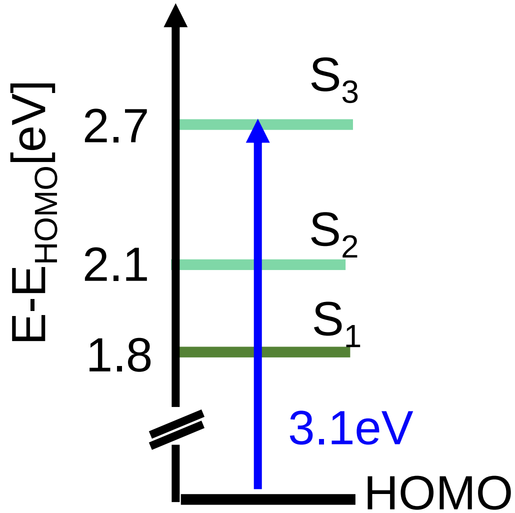

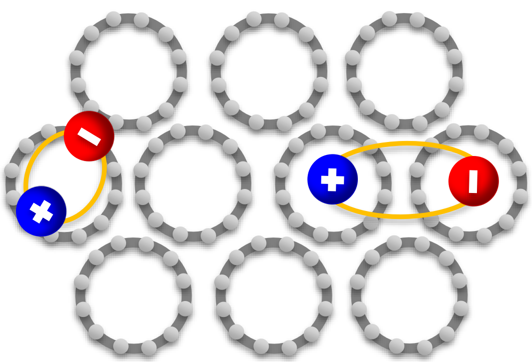

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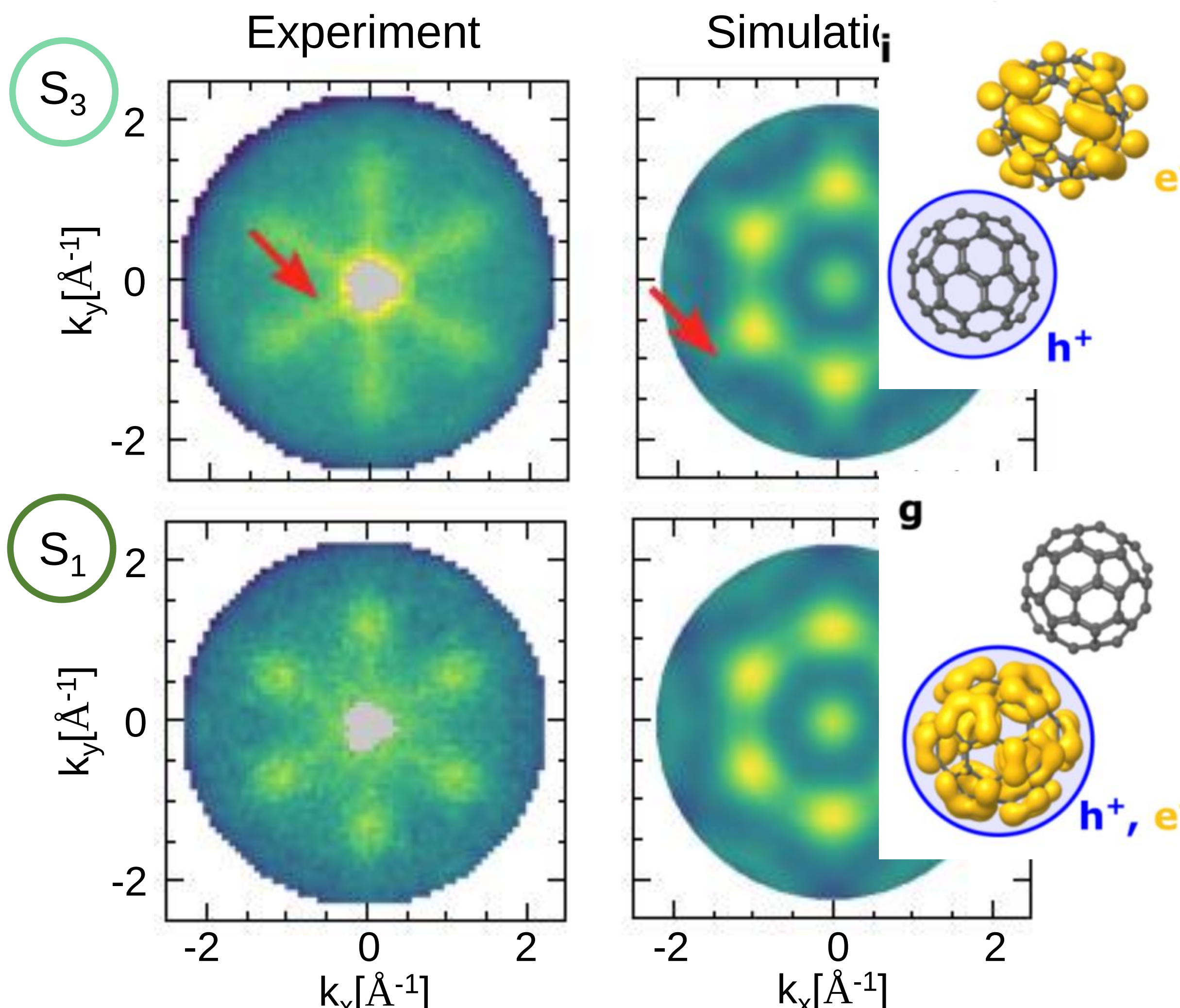
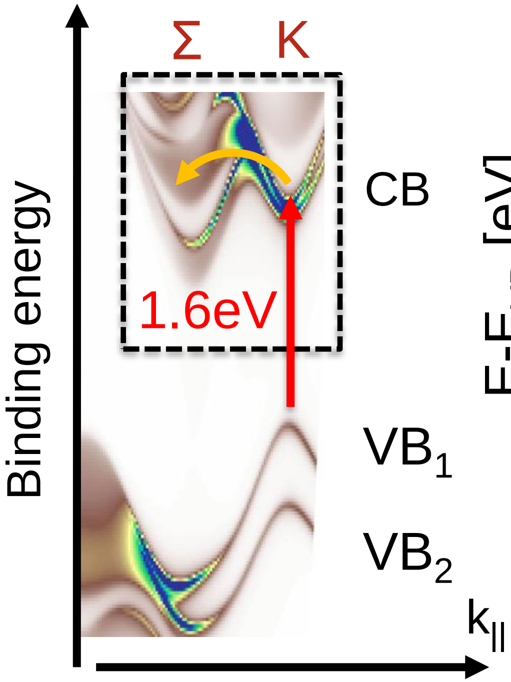
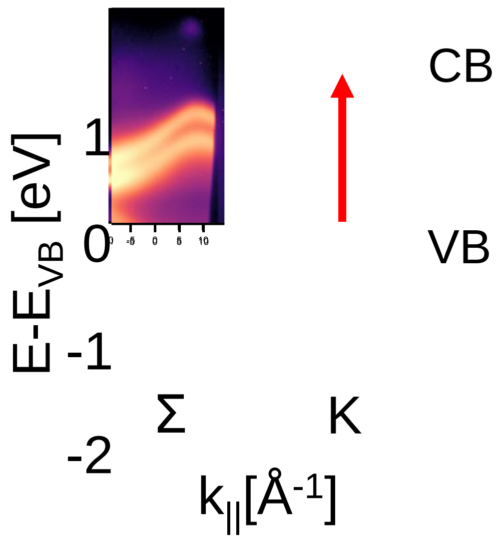


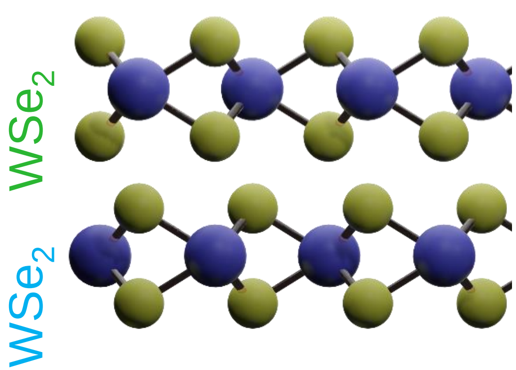
Nat. Commun. 10, 1470 (2019) ; Nat. Commun. 15, 1804 (2024)

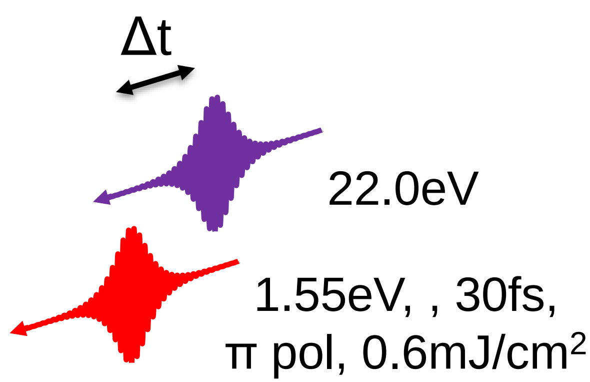
Spatial Confinement of Charge Carriers in Solids

Excited States POT for 2D Materials









Spatial Confinement of Charge Carriers in Solids

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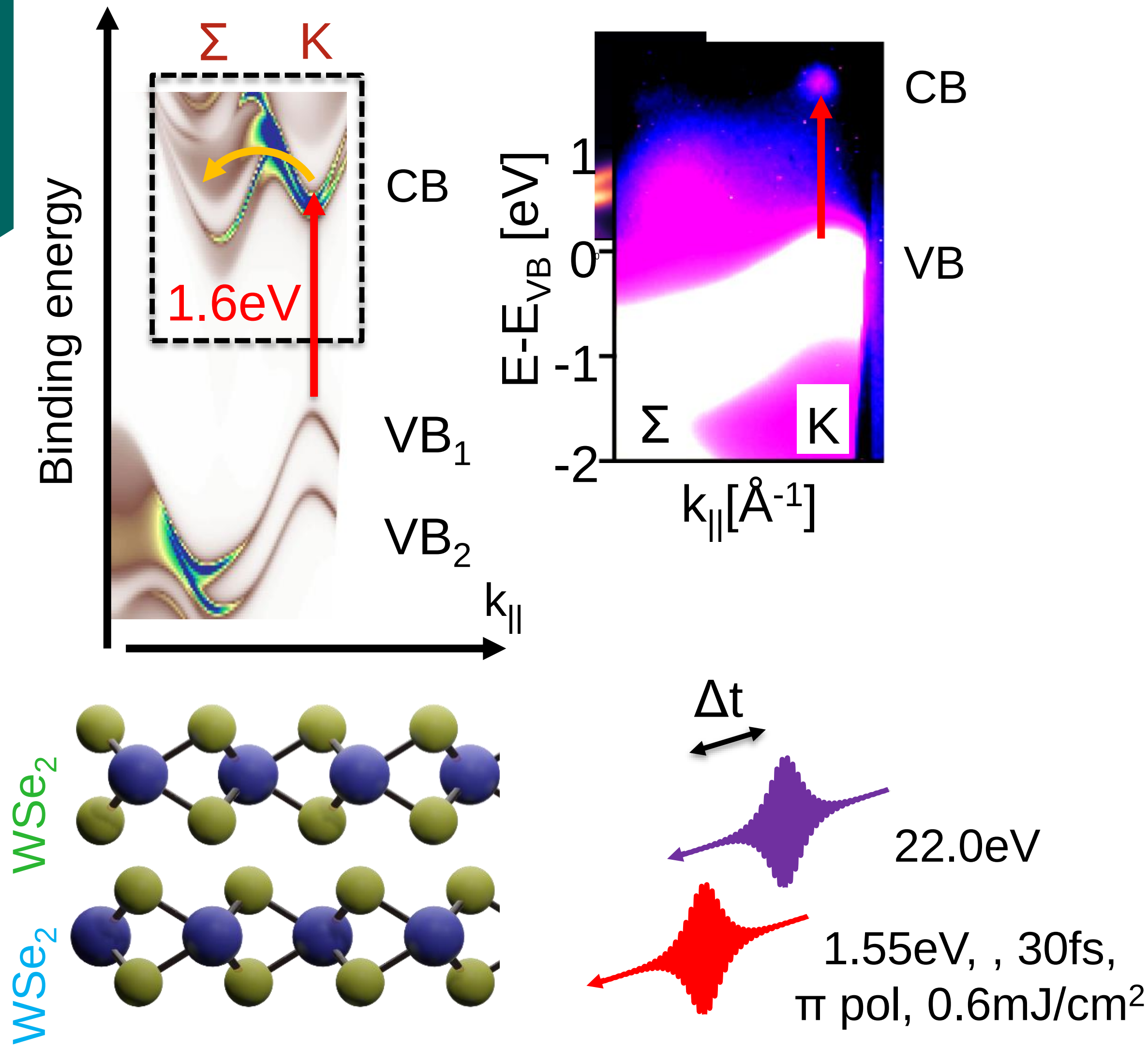

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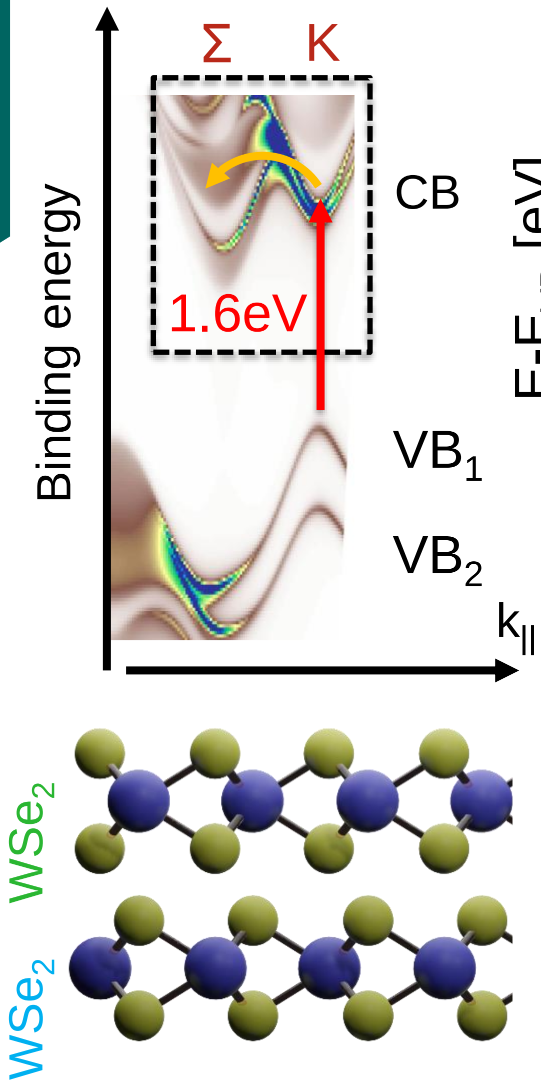
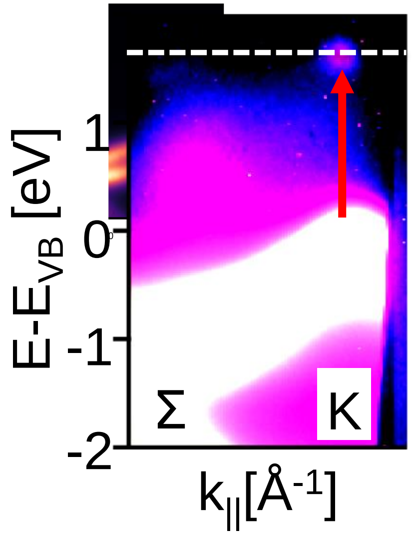
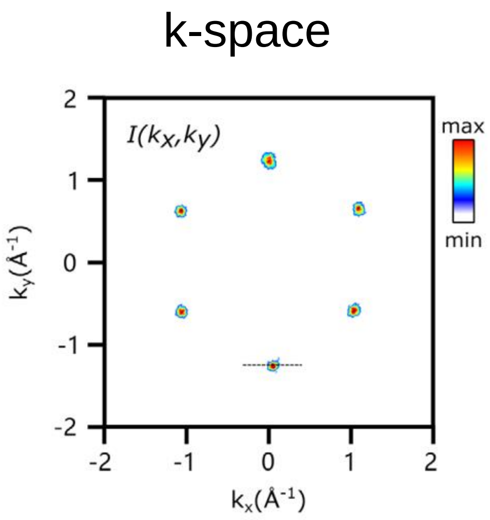
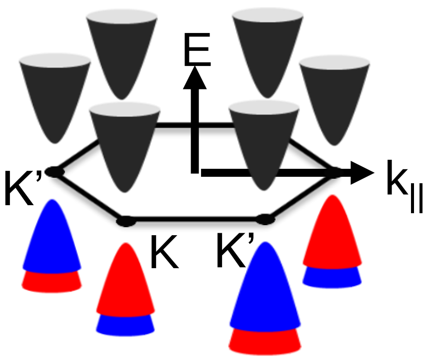




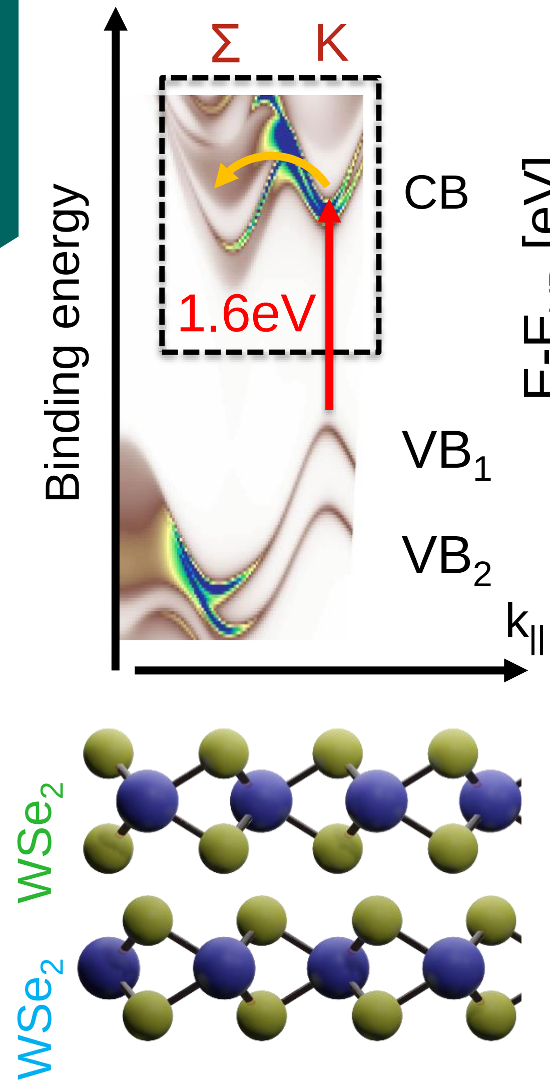


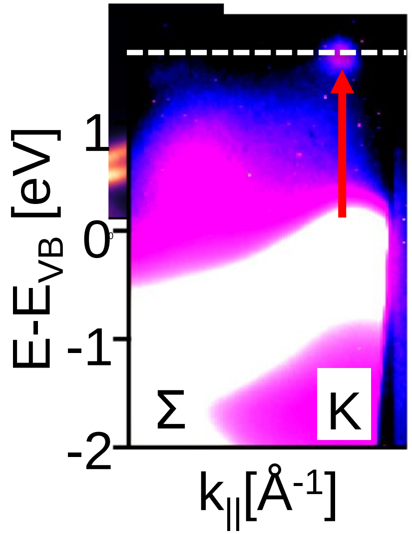


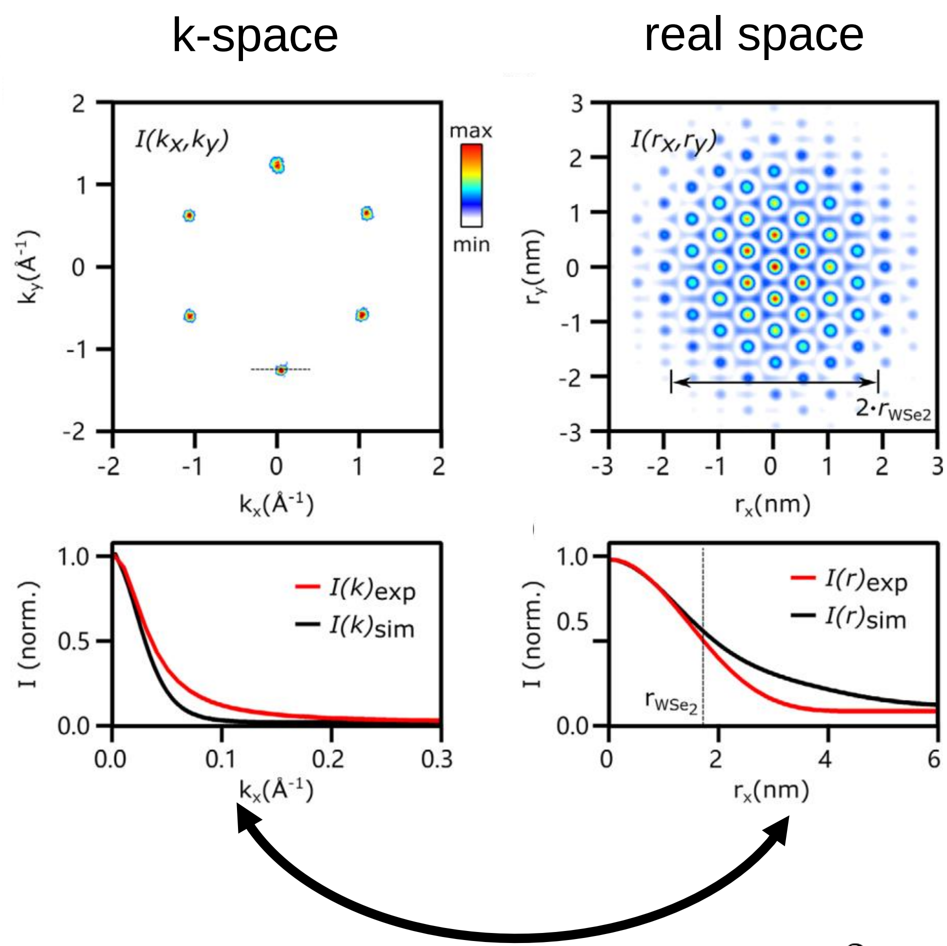
S. Dong et al. Nat Sci 2021; 1:e10010

Spatial Confinement of Charge Carriers in Solids

Excited States POT for 2D Materials







$$\mathrm{I}(E_{kin},\theta,\varphi) \propto |FT[\psi_{MO}]|^2$$

S. Dong et al. Nat Sci 2021; 1:e10010

Conclusions

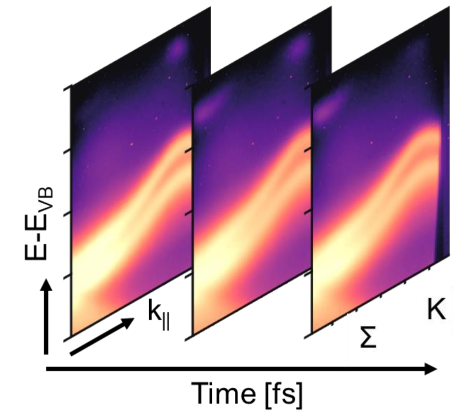
Ultrafast pump-probe photoemission with fs XUV pulses is a powerful tool for imaging

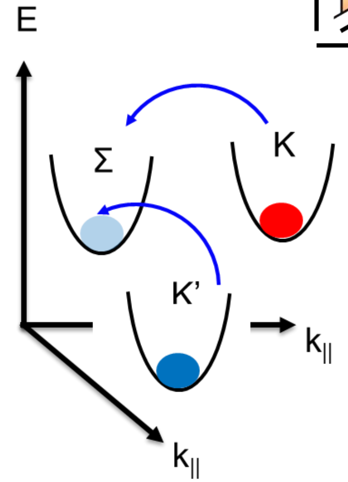
…. inter and interlayer charge and spin transfer processes

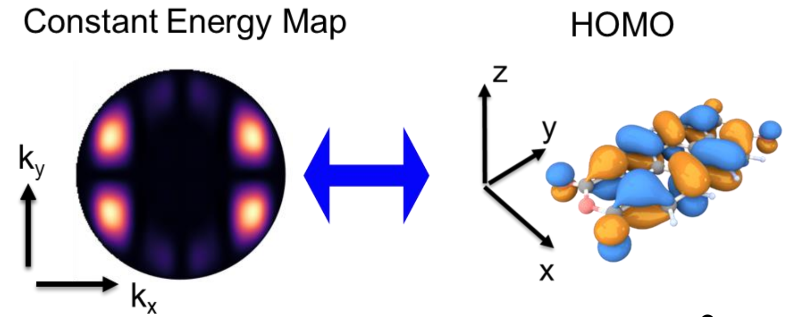
… the orbitals of excited molecular and 2D materials

… the transient charge doping of materials

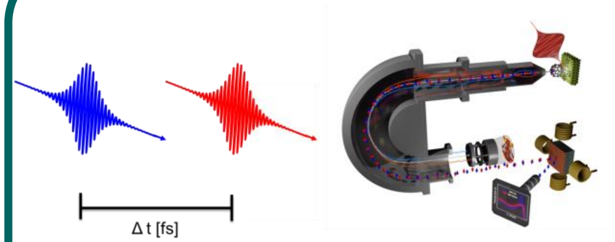
with femtosecond time resolution.



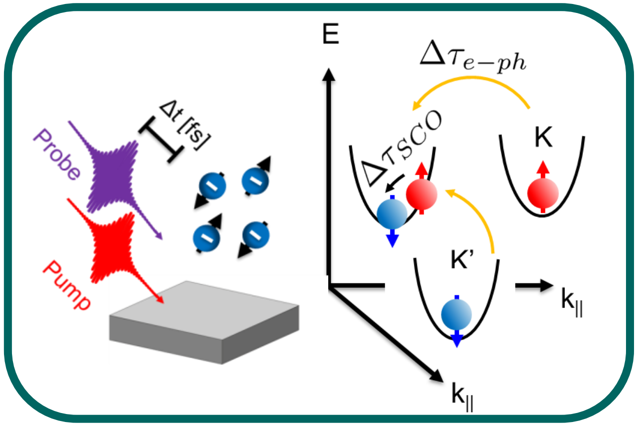




Outline

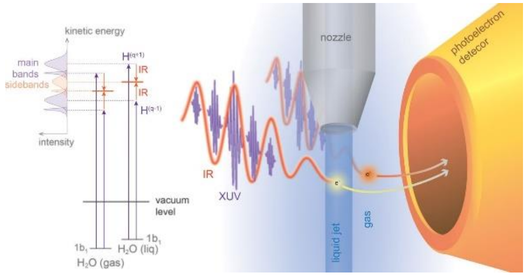


Ultrafast science in solids
… electrons in solids
… time-resolved photoemission spectroscopy



Monitoring….

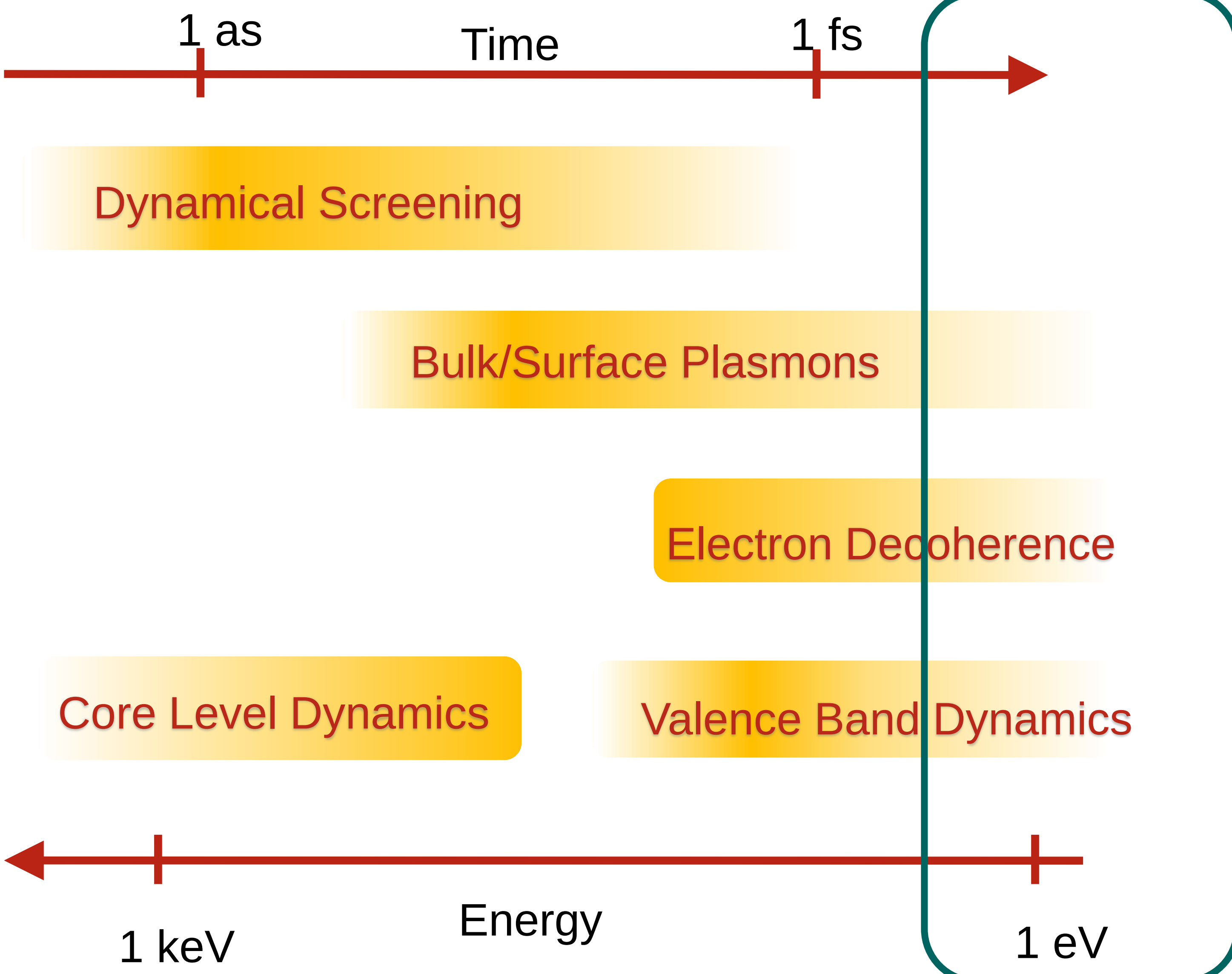
…. the population dynamics of charge and spin carriers in momentum space
… the nature and spatial distribution of charge carriers in direct space
… interlayer charge separation across interfaces



Perspectives and challenges of attosecond surface science

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy



Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

Attosecond pulse train

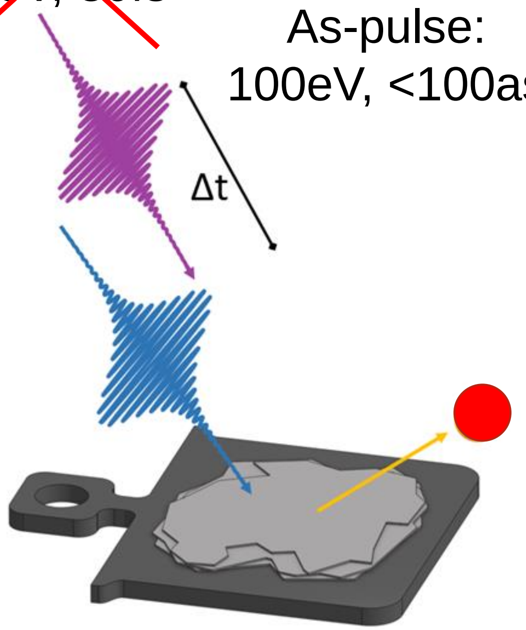
$$\delta t \cdot \delta\omega = const$$

Attosecond burst

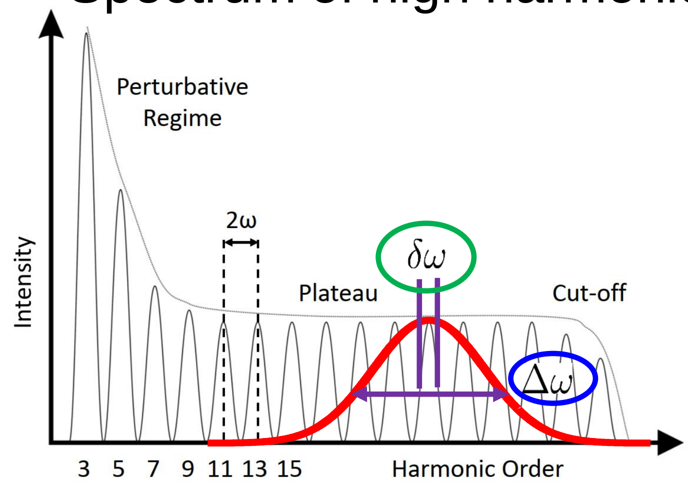
$$\Delta t \cdot \Delta\omega = const$$

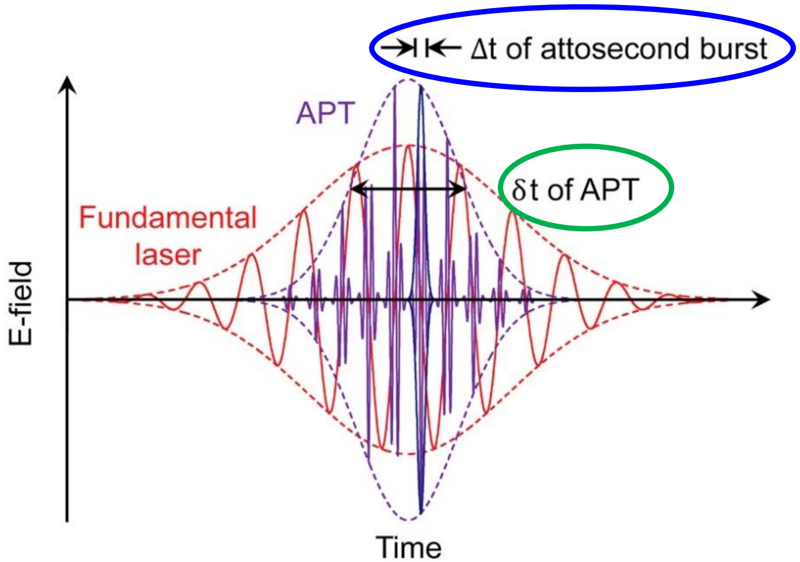
~~Probe pulse: 22eV, 30fs~~

As-pulse:
100eV, <100as



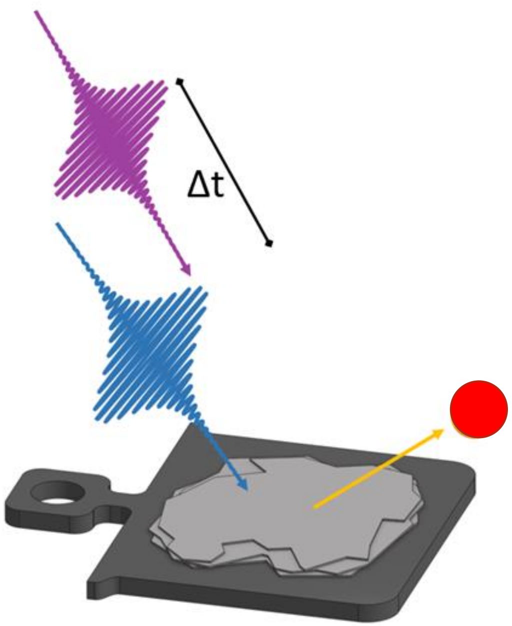
Spectrum of high harmonics



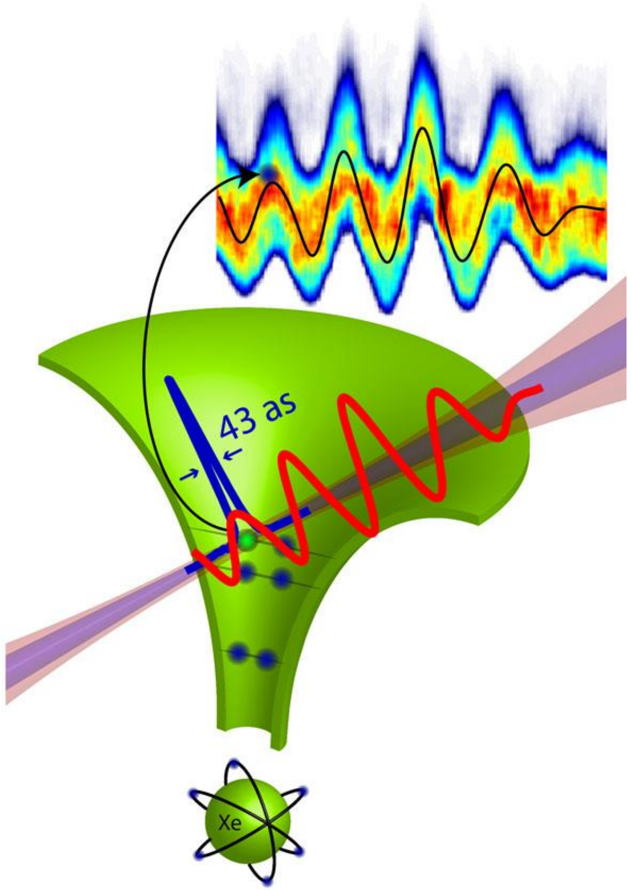


Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy



Attosecond streaking experiment with single attosecond pulses



Optics Express 25, 22 (2017)

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

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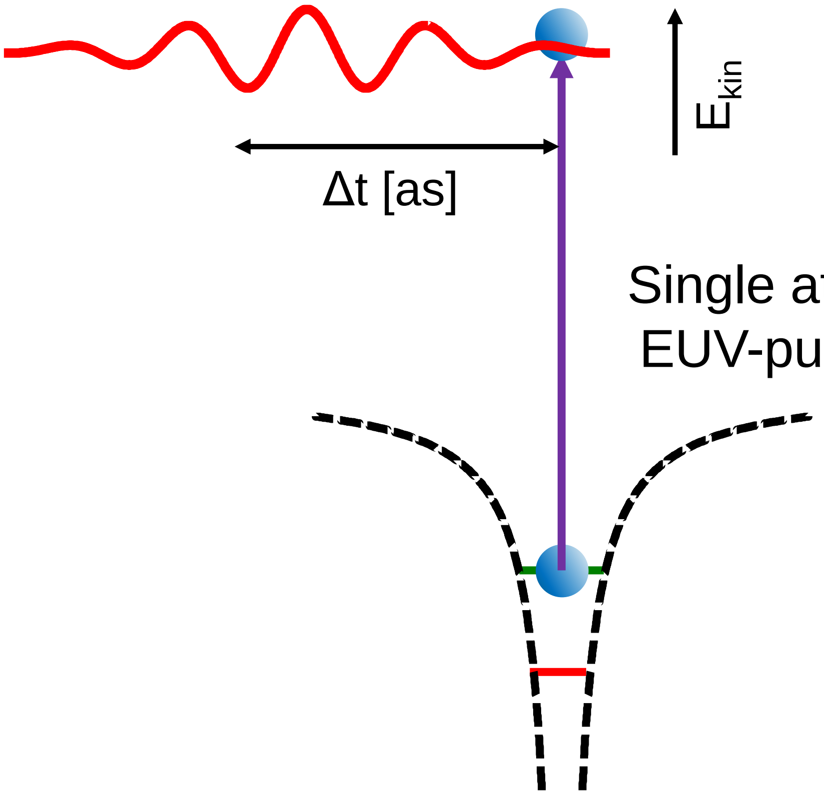

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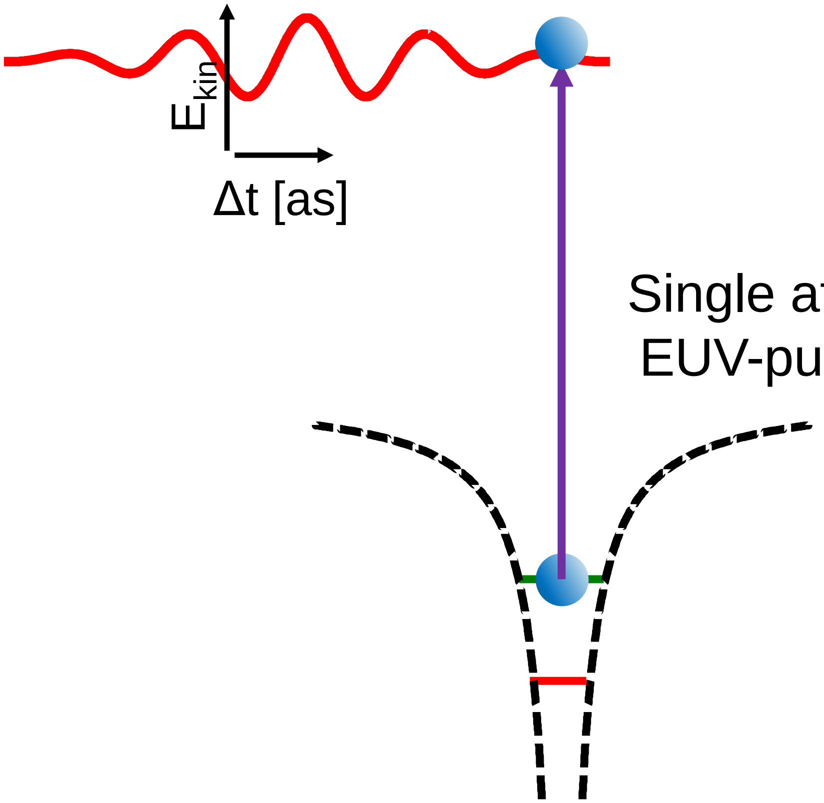
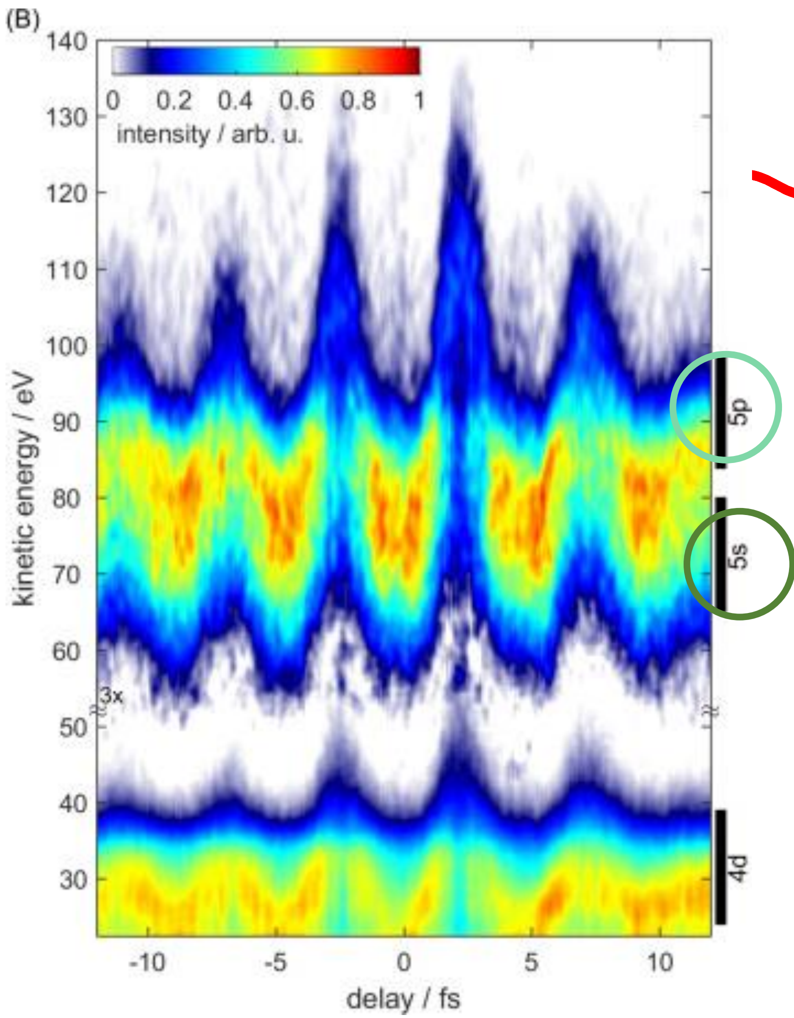


Single attosecond
EUV-pump pulse

Optics Express 25, 22 (2017)

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy



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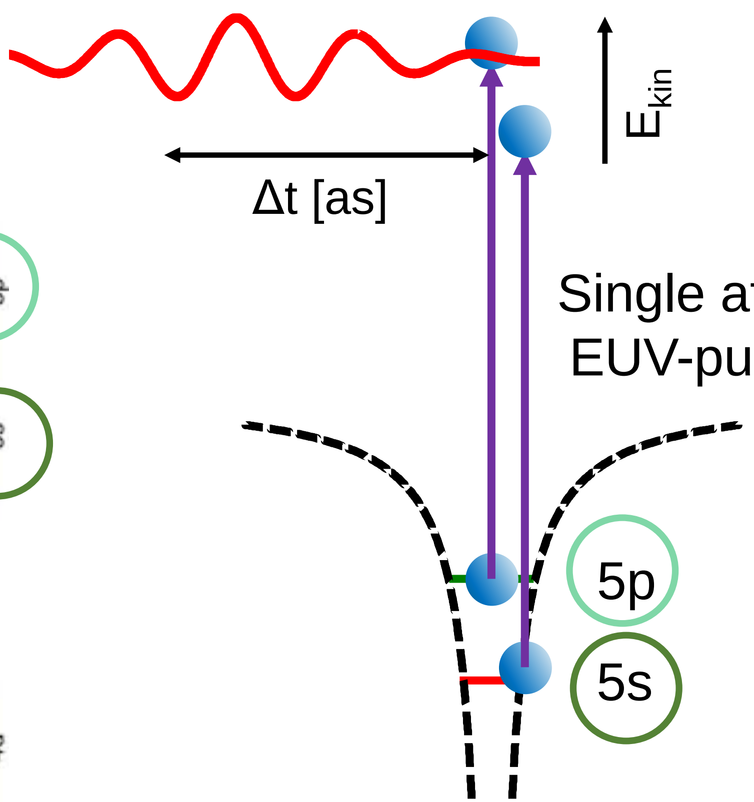
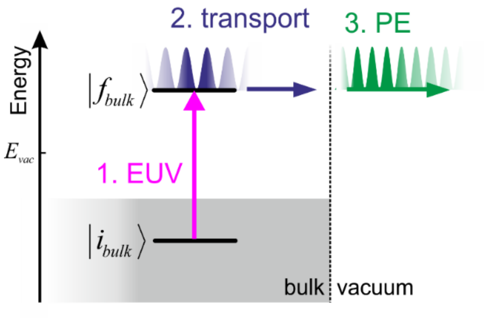


Optics Express 25, 22 (2017)

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

Relative photoemission times in solids



3 step model

W. E. Spicer, *Phys. Rev.* **112** (1958) 114.

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

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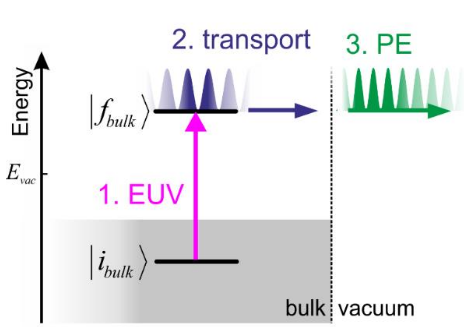

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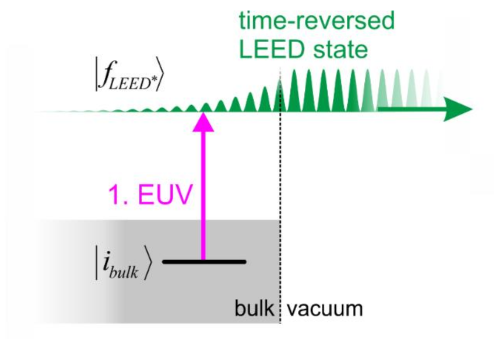
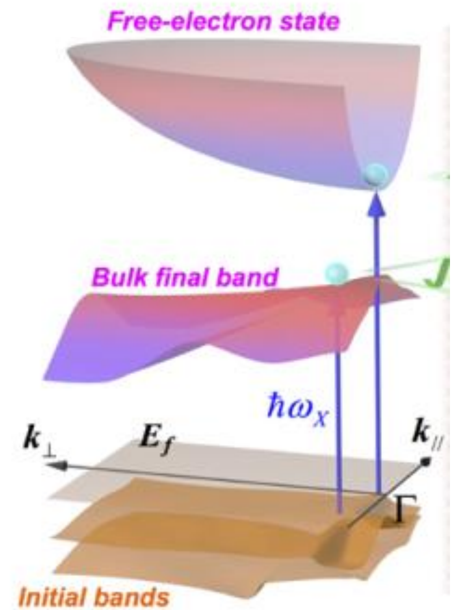


1 step model

G.D. Mahan, Phys. Rev. B **2** (1970) 4334.



Z. Tao, et al., *Science*. **353** (2016) 62

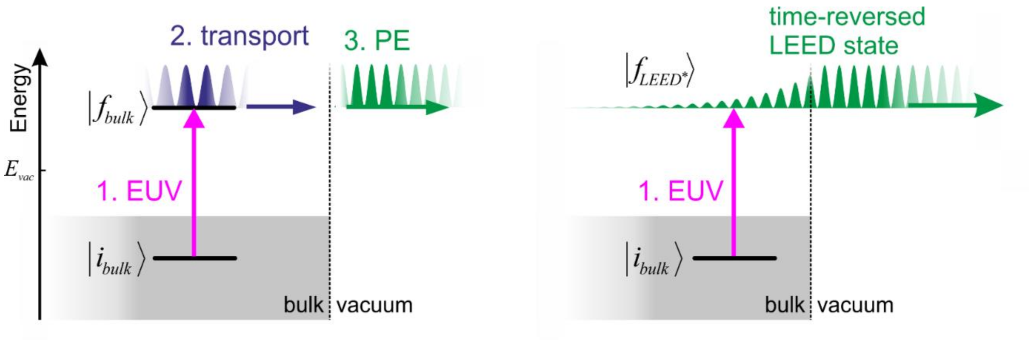
Open Questions:

- What is the role of transport?
- How does the final state influence the photoemission time?
- What about the orbital momentum of the electrons

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

Relative photoemission times in solids



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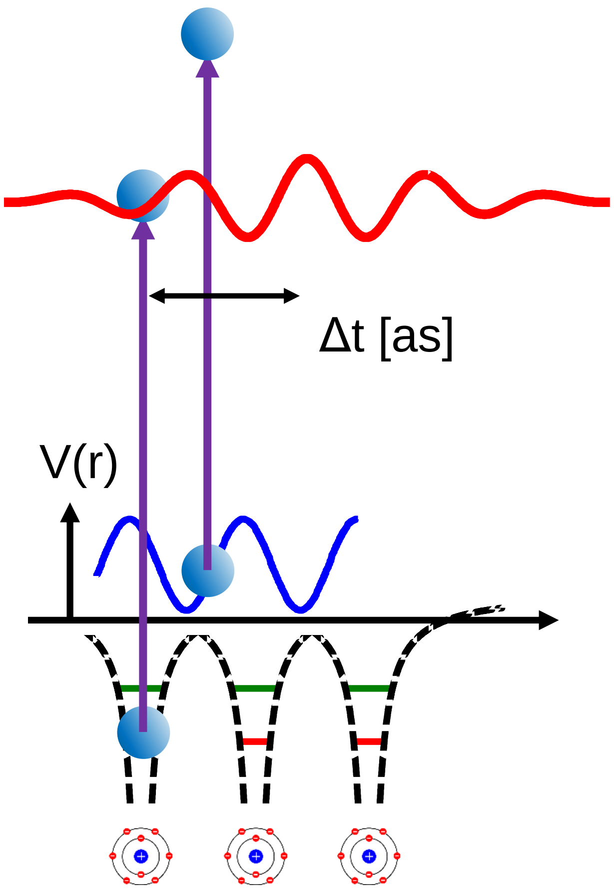
Attosecond streaking experiment in the condensed phase



Nature 449, 1029–1032 (2007)

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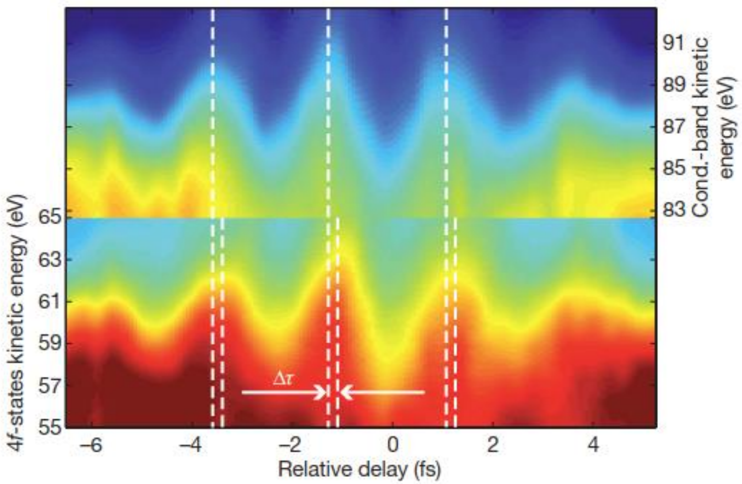
Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

4f and conduction-band spectrogram of single crystal tungsten



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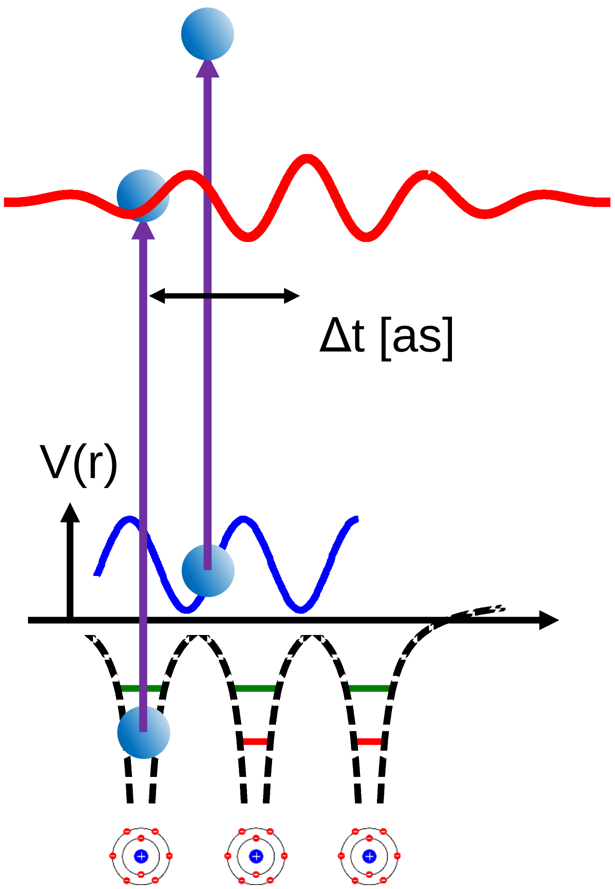

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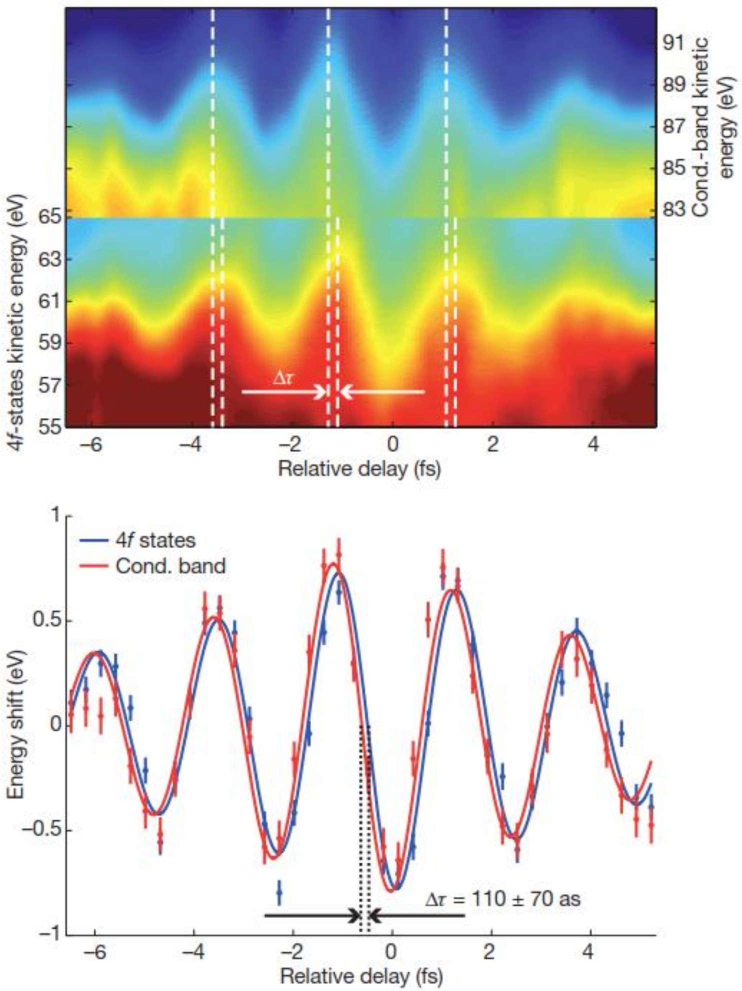

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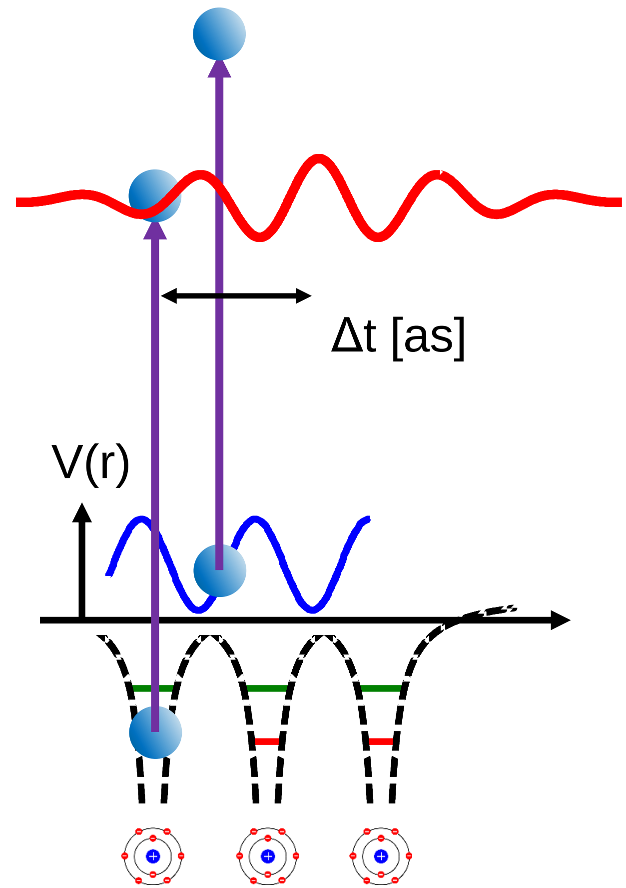

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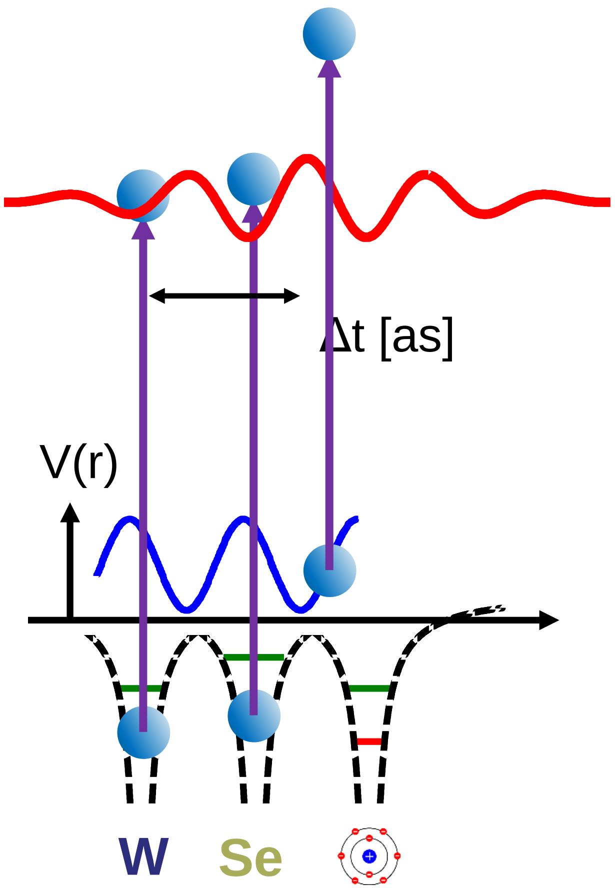
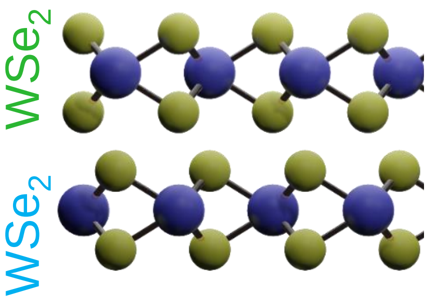
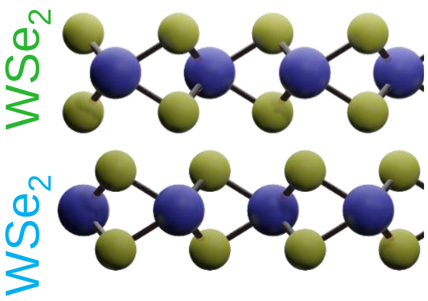


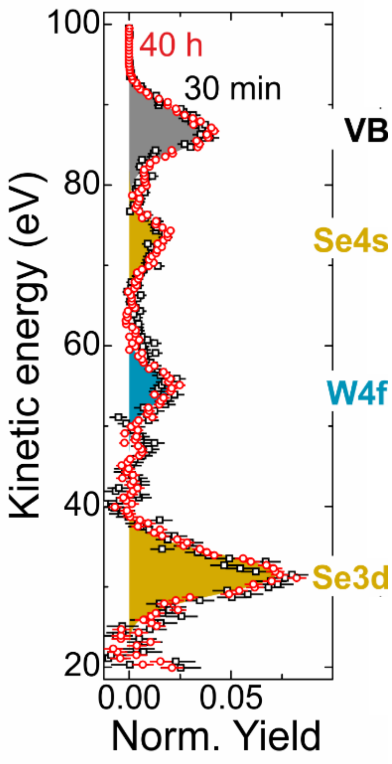


Science 357, 1274 (2017)

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy





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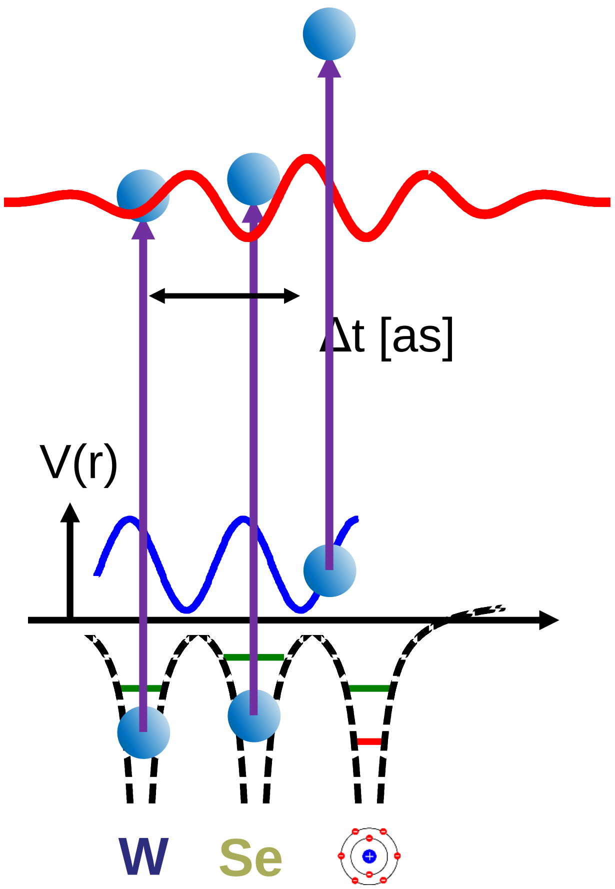

Science 357, 1274 (2017)

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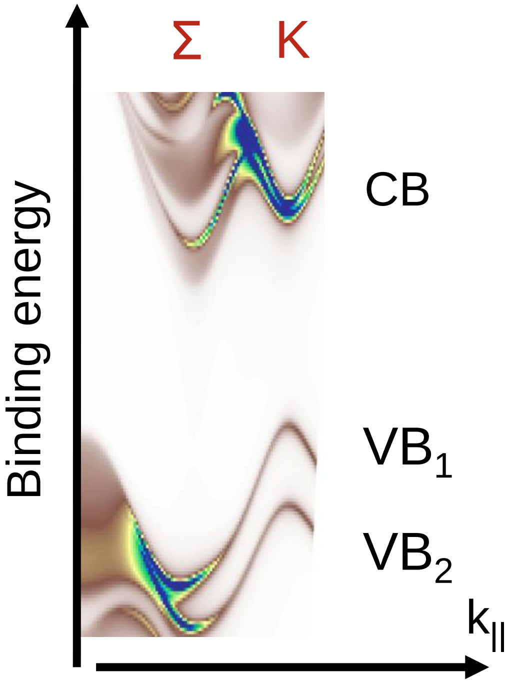
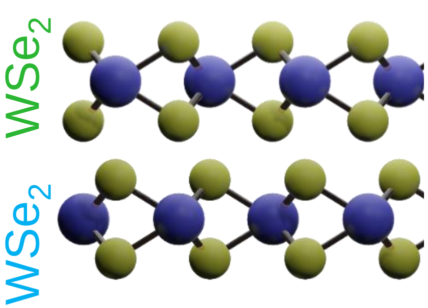
From fs to attosecond spectroscopy





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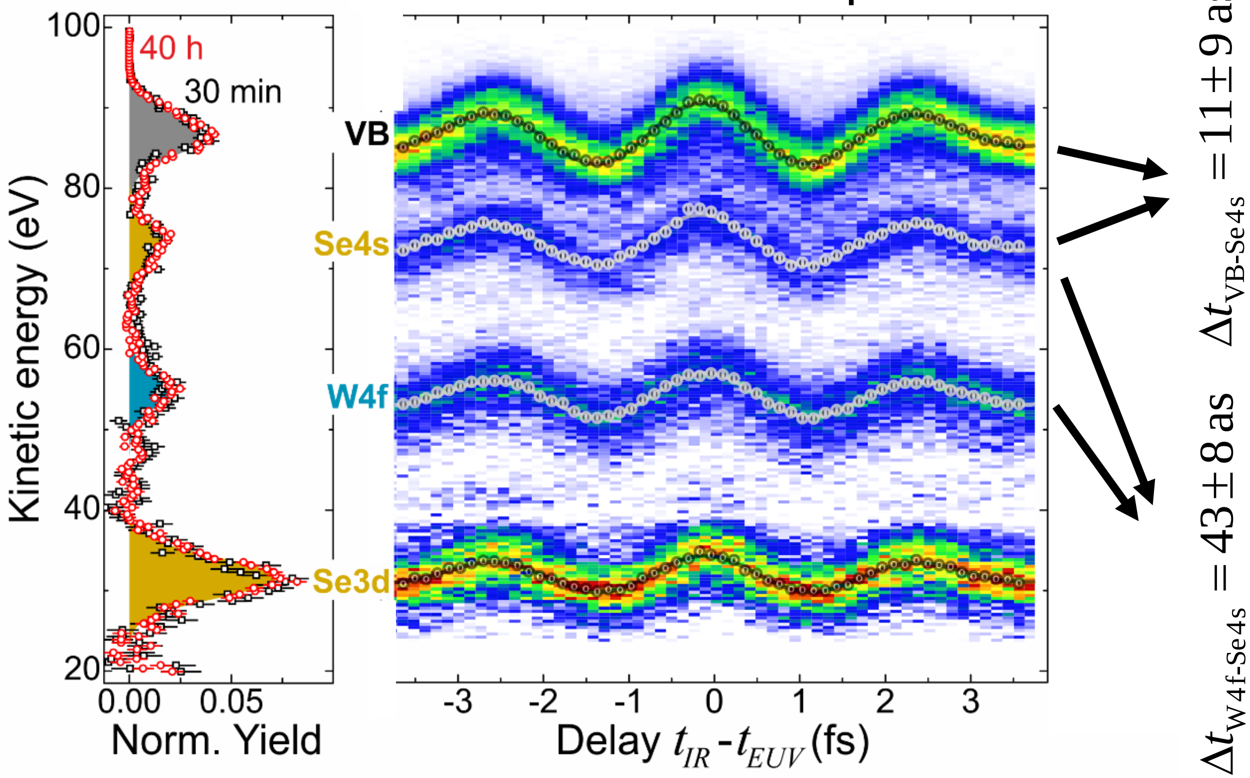
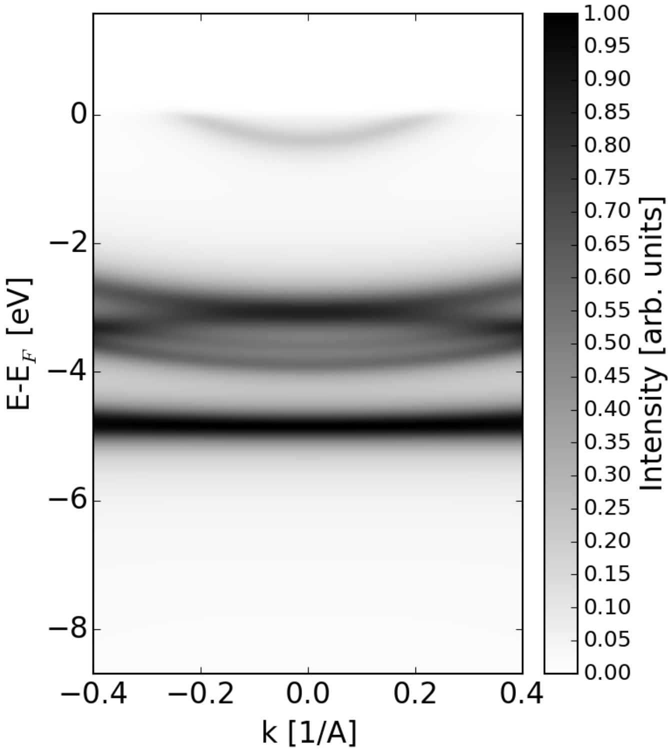


Science 357, 1274 (2017)

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

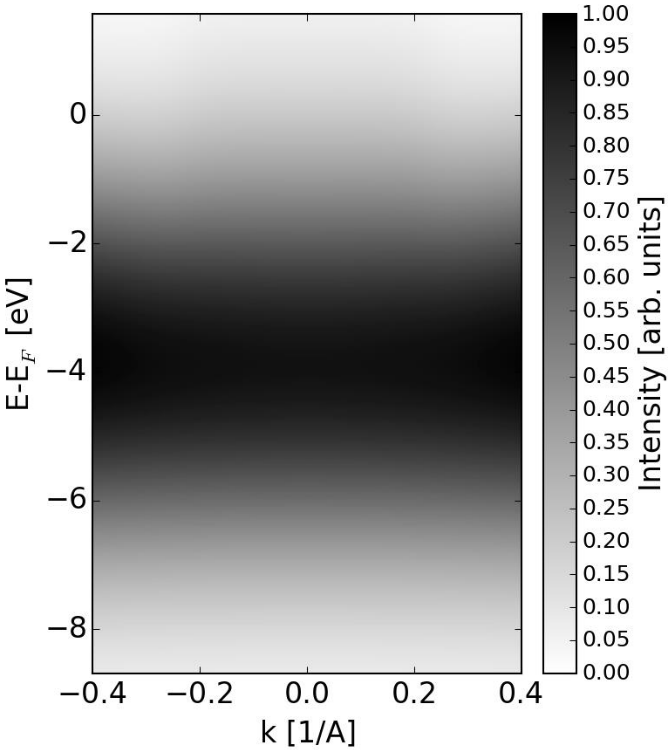
Continuous wave
light source



Loss of energy resolution

$$\Delta t \cdot \Delta E \approx h$$

Single attosecond
pulse

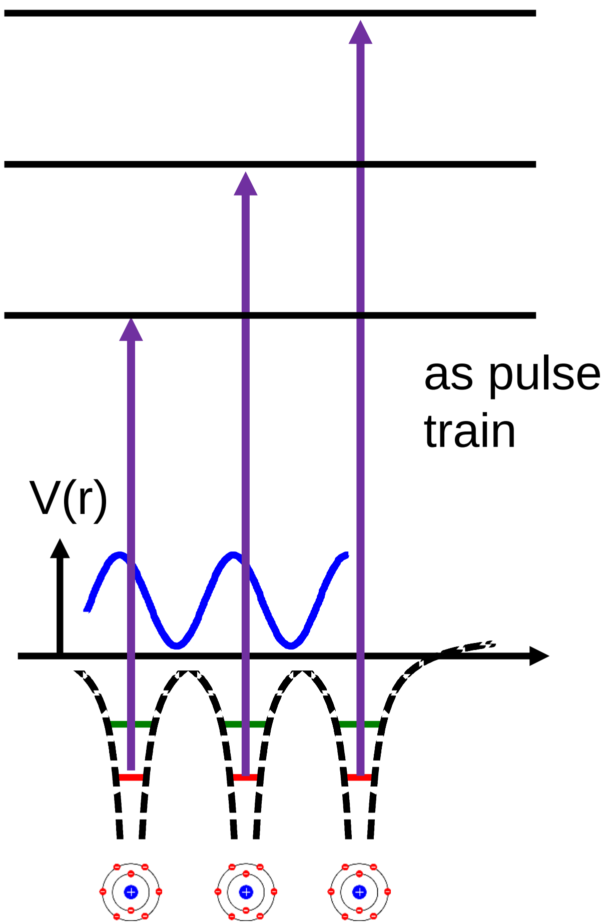


No easy way to conduced attosecond streaking experiment for valence band structure

Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

RABBITT experiment



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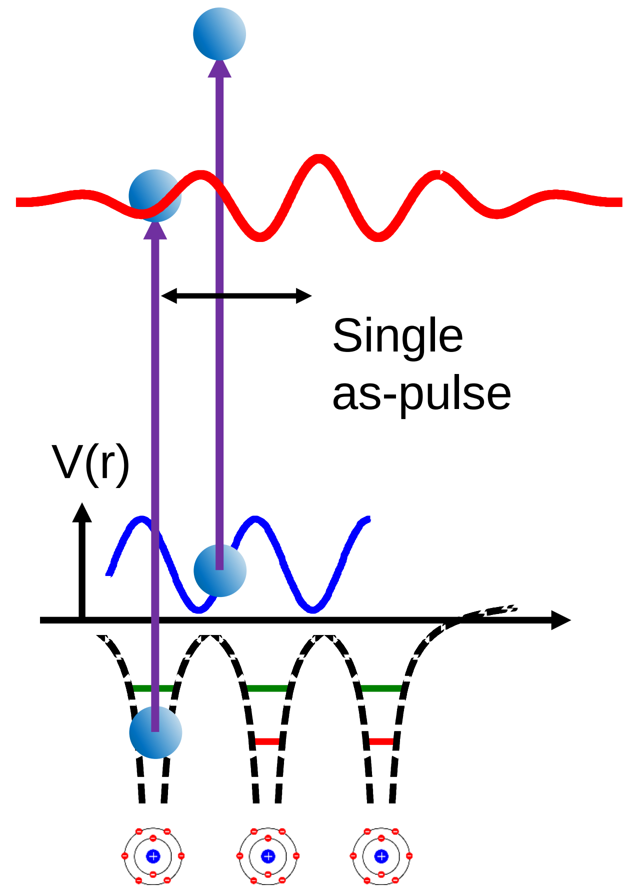
Attosecond streaking experiment in the condensed phase

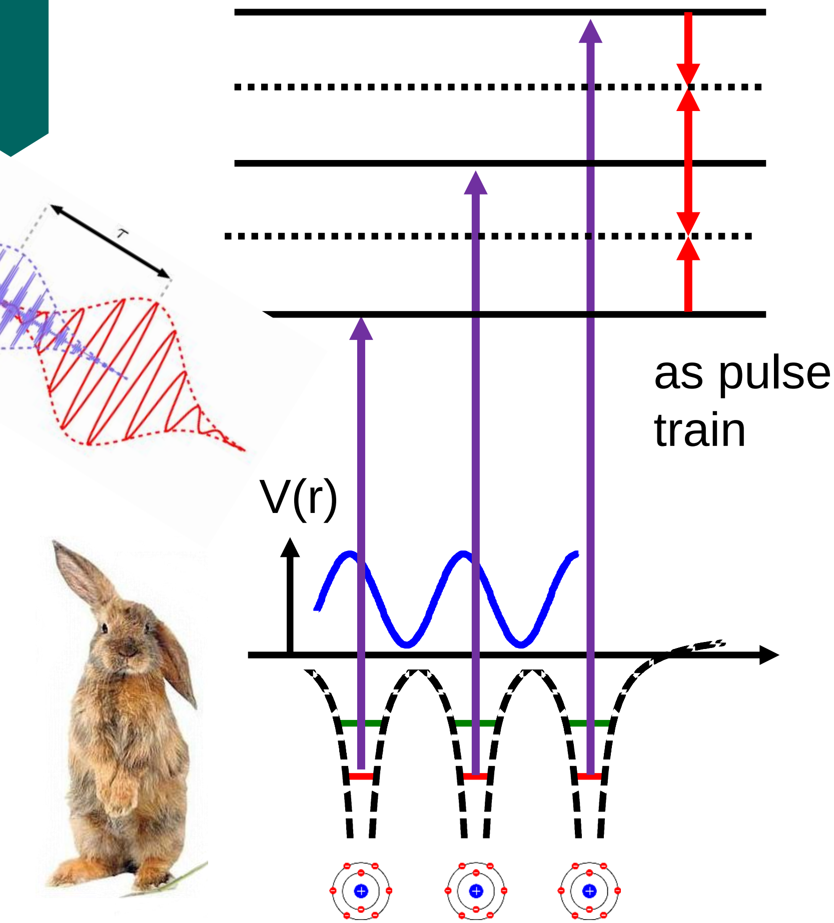


Nature 449, 1029–1032 (2007)

Towards Attosecond Surface Science Spectroscopy

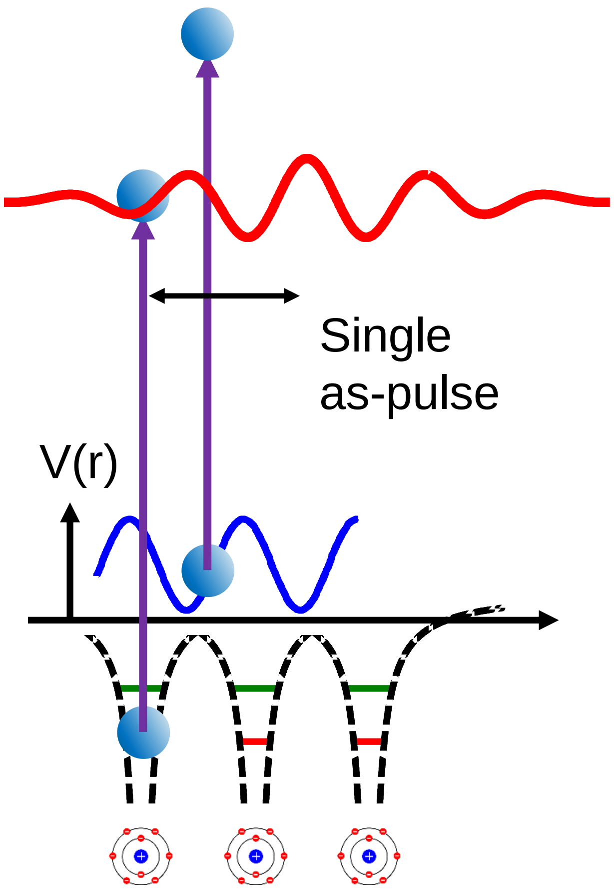
From fs to attosecond spectroscopy

RABBITT experiment



Science. **353** (2016) 62

Attosecond streaking experiment in the condensed phase

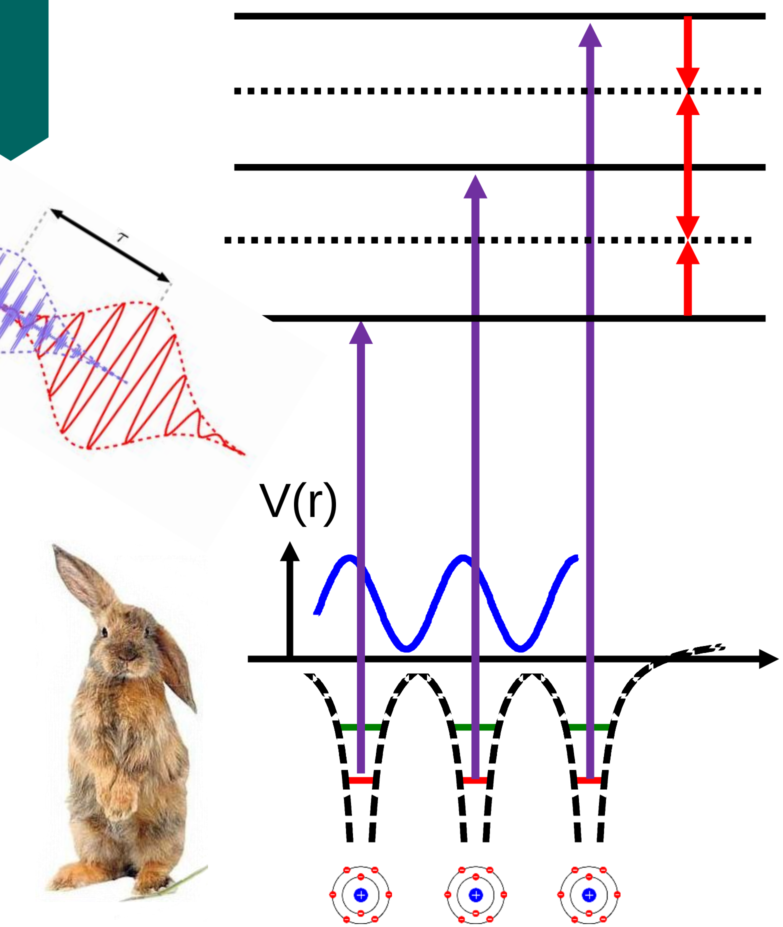


Nature 449, 1029–1032 (2007)

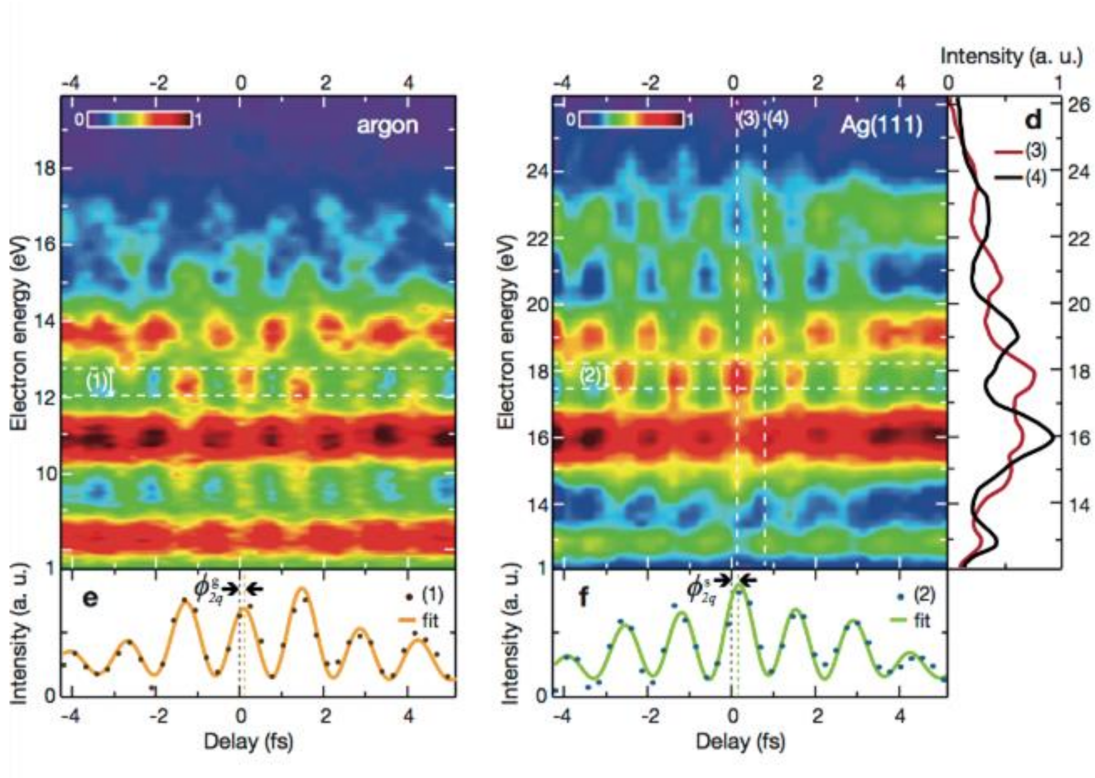
Towards Attosecond Surface Science Spectroscopy

From fs to attosecond spectroscopy

RABBITT experiment



Gas phase (argon) Solid State(silver)



Works only very well for isolated initial states

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R. Locher et al., Optica 2 (2015)

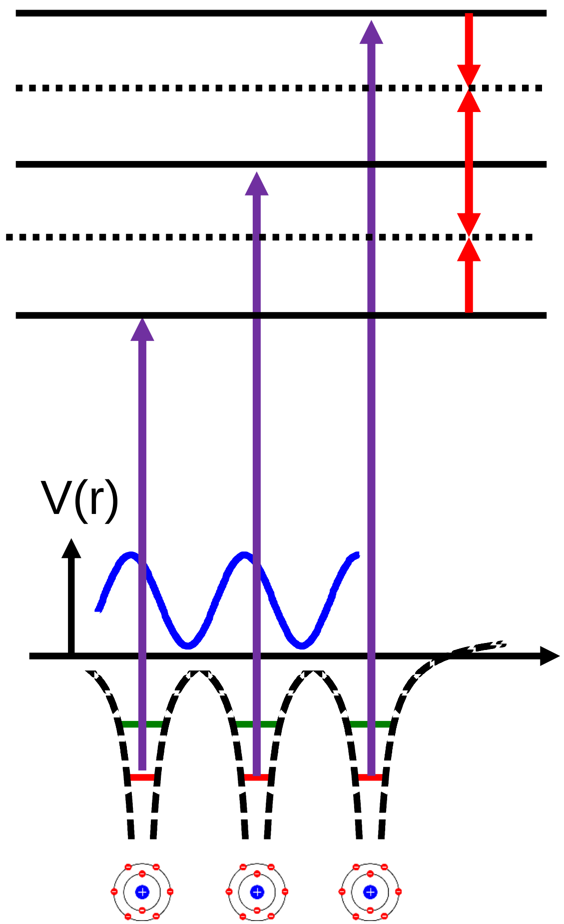
Towards Attosecond Surface Science Spectroscopy

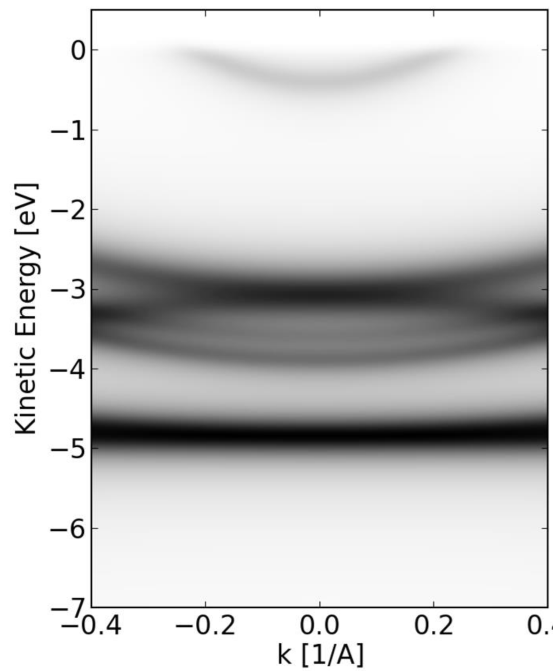
From fs to attosecond spectroscopy

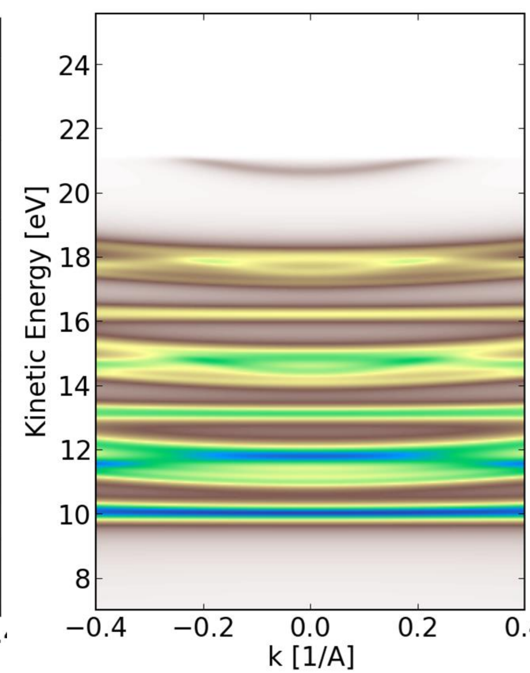
RABBITT experiment

Single HHG line

HHG spectrum







Is there no chance for RABBITT for valence states?

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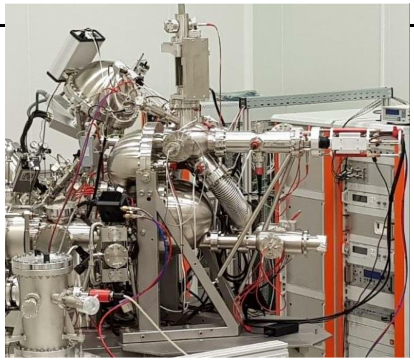
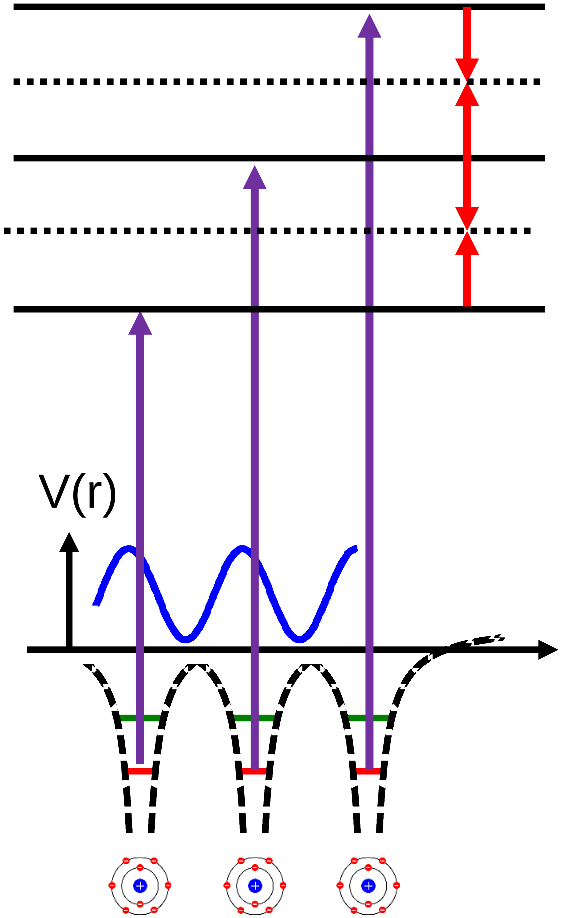
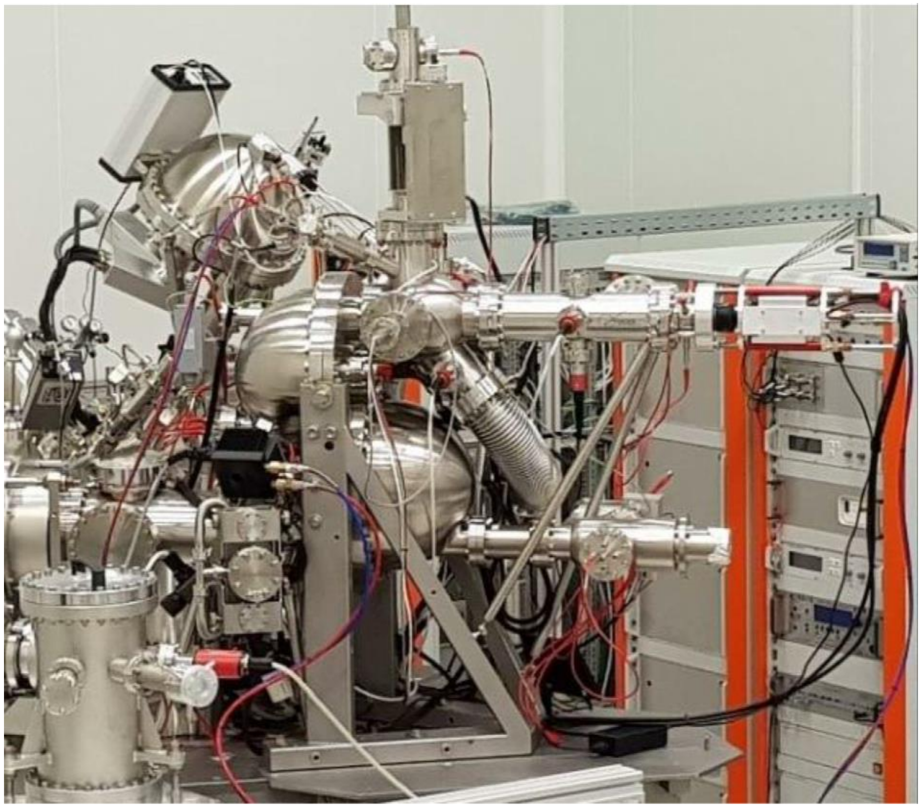
R. Locher et al., Optica 2 (2015)

Towards Attosecond Surface Science Spectroscopy

Attosecond Momentum Microscopy







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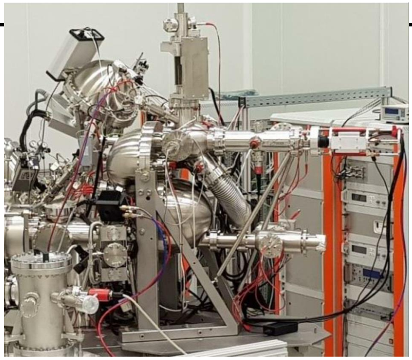
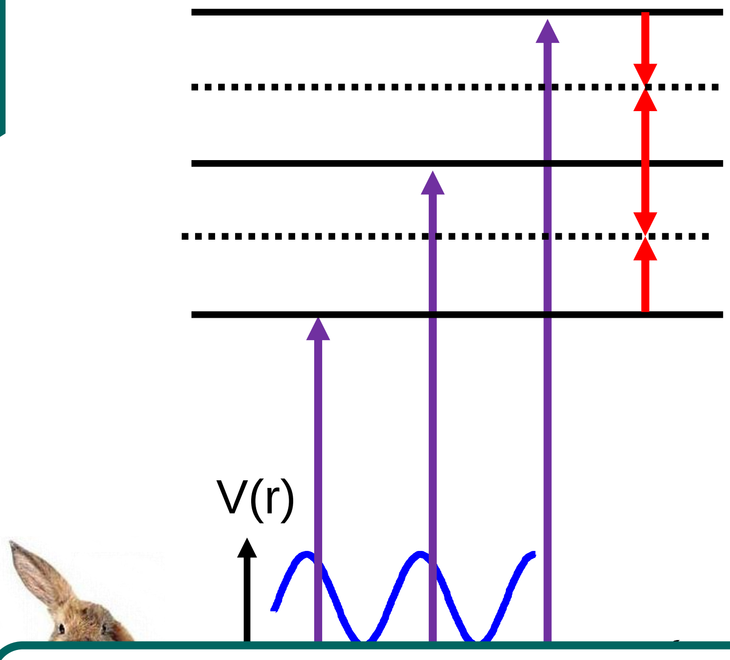
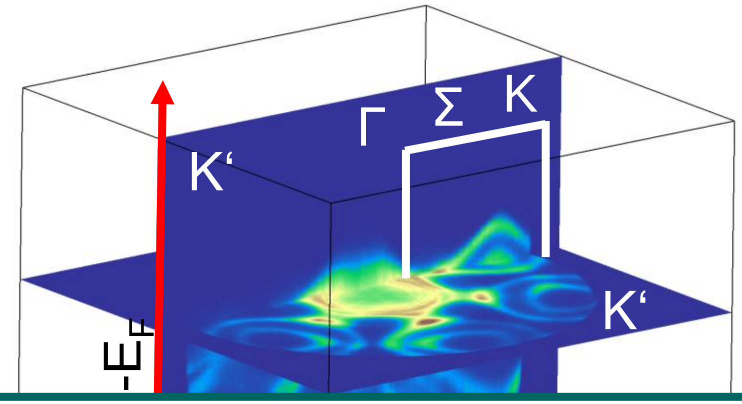
Towards Attosecond Surface Science Spectroscopy



Attosecond Momentum Microscopy



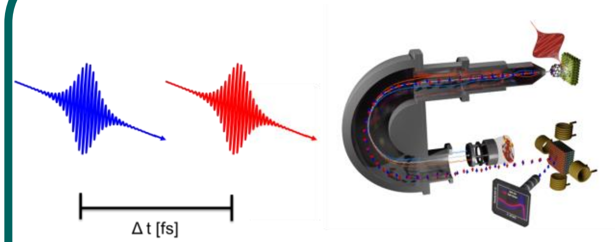
Full energy and momentum information



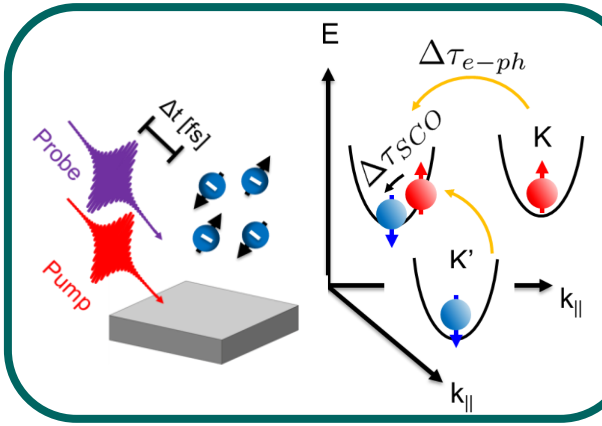
Disentangle photoemission yield of different valence band states and HHG lines by their characteristic signatures in

- Energy
- Momentum space
- Spin

Outline

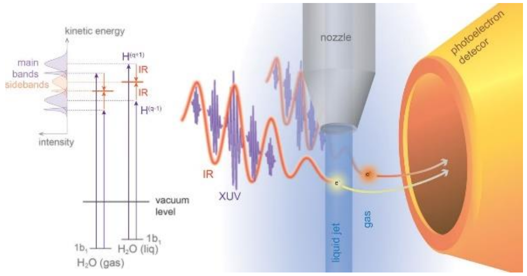


Ultrafast science in solids
… electrons in solids
… time-resolved photoemission spectroscopy



Monitoring….

…. the population dynamics of charge and spin carriers in momentum space
… the nature and spatial distribution of charge carriers in direct space
… interlayer charge separation across interfaces



Perspectives and challenges of attosecond surface science