Time-resolved experiments with HHG tabletop lasers compared to FEL and SR measurements

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School on Synchrotron and FEL Methods for Multidisciplinary Applications

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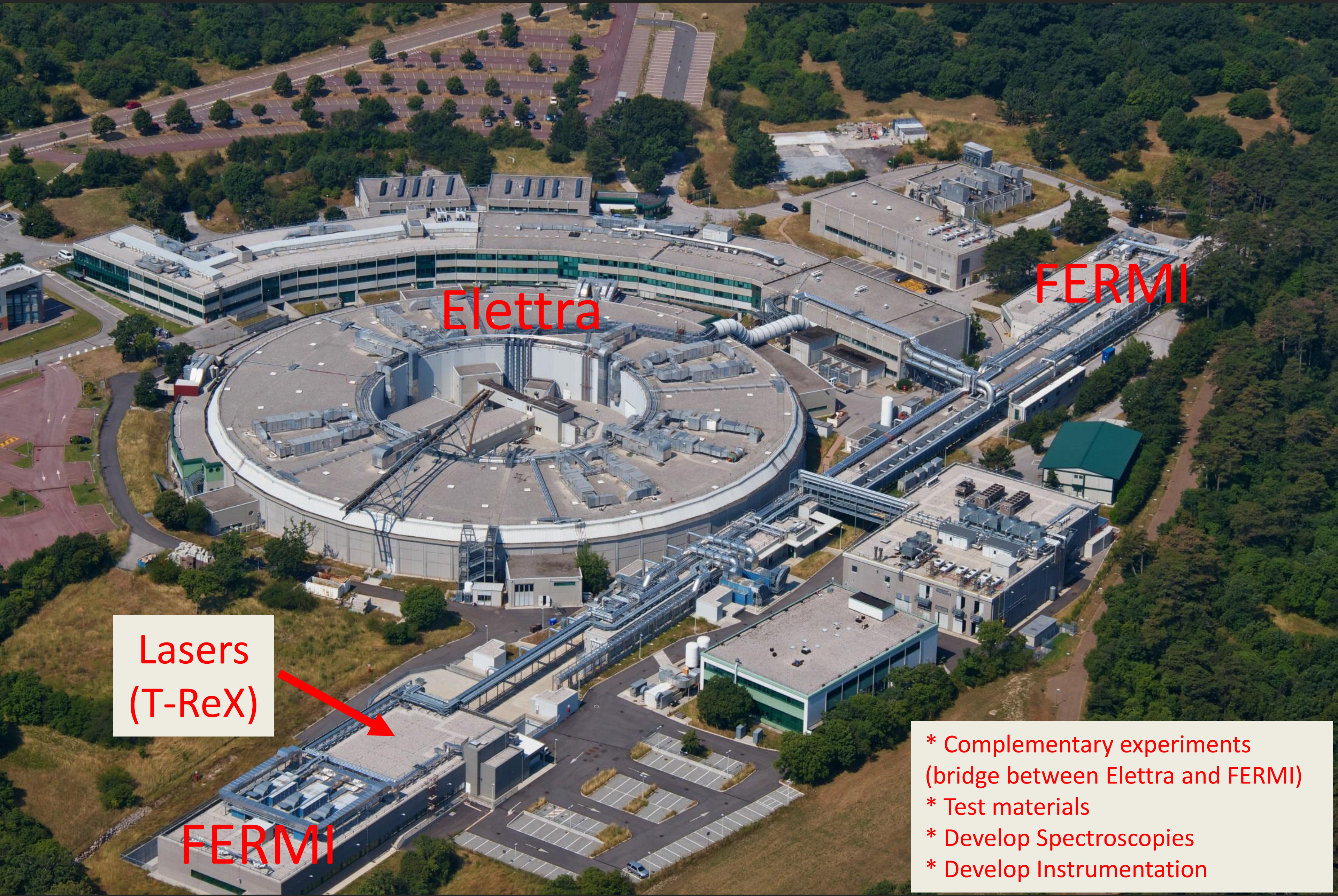
The Elettra-ST Campus: Synchrotron, FEL, Lasers

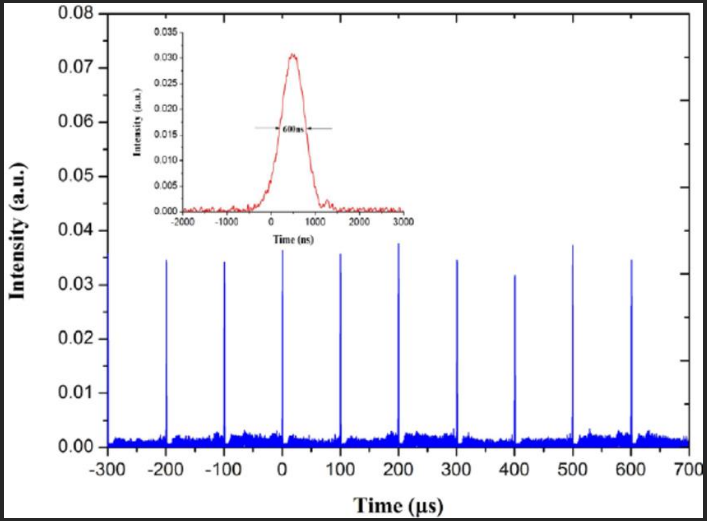


* Complementary experiments (bridge between Elettra and FERMI)
* Test materials
* Develop Spectroscopies
* Develop Instrumentation

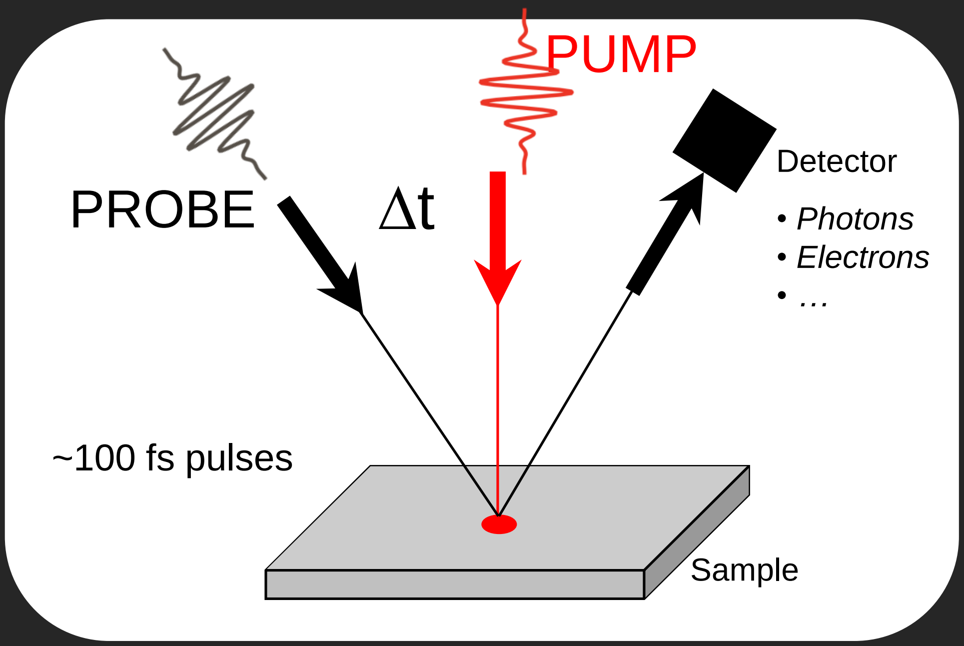
Synchrotron, FEL, Lasers: basic parameters

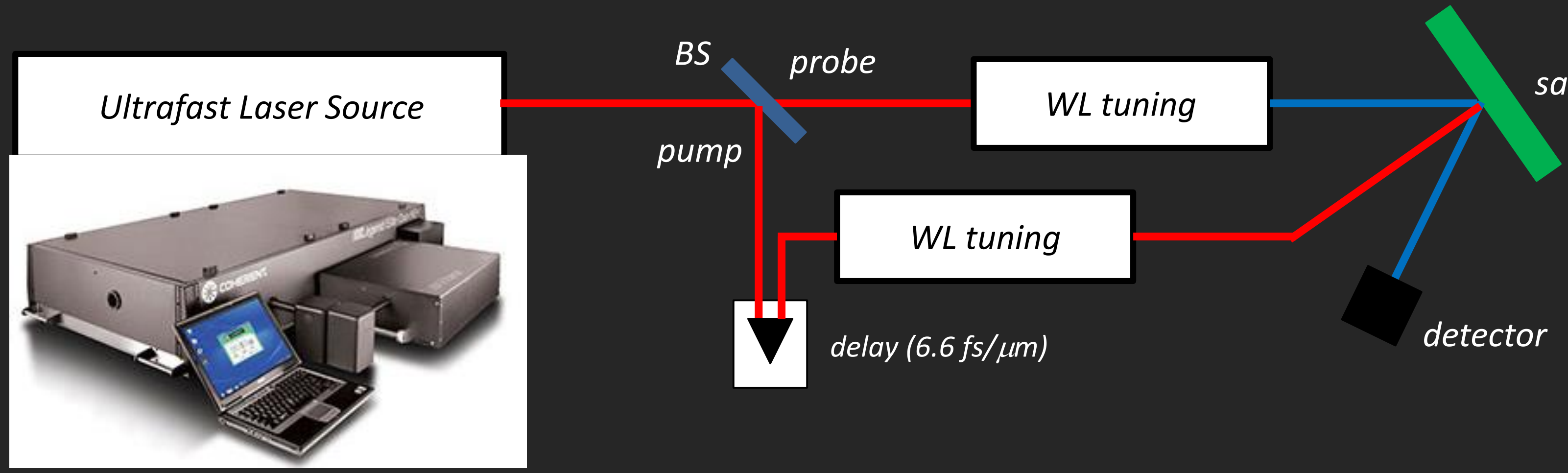
	Synchrotron	FEL	Laser
Energy range (eV)	8-10000	12-1000	0-70
Energy Bandwidth	<1 meV	20-40 meV	1-1000 meV
Pulse Duration	≈100 ps	20-50 fs	5-300 fs
Repetition Rate	500 MHz	50 Hz (kHz)	kHz-MHz
Brightness	10^{12} ph/s	10^{20} ph/s	10^{16} ph/s
Polarization	tunable	tunable	tunable

$$\Delta E \Delta \tau \geq \frac{\hbar}{2}$$



Time-resolved experiments

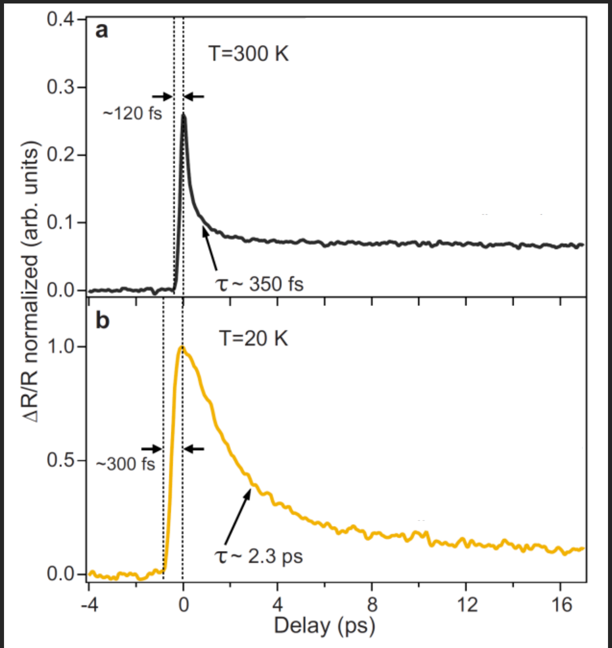




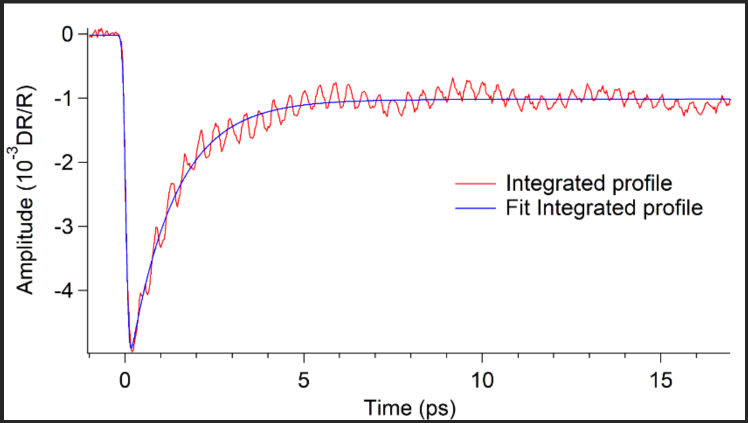
Time-resolved experiments

In principle, time resolved experiments can be performed with every pulsed source, but different timescales (and underlying physics) can be accessed.

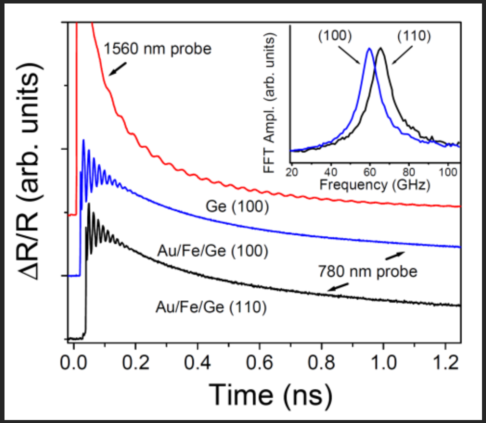
Dynamics across phase transition



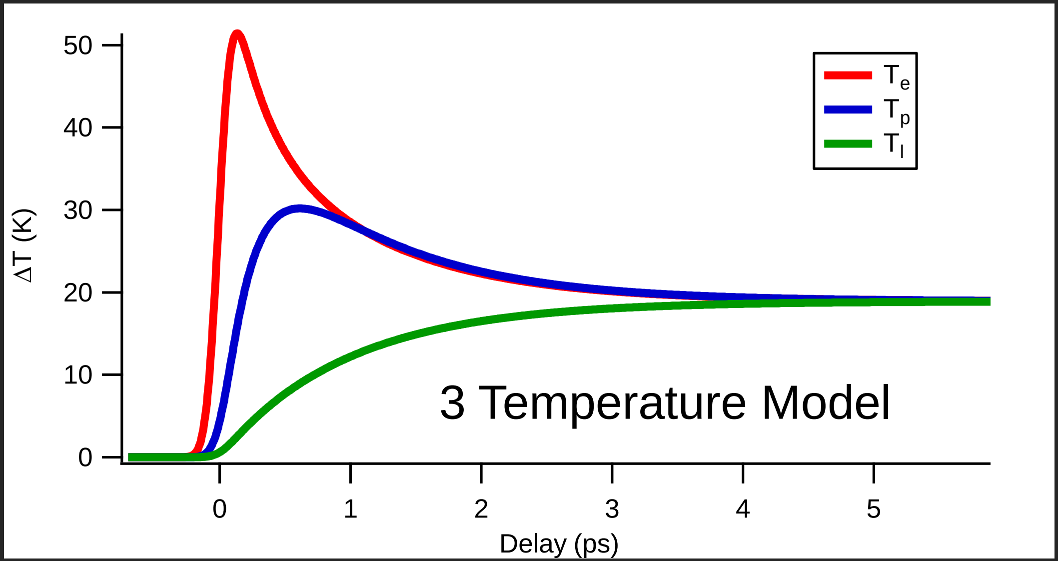
Coherent Phonon



Strain Waves



TR SPECTROSCOPIC experiments reveal the ground state of a material by its dynamics!



Time-resolved experiments: laser sources

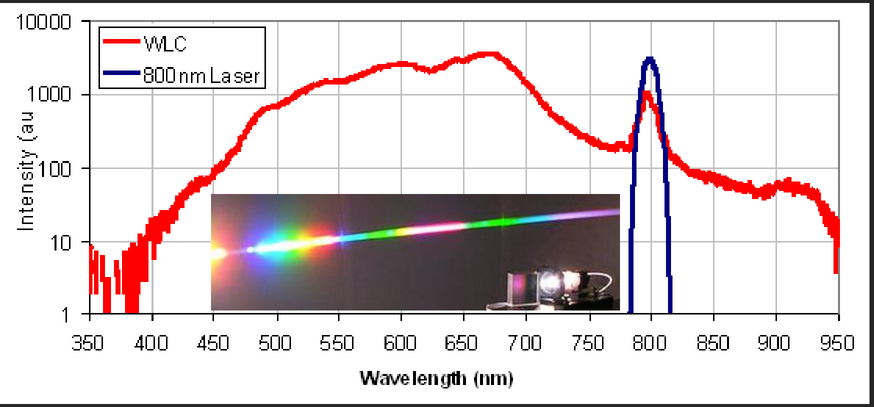
	Multipass	Regenerative	Oscillators
Energy/pulse	1-10 mJ	5-500 uJ	10 nJ
Repetition rate	1-10 kHz	50-500 kHz	50-100 MHz
	'Probe' limit	ok	'Pump' limit

When we consider a laser, the *number of pulses per second* and their *energy* are inversely proportional (their product is the average power, which is of the order 1-40 W)

→ Several nonlinear phenomena are accessible only with 0.5 -1 mJ/pulse:

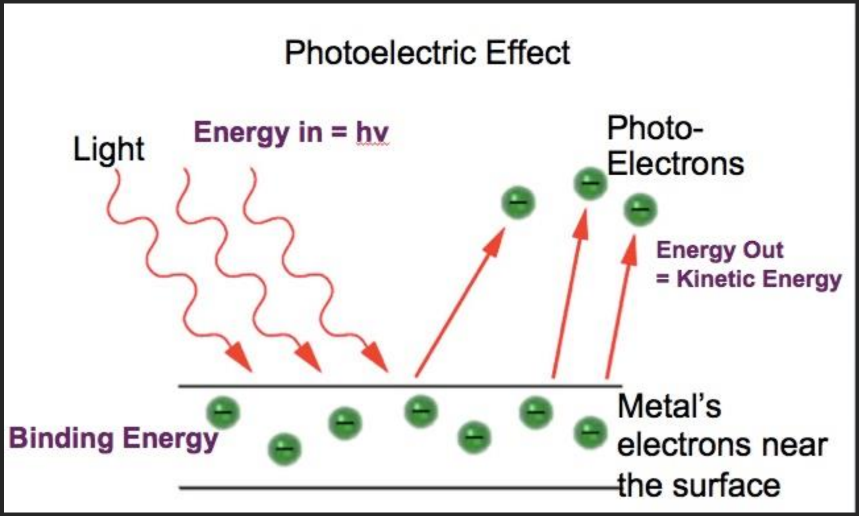
- HHG
- Harmonics in gases

→ Supercontinuum generation (used in OPAs) has a threshold at 1 uJ/pulse

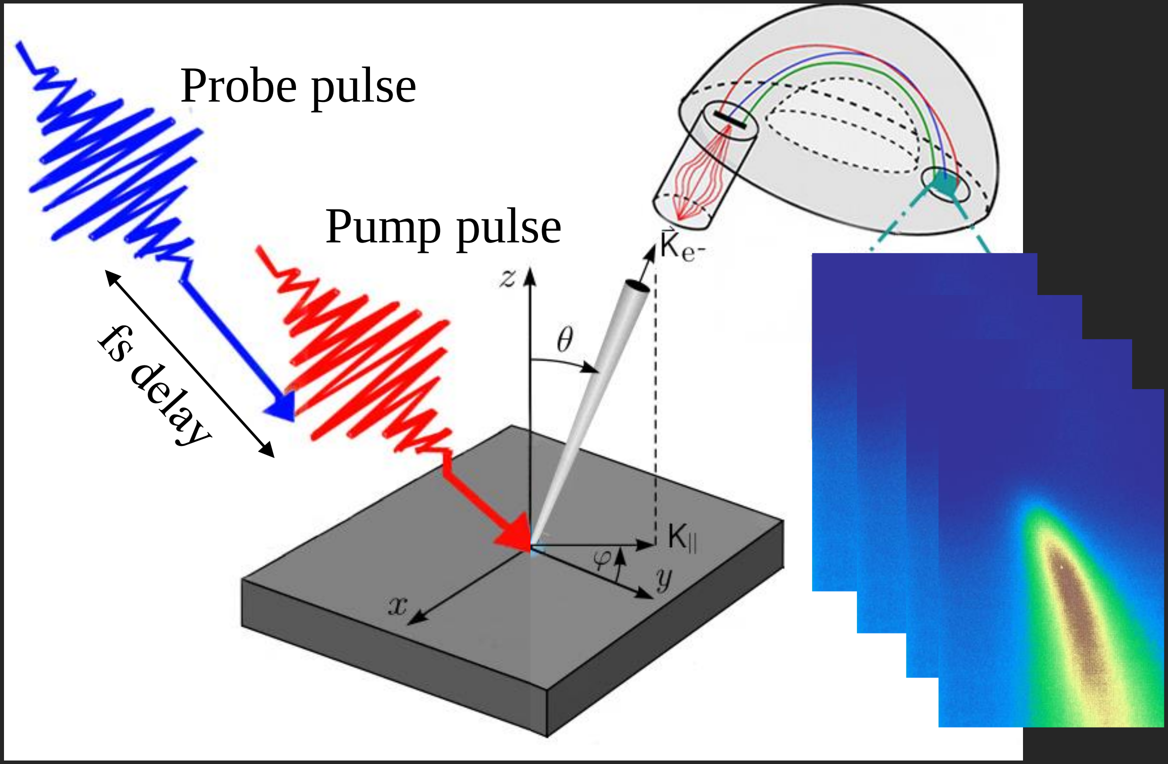


Photoemission and Time-resolved photoemission (ARPES, TR-ARPES)

Photoelectric Effect:
photon-in, electron-out



For non-equilibrium experiments, a pump pulse is added. This should not produce photoelectrons



Time-resolved photoemission (TR-ARPES): transient effects

Photoemission Intensity:

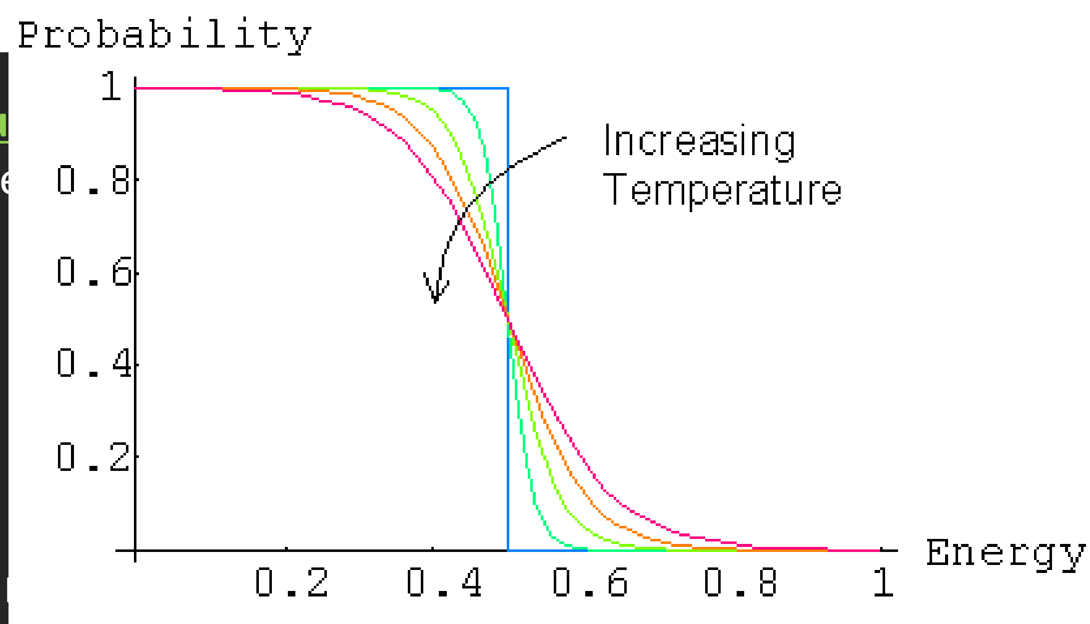
$$I\,(E, \mathbf{k}) = I_0(E, \mathbf{k}, pol) \cdot A\,(E, \mathbf{k}) \cdot f\,(E)$$

Matrix element: contains information on the geometry of the experiment and on the light polarization

One particle spectral function: describes the band structure

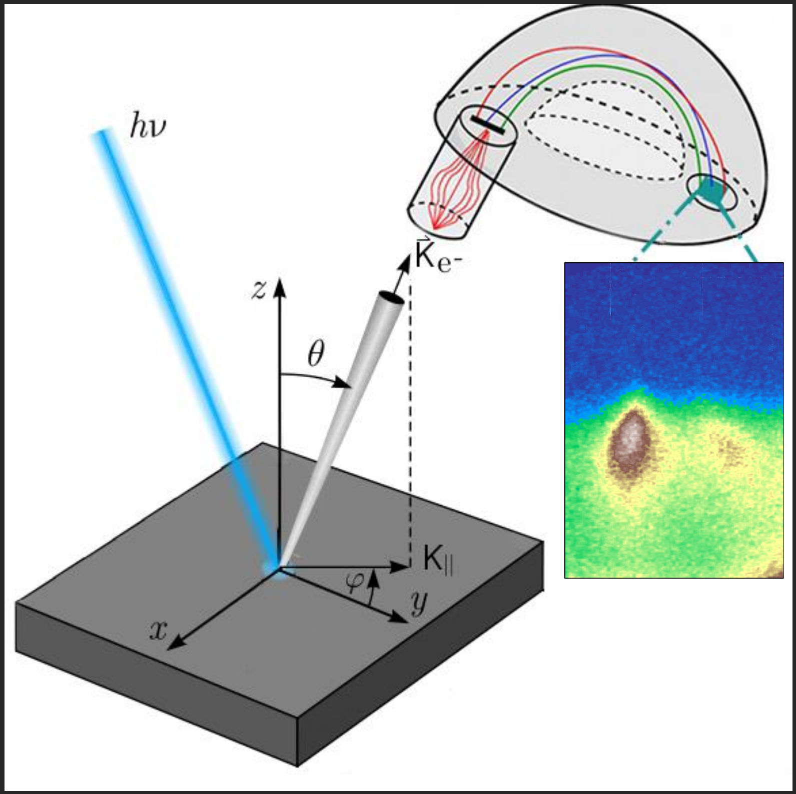
$A(E,k) \rightarrow A(E,k,\tau)$ with $\Sigma'(\tau)$ and $\Sigma''(\tau)$

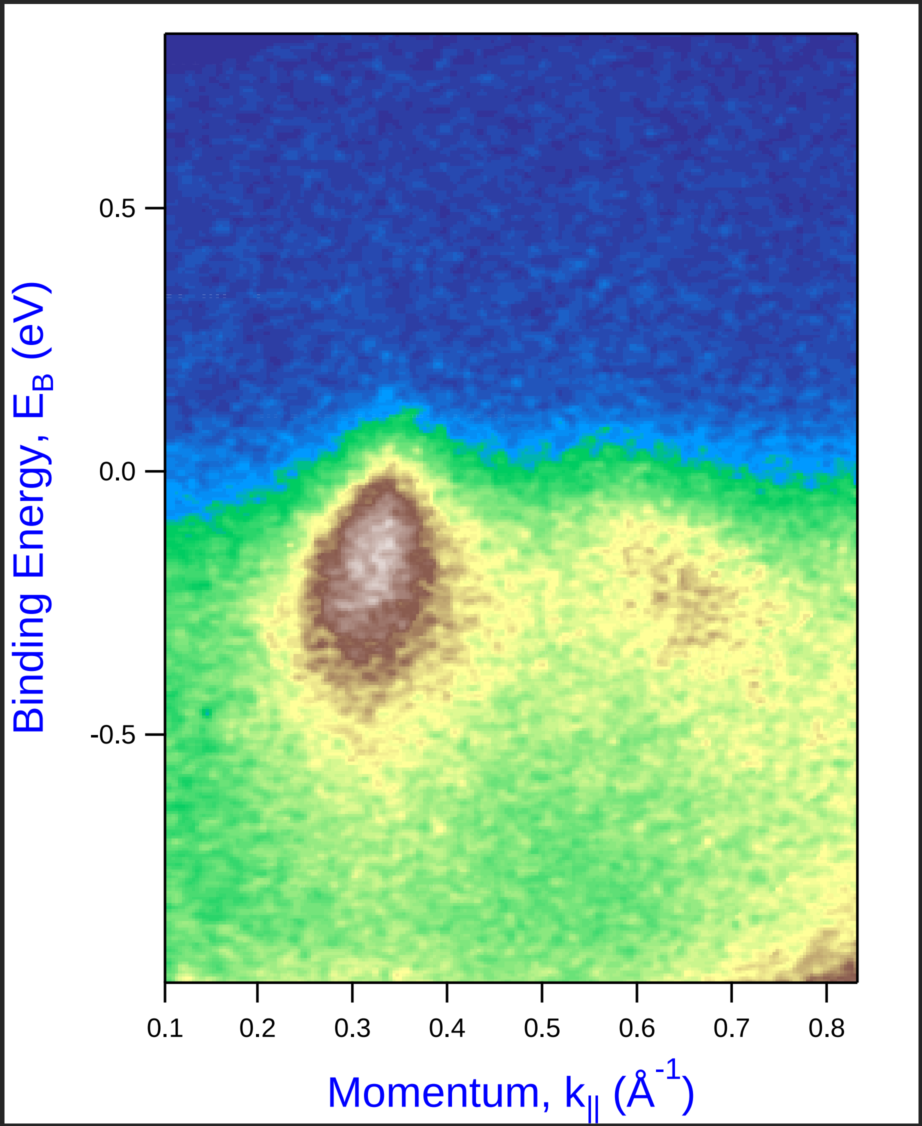
Fermi Dirac distribu… , the occupation (population) of the e…



The excitation ca… ndstructure itself

Time and Angular Resolved Photoelectron Spectroscopy (TR-ARPES)





Conservation Laws

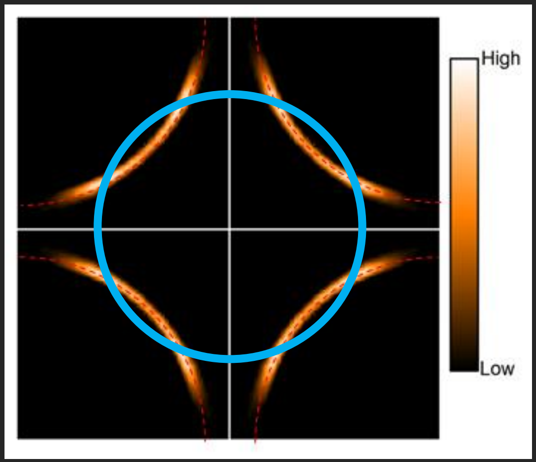
$$E_{kin} = h\nu - \phi - E_B$$

$$\vec{k}_{||} = \vec{K}_{||} = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\theta$$

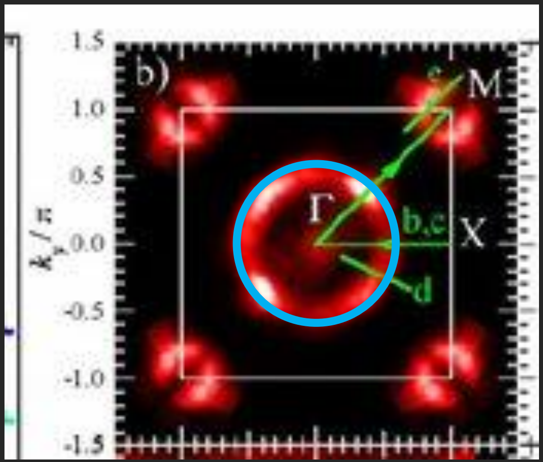
Photoemission with ultrashort pulses: 1) photon energy

We just saw that: $h\nu > \phi$. Usually, $\phi \approx 4$-5 eV. This sets a limit in TR experiments

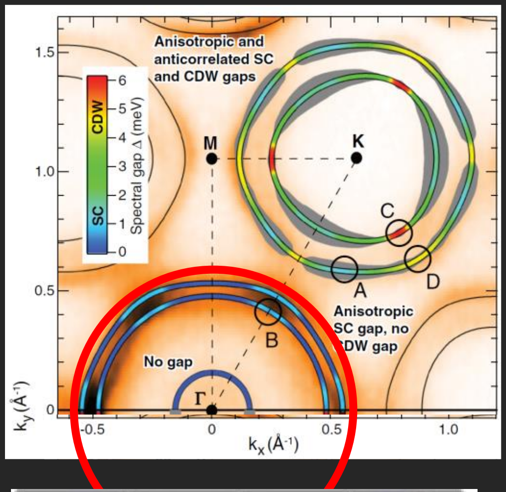
$Bi_2Sr_2CaCu_2O_8$

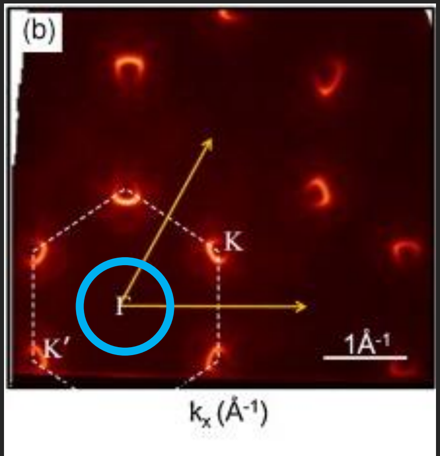


NdFeAsOF

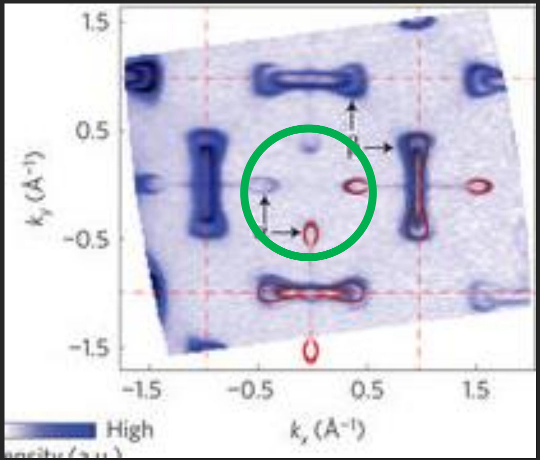


$NbSe_2$

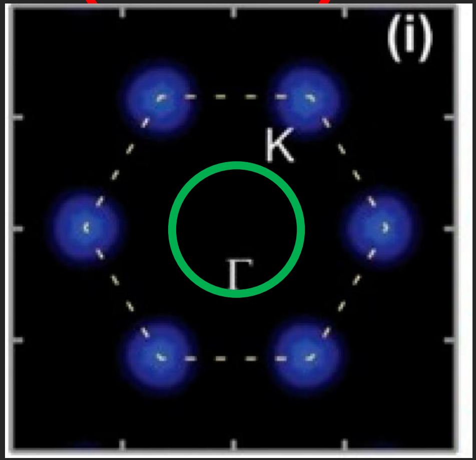




Graphene



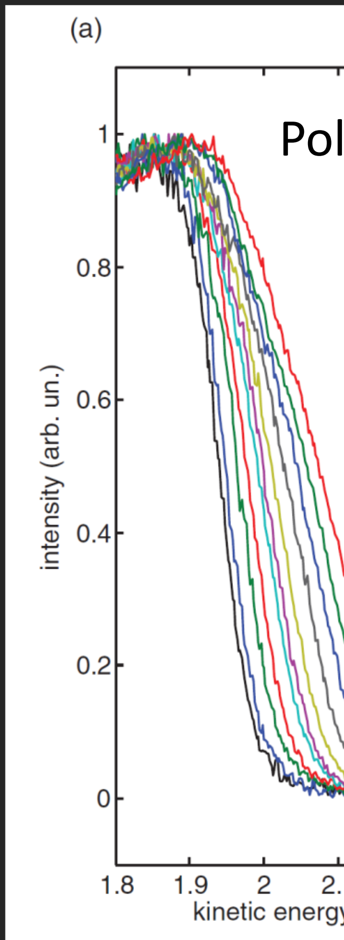
NbP

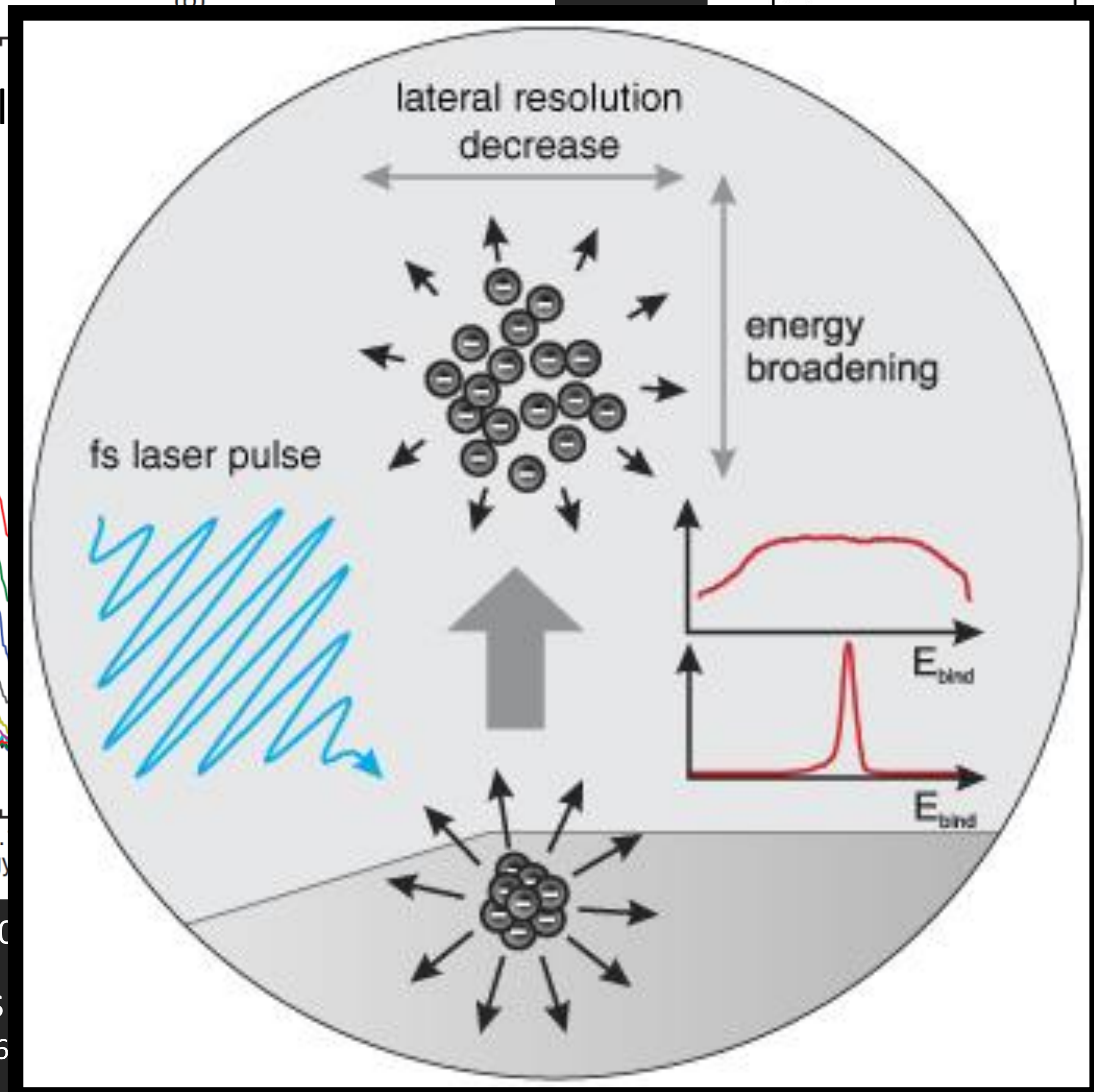


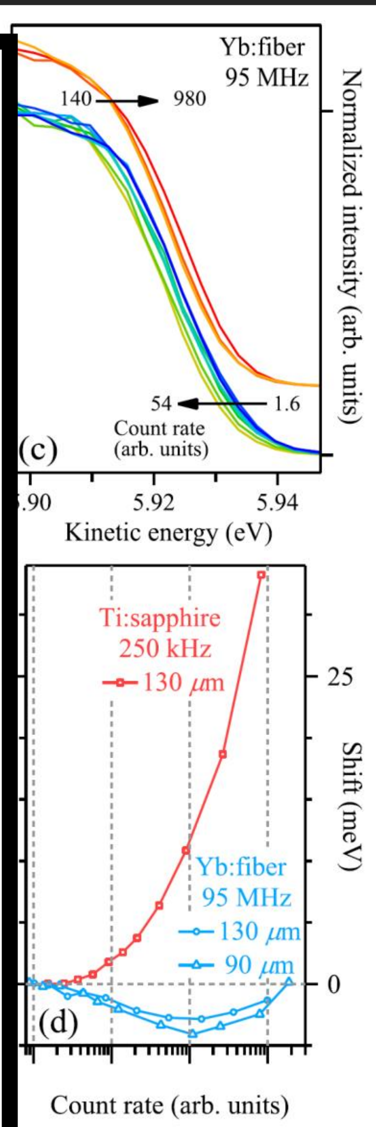
MoS2

Colored circles are drawn for $h\nu = 6.2$ eV, the most typical energy for TR-ARPES

Photoemission with ultrashort pulses: 2) space charge







J. Faure et al., RSI **83**, (

Fermi edge meas
N=2x10^5 to 6x10^6

Y. Ishida et al., RSI **87**, 123902 (2016)

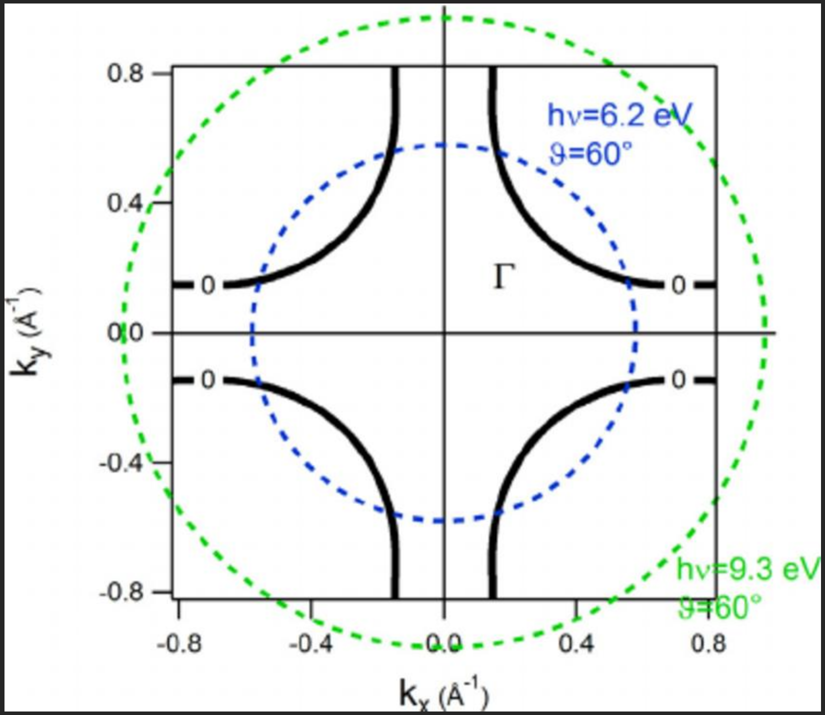
THUMB RULE: *maximum 1 e- per pulse*: <u>count rate problem with low rep. rate lasers</u>

Toward the TR-ARPES Mapping of the Entire Fermi surface of materials

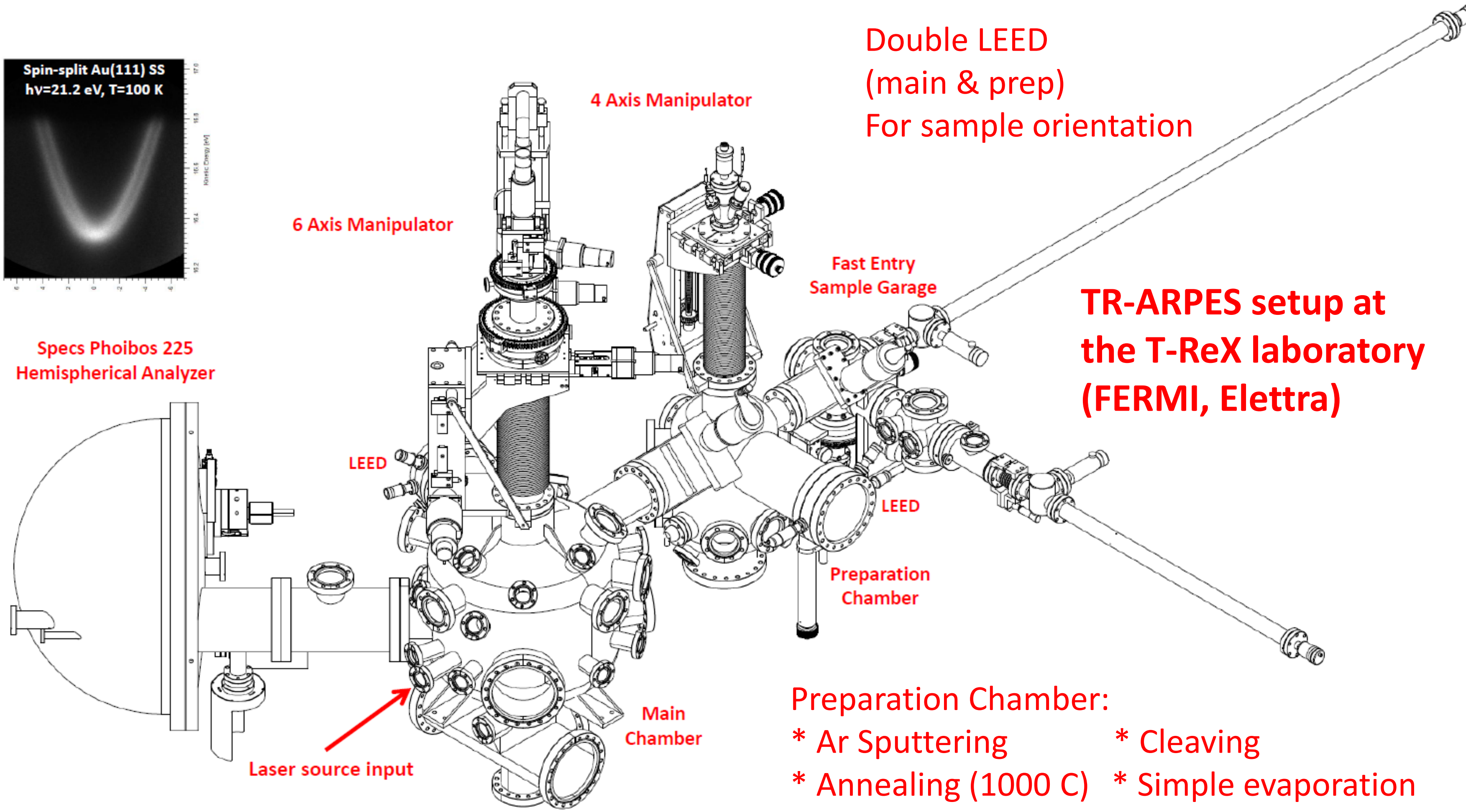
QUEST: probe large portions of binding energy and momentum space, with high statistics.

* High rep. rate
* Low space charge
* High energy and momentum resolution
* High S/N ratio, to reveal trends as a function of T, doping, fluence
* Fully tunable polarization state (linear horizontal and vertical, circular left and right).

At >9 eV and ≈60° emission angle, the entire BZ of cuprates can be accessed!



The T-ReX TR-ARPES Endstation



Double LEED
(main & prep)
For sample orientation

TR-ARPES setup at the T-ReX laboratory (FERMI, Elettra)

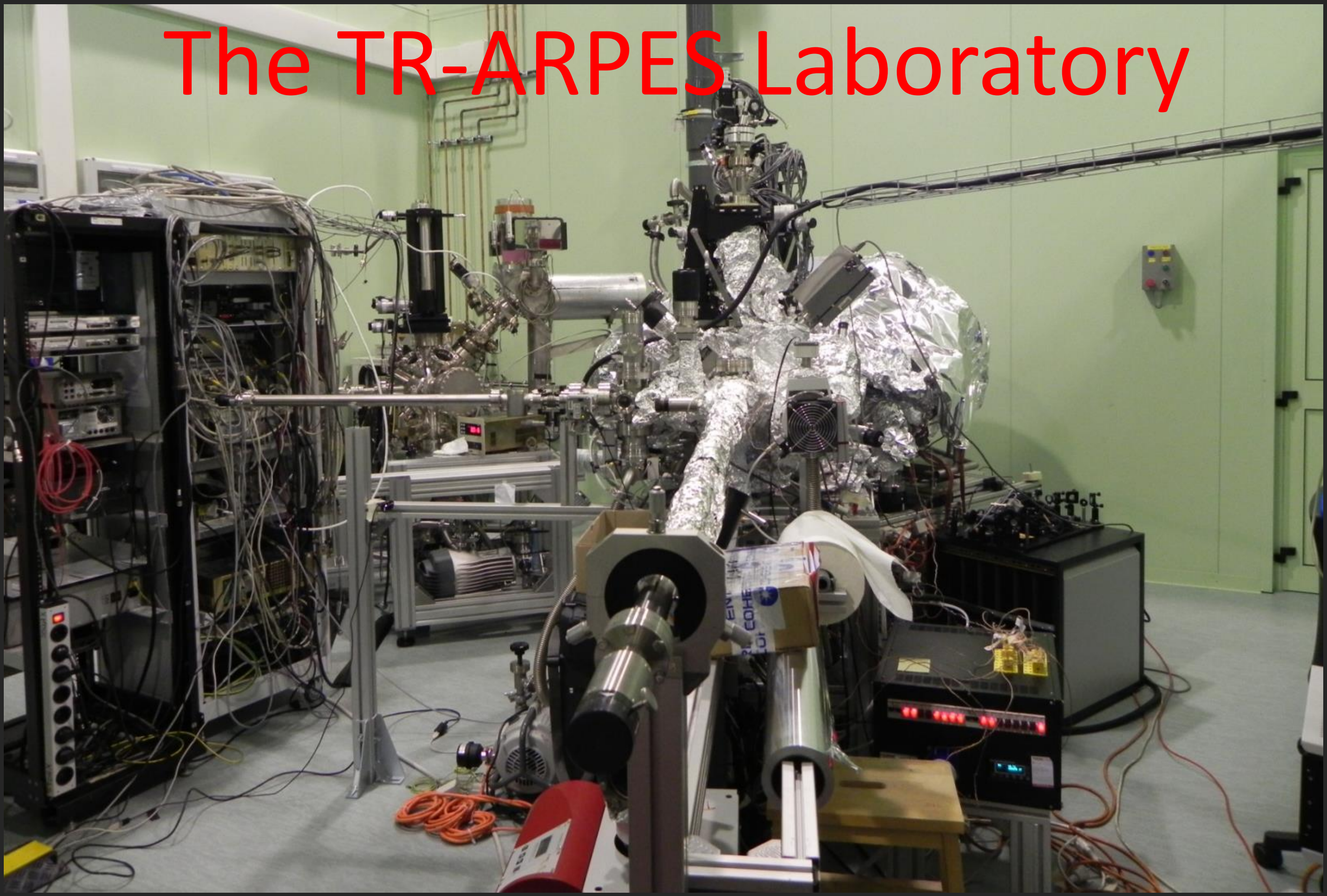
Preparation Chamber:
* Ar Sputtering
* Annealing (1000 C)
* Cleaving
* Simple evaporation

Sources (equilibrium):
* UV Source (21.2 eV)
* X-Ray Source
* e-gun

Laser (non-equilibrium):
* Probe: 6.2 eV, 8.5 eV, 9.3 eV
1.5, 3.1, 4.5 eV and OPA for NL-PES
in situ TR-Optics
* Pump: 1.5 eV, 3.1 eV, 0.5-1.2 eV

SPECS Phoibos 225
Photoelectron Analyzer
~20 meV energy resolution
±20° acceptance (‘SWAM’)

The TR-ARPES Laboratory



MAIN CHAMBER

- SPECS Phoibos 225 hemis. analyzer (angular res. 0.1°, energy res. 20 meV)
- DelayLine Detector (DLD), low noise
- Six DOF cryo-manipulator (T>30 K)
- $5*10^{-11}$ mbar
- LEED
- He-I lamp, 21.2 eV

PREPARATION CHAMBER

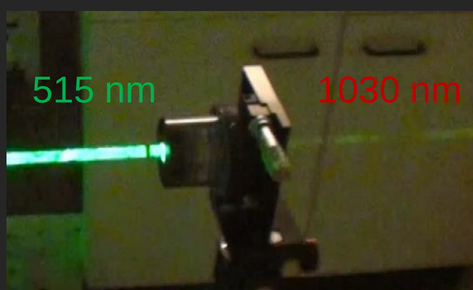
- Sputtering and annealing (T<800°C)
- LEED
- RGA
- Cleaving in UHV via scotch tape or pin
- $<10^{-10}$ mbar
- Fast entry (10^{-7} mbar in 30')
- Sample growth possible

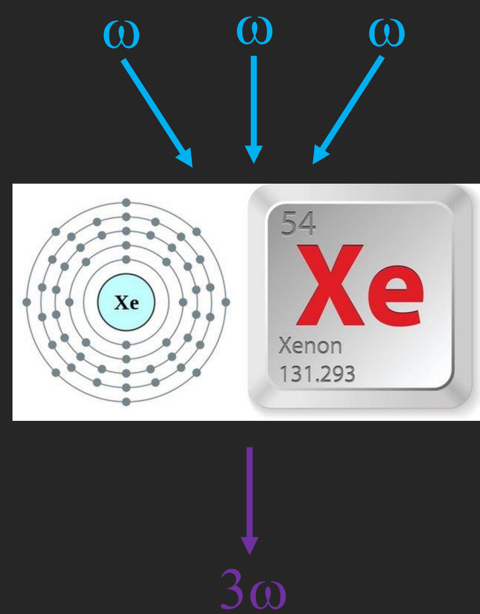
LASER SOURCES

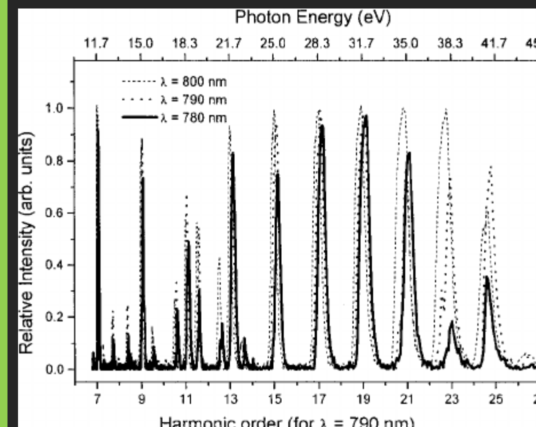
- FHG at 6.2 eV (up to 700 kHz)
- 8.5-9.3 eV in Xe (up to 250 kHz)
- HHG (>50 kHz)
- TR-OPTICS on same crystal
- 2PPE
- Polarization control
- OPA pumping up to mid-IR

Generation of ultrashort UV photons for TR-ARPES

Harmonics in Crystals	Harmonics in Gases	HHG in Gases

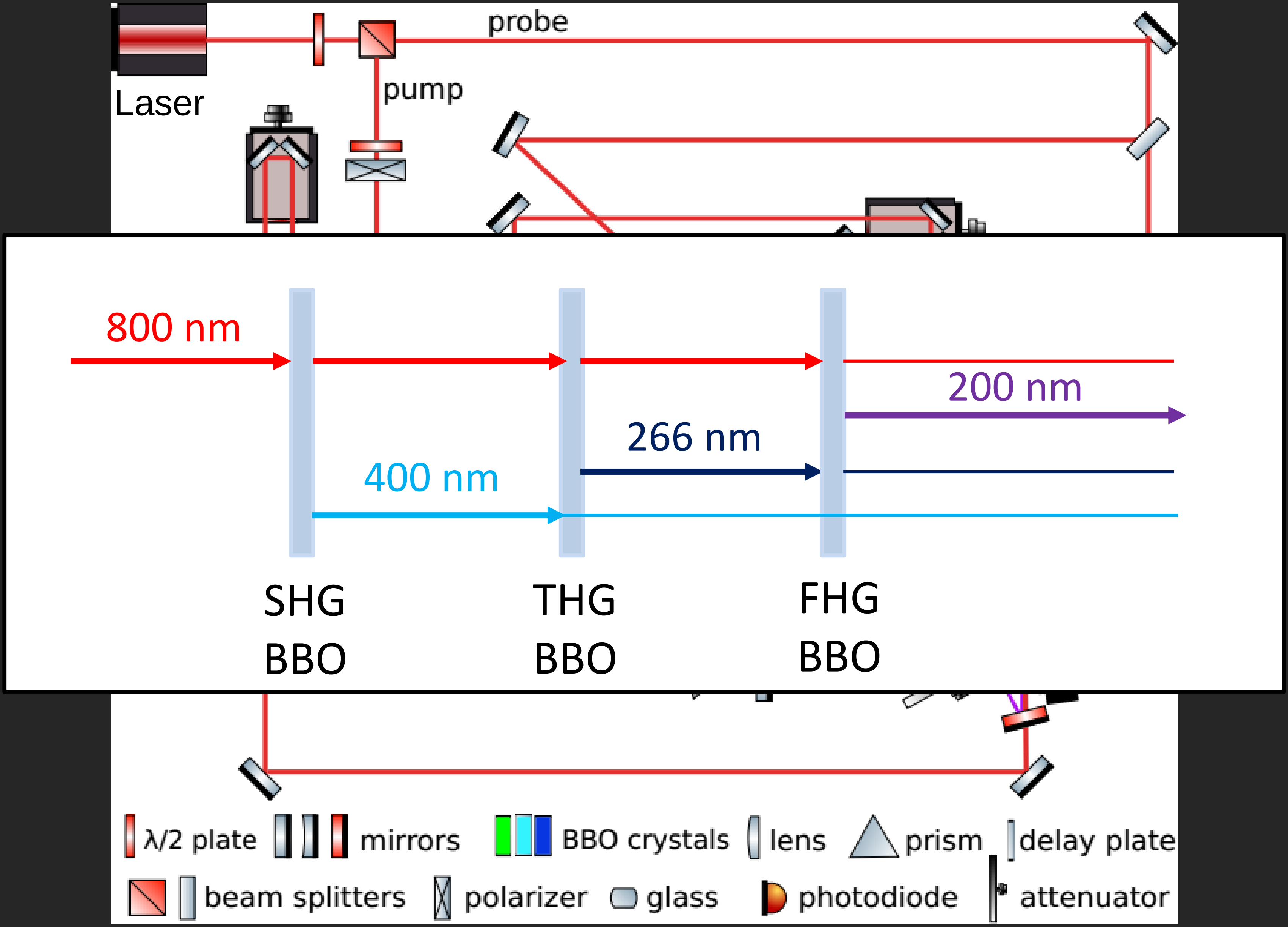




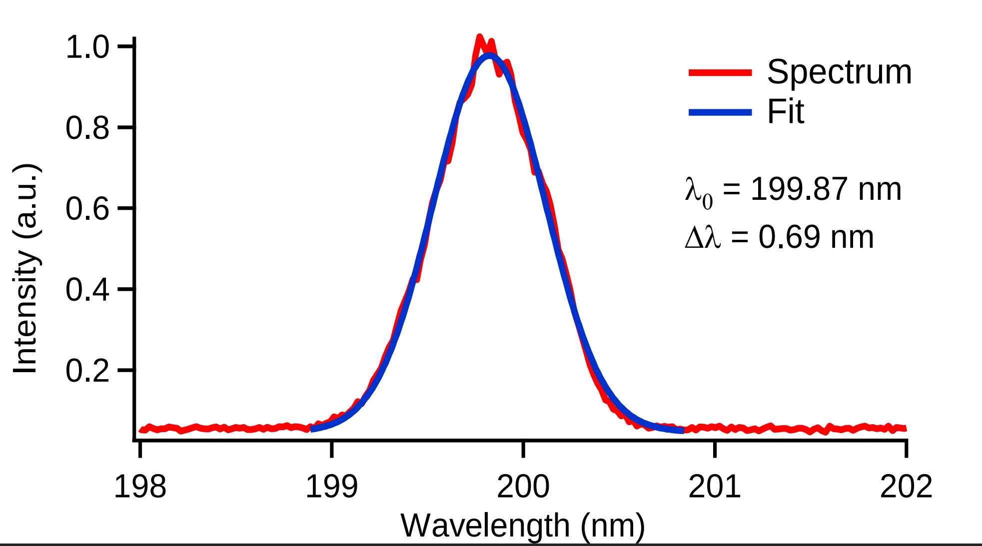


4-5th harmonics SHG or frequency-mixing	6-9th harmonics THG, 4wave mixing	(2n+1)th harmonics Three-step process
no threshold up to 10s MHz	10^{12} W/cm^2 100s kHz - MHz	10^{14} W/cm^2 1-100s kHz

1) Harmonics in Crystals (6-7 eV): generation



1) Harmonics in Crystals (6-7 eV): pulse characterization

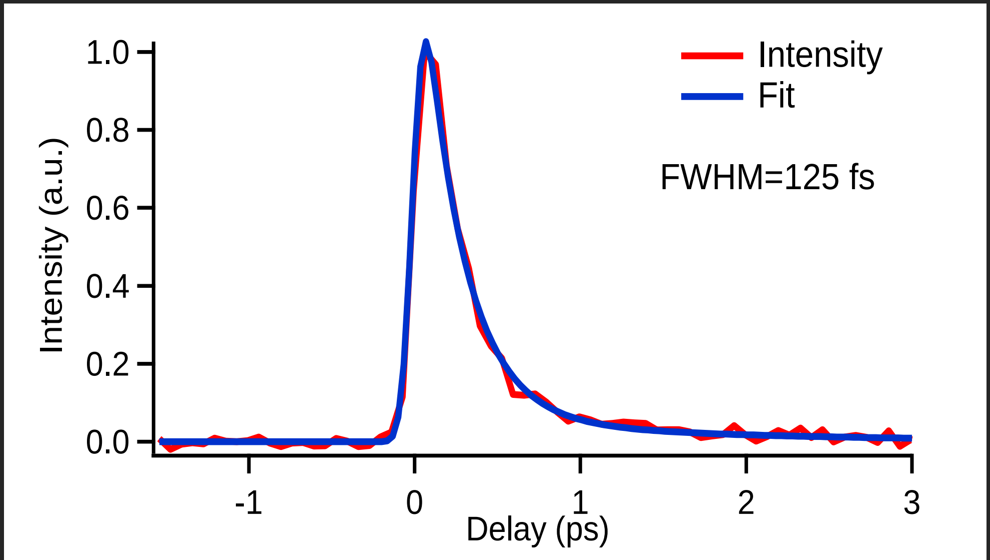


Spectrum measured via high-resolution (0.1 nm) monochromator

$$h\nu_{IV} = 6.2\ \mathrm{eV}$$

$$\Delta E_{IV} = 21\ \mathrm{meV}$$

$$\Delta\tau_{IV,TL} = 85\ \mathrm{fs}$$



Time-resolution measured via TR-ARPES experiment

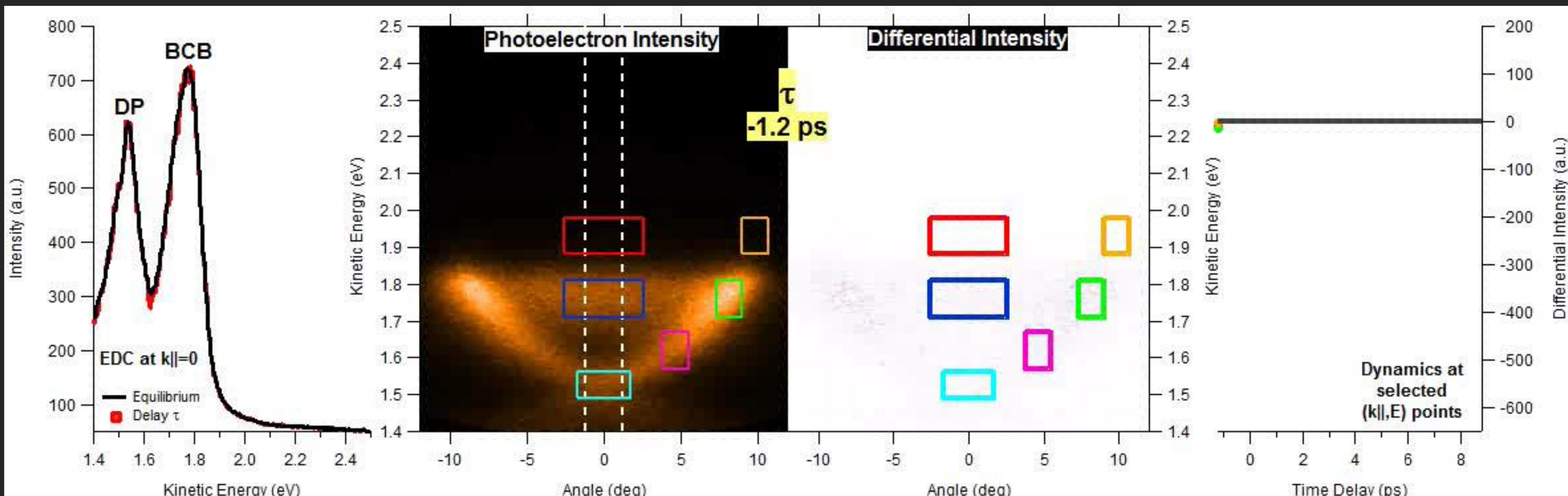
$$\Delta t_{tot} = \sqrt{\Delta t_{IV}^2 + \Delta t_{PUMP}^2} = 125\ \mathrm{fs}$$

$$\Delta t_{PUMP} \sim 60\ \mathrm{fs}$$

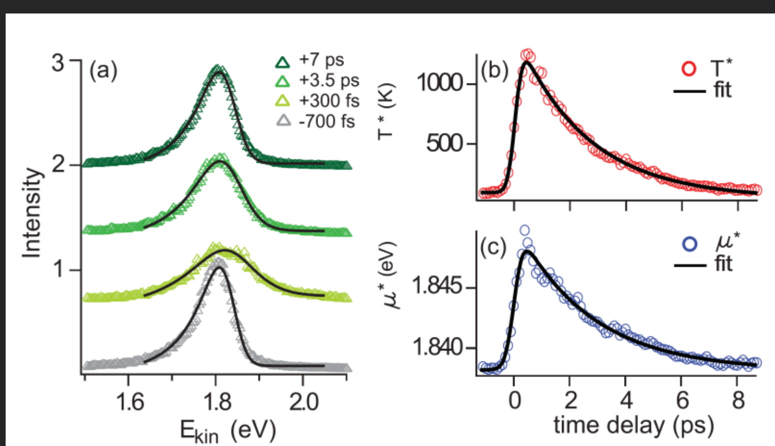
$$\Delta t_{IV} \sim 110\ \mathrm{fs}$$

1) Harmonics in Crystals (6-7 eV): results

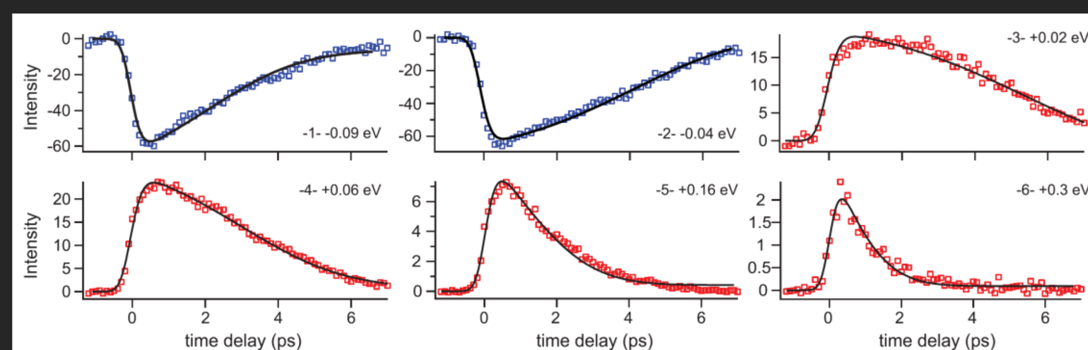
TR-ARPES on Topological Insulators: Relaxation Dynamics on Bi_2Se_3



Electronic Temperature Te(t) can be extracted



Te(t) suffices to reproduce I(E,t) with a time-dependent FD

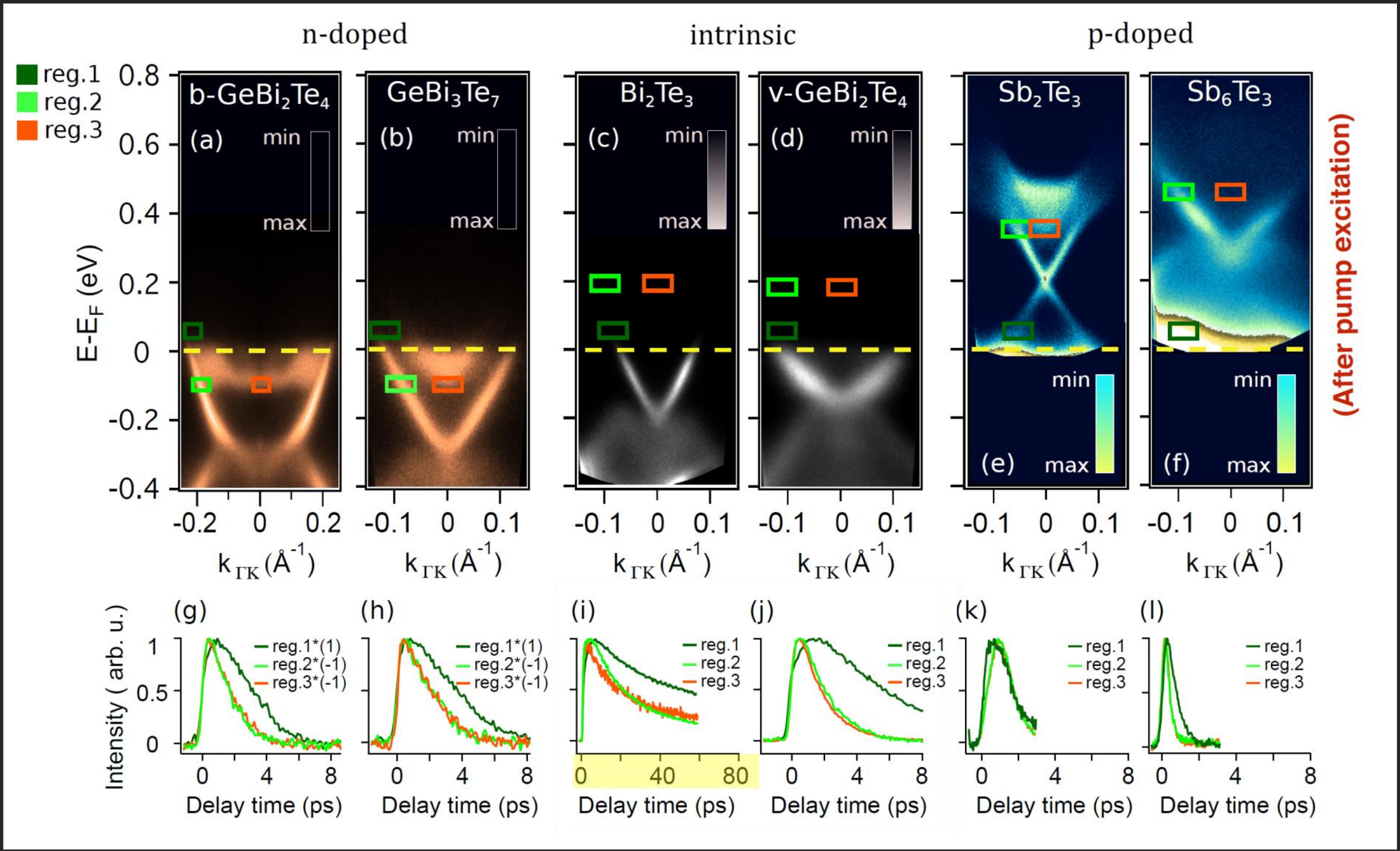


τ(Te)≈2.5 ps

A. Crepaldi et al., PRB **86**, 205133 (2012)

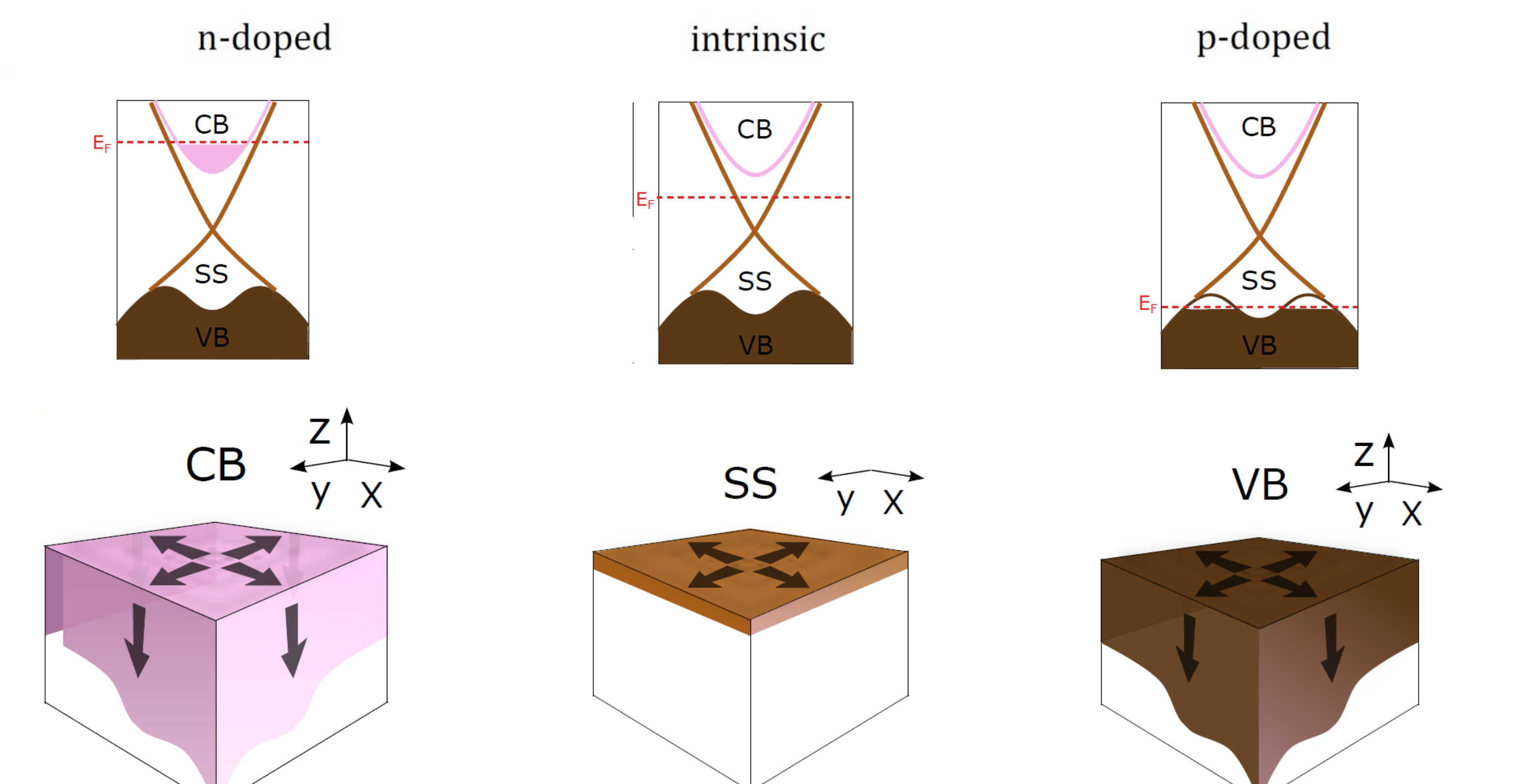
1) Harmonics in Crystals (6-7 eV): results

TR-ARPES on Topological Insulators: comparing different TIs



A. Sterzi et al., PRB **95**, 115431 (2017)

1) Harmonics in Crystals (6-7 eV): results

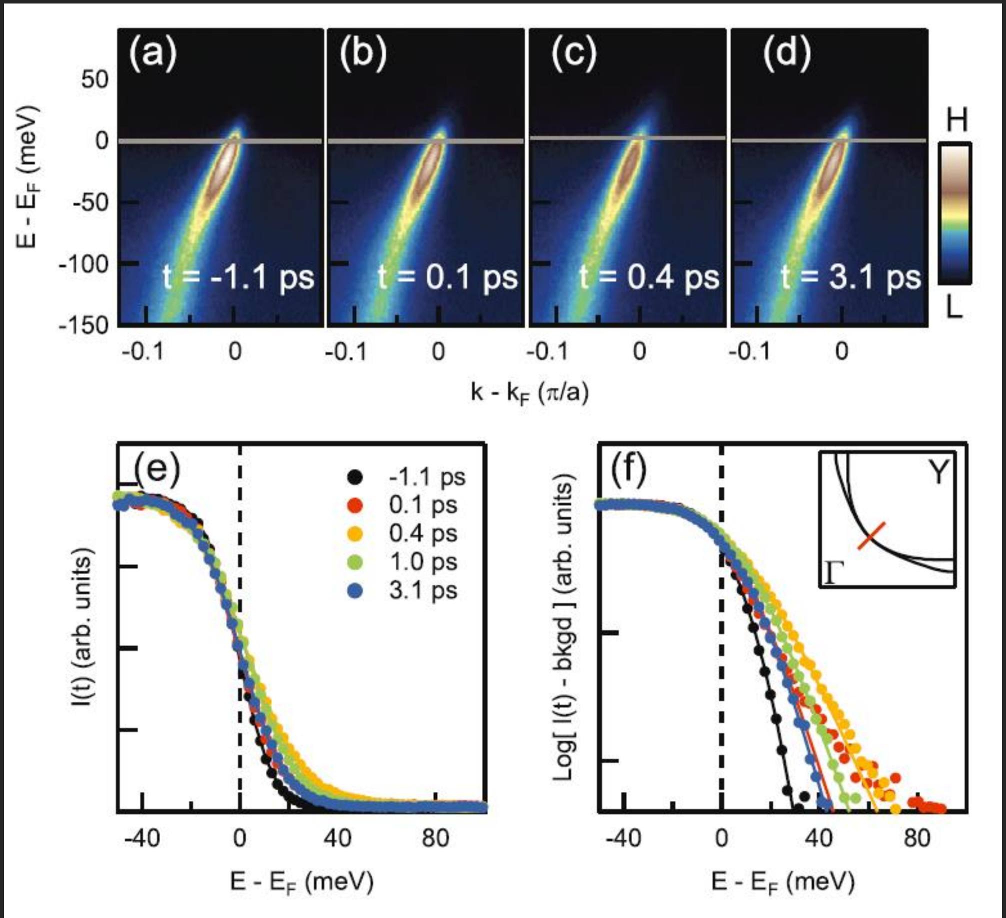


PRB **95**, 115431 (2017)

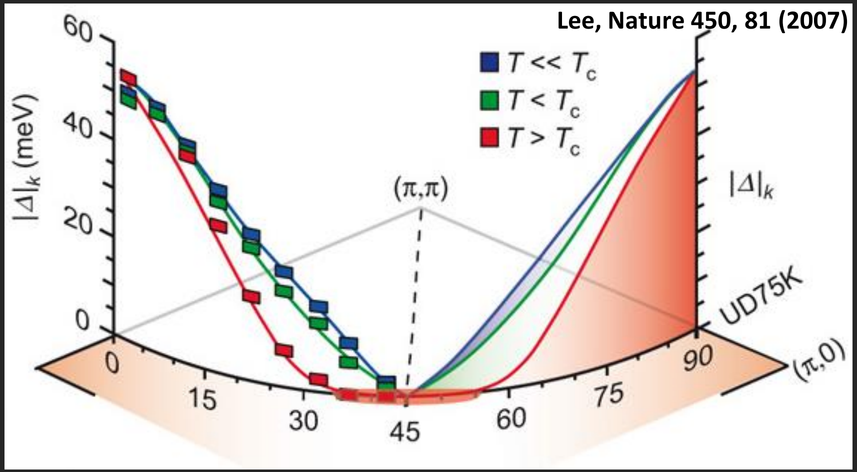
<u>Observation</u>: when no bulk states cross the Fermi level, carriers excited in the topological surface state cannot decay into bulk states, hence the relaxation of carriers is less efficient.

1) Harmonics in Crystals (6-7 eV): results

Results on High-Critical Temperature (Cuprate) Superconductors:
the case of Bi2212 ($Bi_2Sr_2CaCu_2O_{8+\delta}$, Tc=96 K at Optimal Doping)



C. Smallwood et al, PRB 89, 115126 (2014)

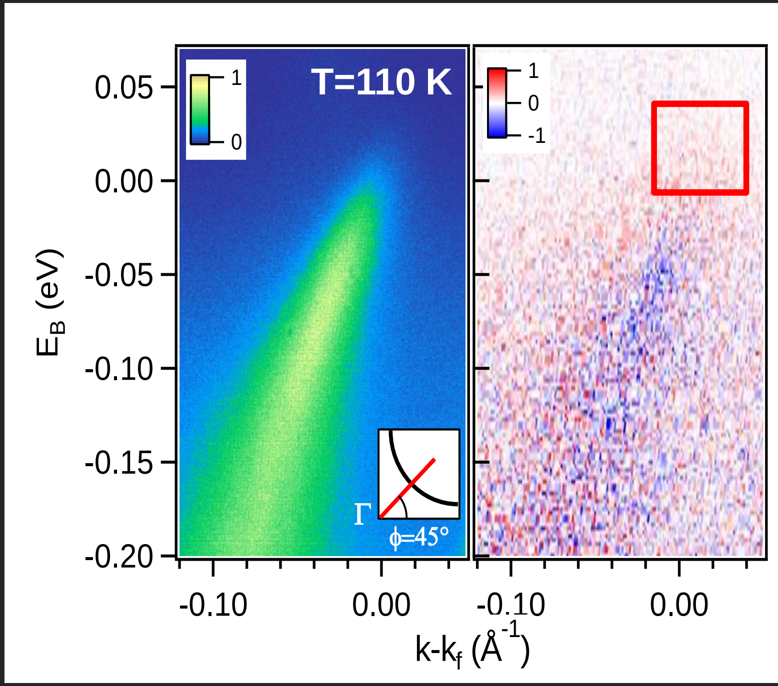


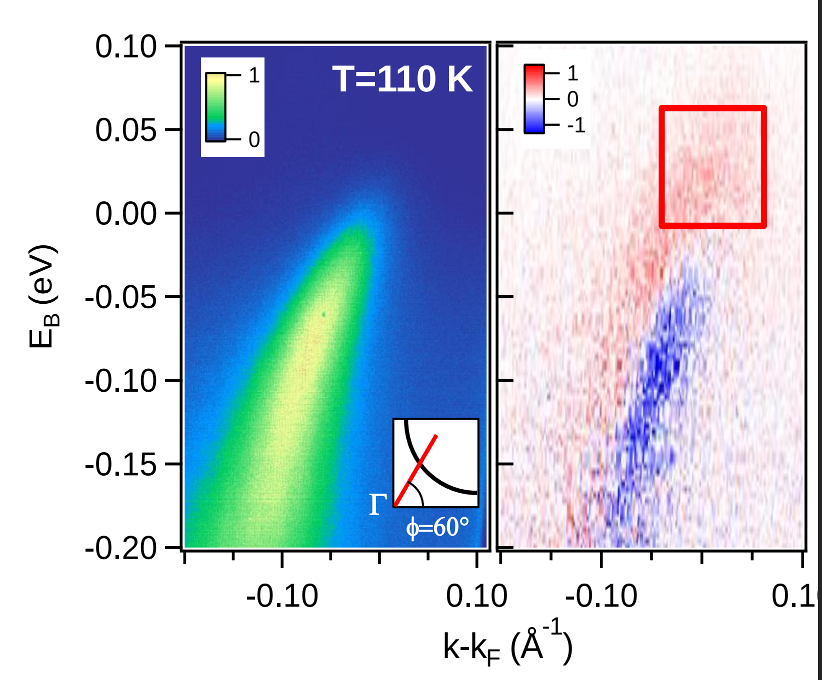
- The FS is an 'arc'
- The GAP in the DOS is anisotropic
- At the AN, the gap is open also above Tc

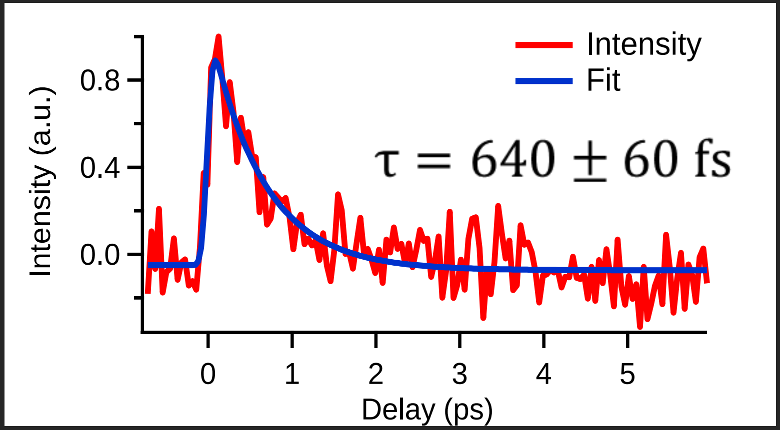
ultrahigh resolutions
ΔE_B~10 meV
$\Delta\vartheta$~0.1° ($\Delta k_{||}$~0.003 Å^{-1})

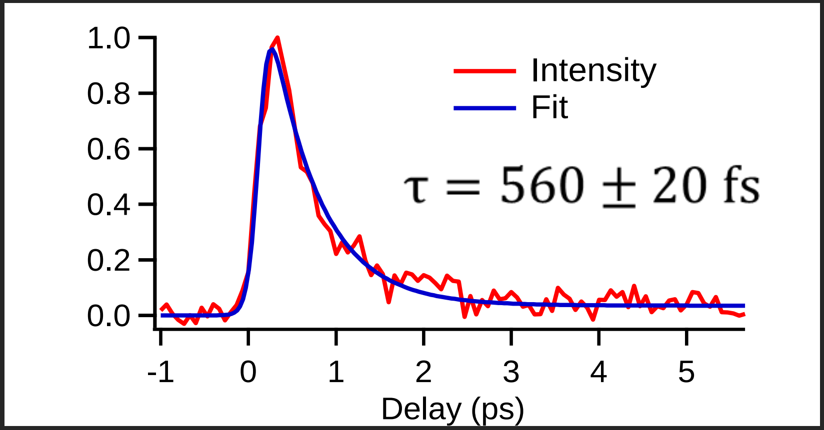
1) Harmonics in Crystals (6-7 eV): results

In the pseudogap state, the dynamics are similar in different regions of the FS





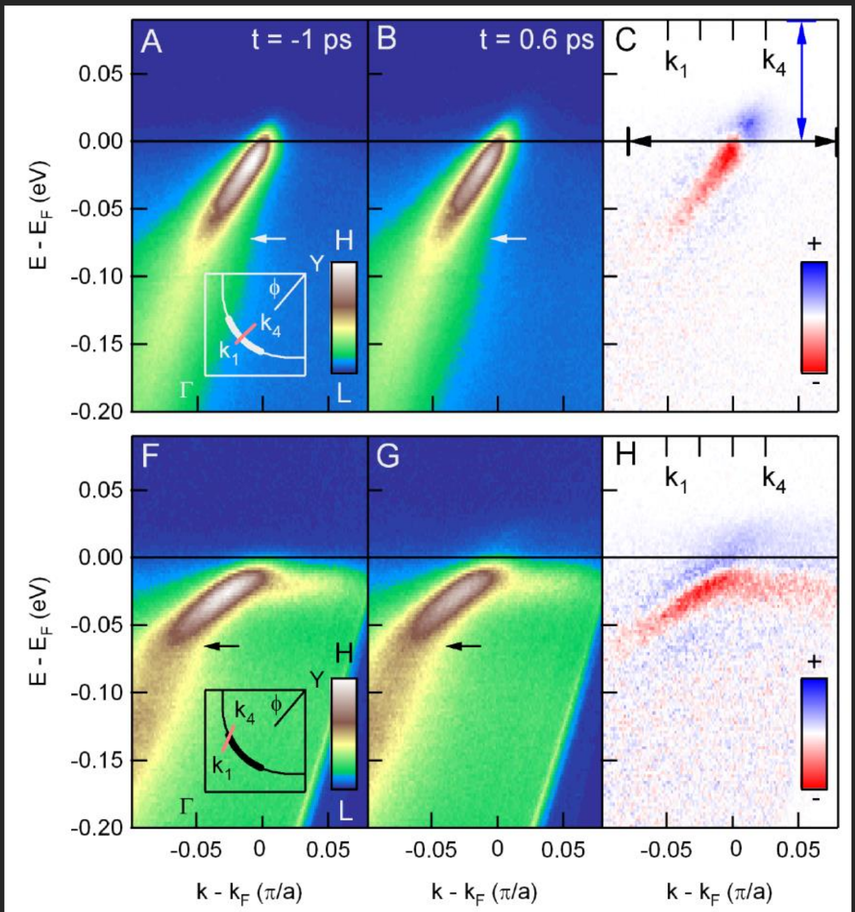




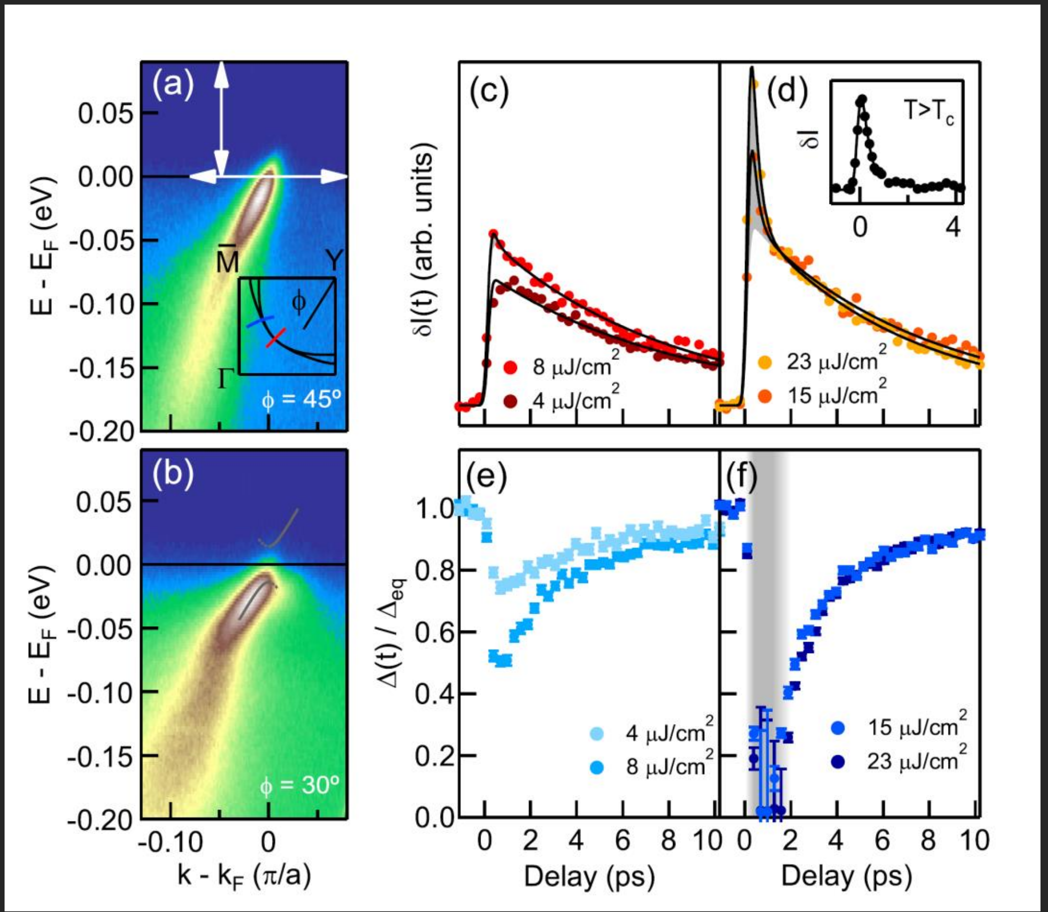
1) Harmonics in Crystals (6-7 eV): results

Non-equilibrium electron dynamics in Bi2212, superconducting state:

- At different momenta on the FS
- At *low* (5 uJ/cm^2) and *high* (15 uJ/cm^2) fluence: melting superconductivity



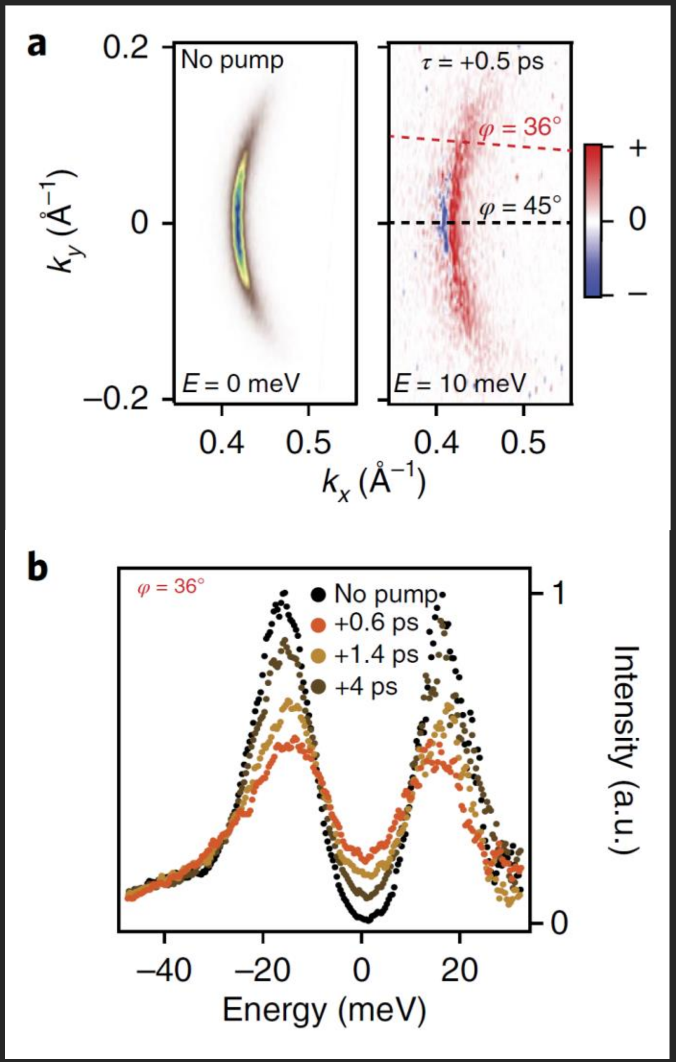
C. Smallwood et al., Science 336, 1137 (2012)

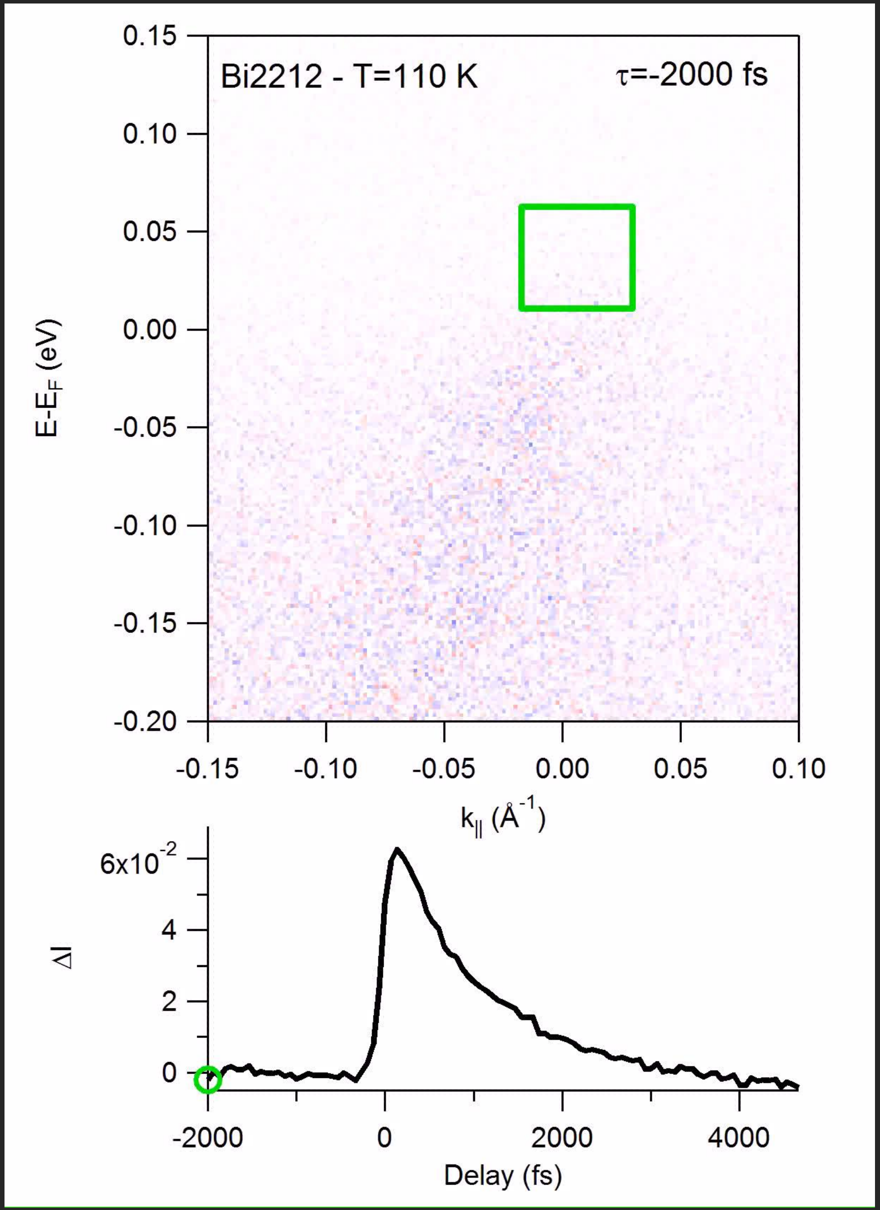


C. Smallwood et al., Phys. Rev. B 92, 161102(R) (2015)

1) Harmonics in Crystals (6-7 eV): results

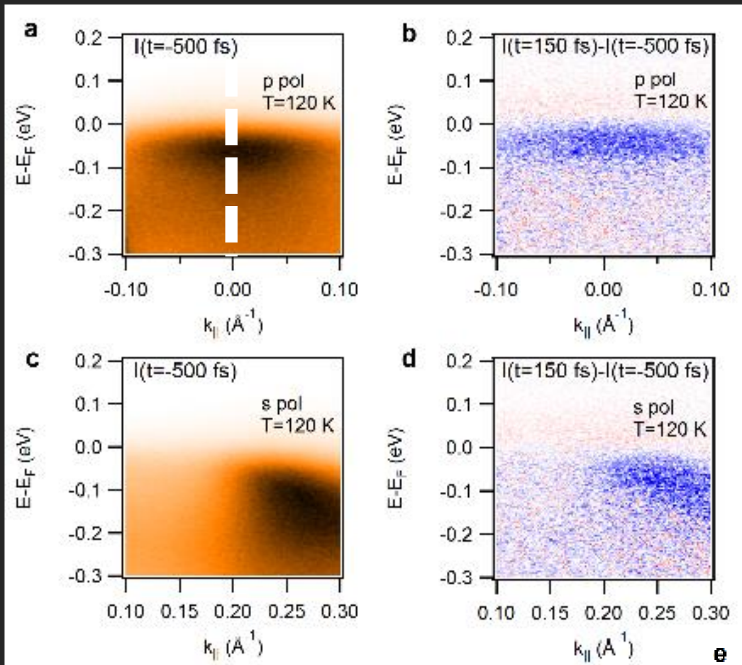
Non-equilibrium FS and Gap dynamics:
<u>the gap '*fills*' without closing</u>





F. Boschini et al., Nature Materials **17**, 416 (2018)

1) Harmonics in Crystals (6-7 eV): results on FeSeTe, selection rules



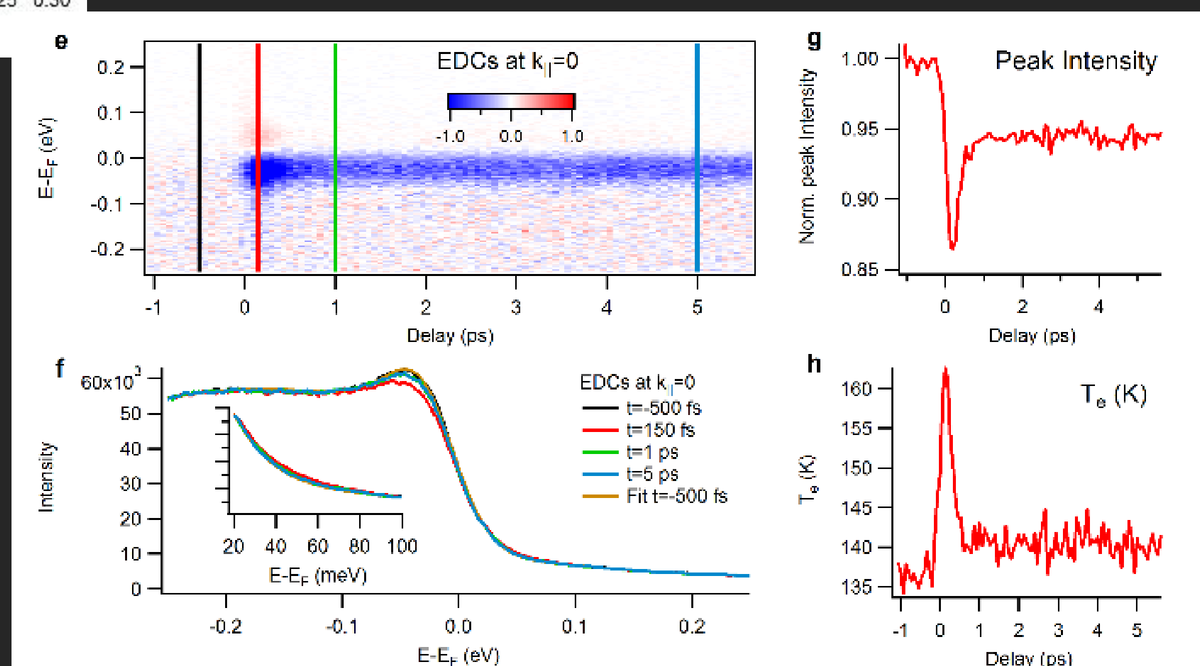
$FeSe_{0.4}Te_{0.6}$, T_c~14 K, T=120 K, hν=6.2 eV

Two h-pockets at Γ disentagled by selection rules (probe polarization).

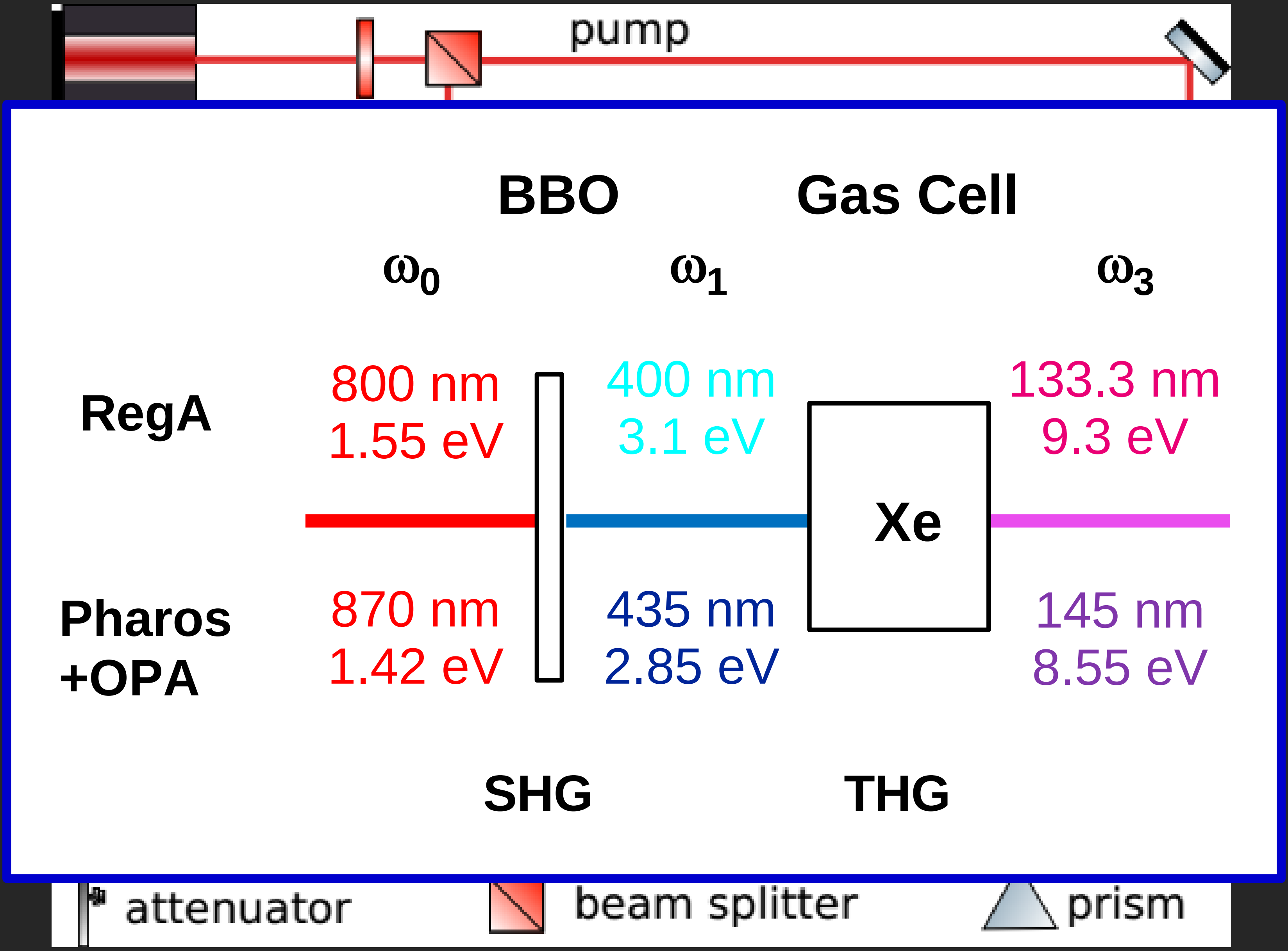
Both display similar dynamics, within a ~250 fs time resolution

Via EDCs analysis we disentangled two effects:

- Ultrafast increase and relaxation of Te
- Long-lasting reduction of the peak intensity

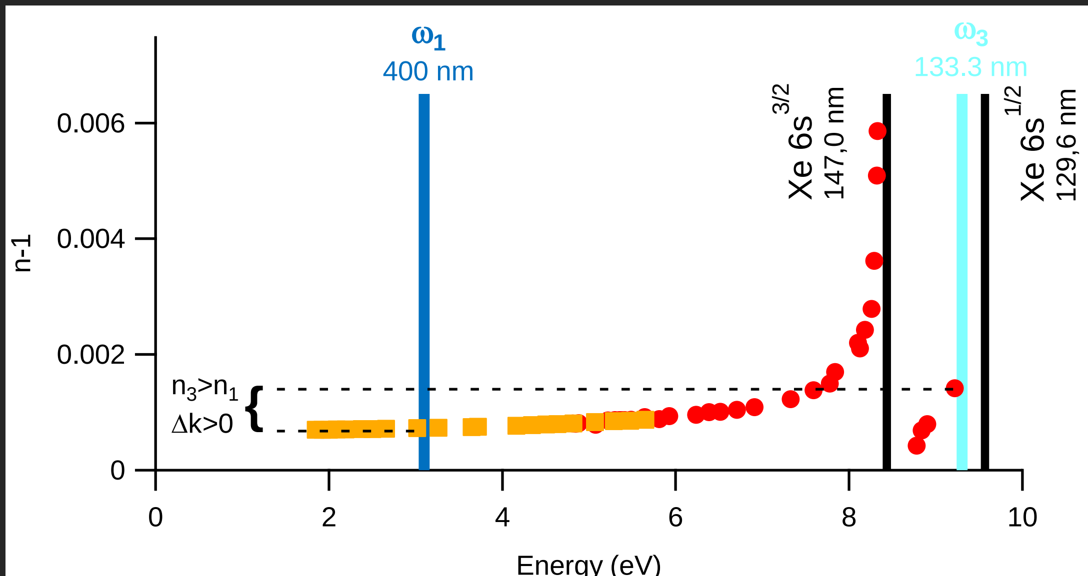


2) Harmonics in Gases (8-11 eV): generation



2) Harmonics in Gases (8-11 eV): generation, as 4WM or 6WM

Xe refractive index



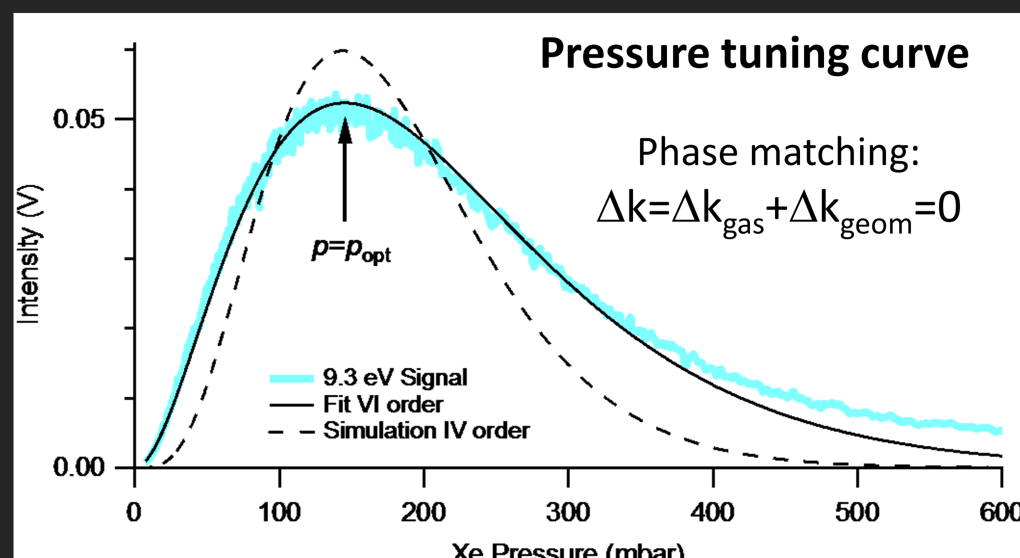
$\Delta k_{gas}>0$

Positive dispersion for THG of 3.1 eV

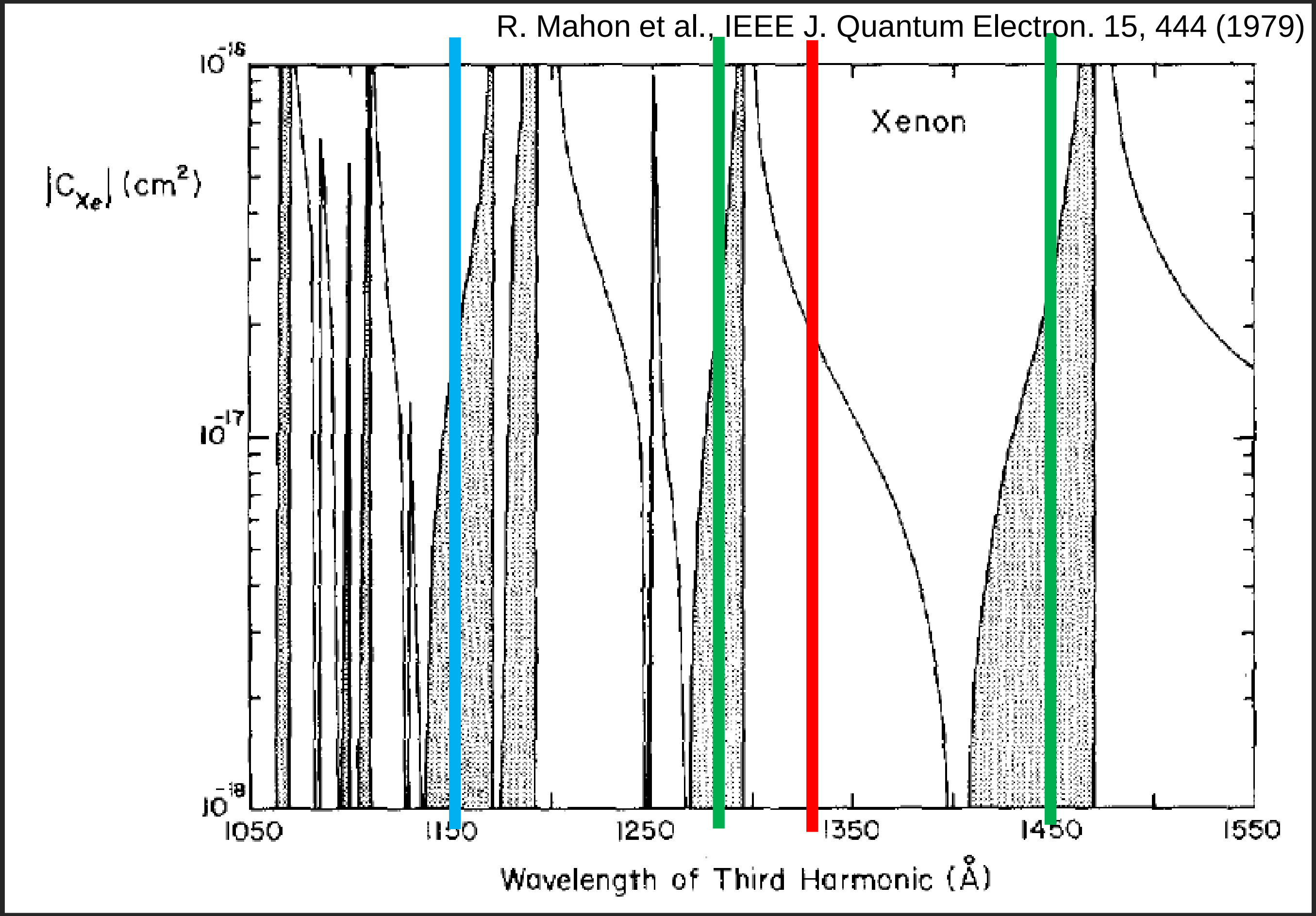
F. Cilento et al., J. Electr. Spectrosc. Relat. Phenom. 207, 7 (2016)

When focusing, a phase shift (Gouy term) depending on focusing geometry and NL interaction is introduced. Only a 6-wave mixing introduces a $\Delta k_{geom}<0$, compensating $\Delta k_{gas}>0$.

~~$\omega_3=3\omega_1$~~ $\omega_3=4\omega_1-\omega_1$



2) Harmonics in Gases (8-11 eV): generation, maximize efficiency

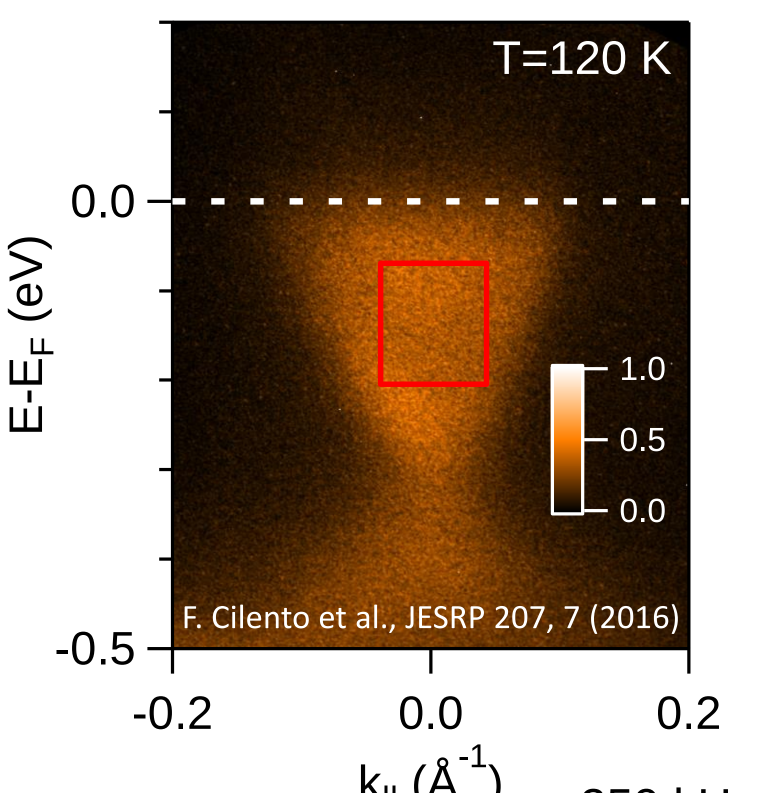


<u>YES</u>, by working in the negative-dispersion regime (green), where THG as a 4-wave mixing is allowed. In red, THG as 6-wave mixing from 800 nm pulses.

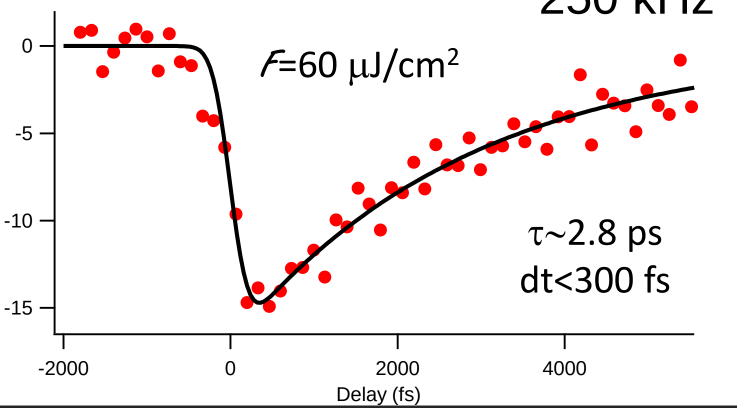
a) TUNE seed energy (w. OPA) to 770 nm or <u>870 nm</u> and make 6HG *green*
b) Use 1035 nm seed (fundamental) and make 9HG *blue*

2) Harmonics in Gases (8-11 eV): results on Bi_2Se_3

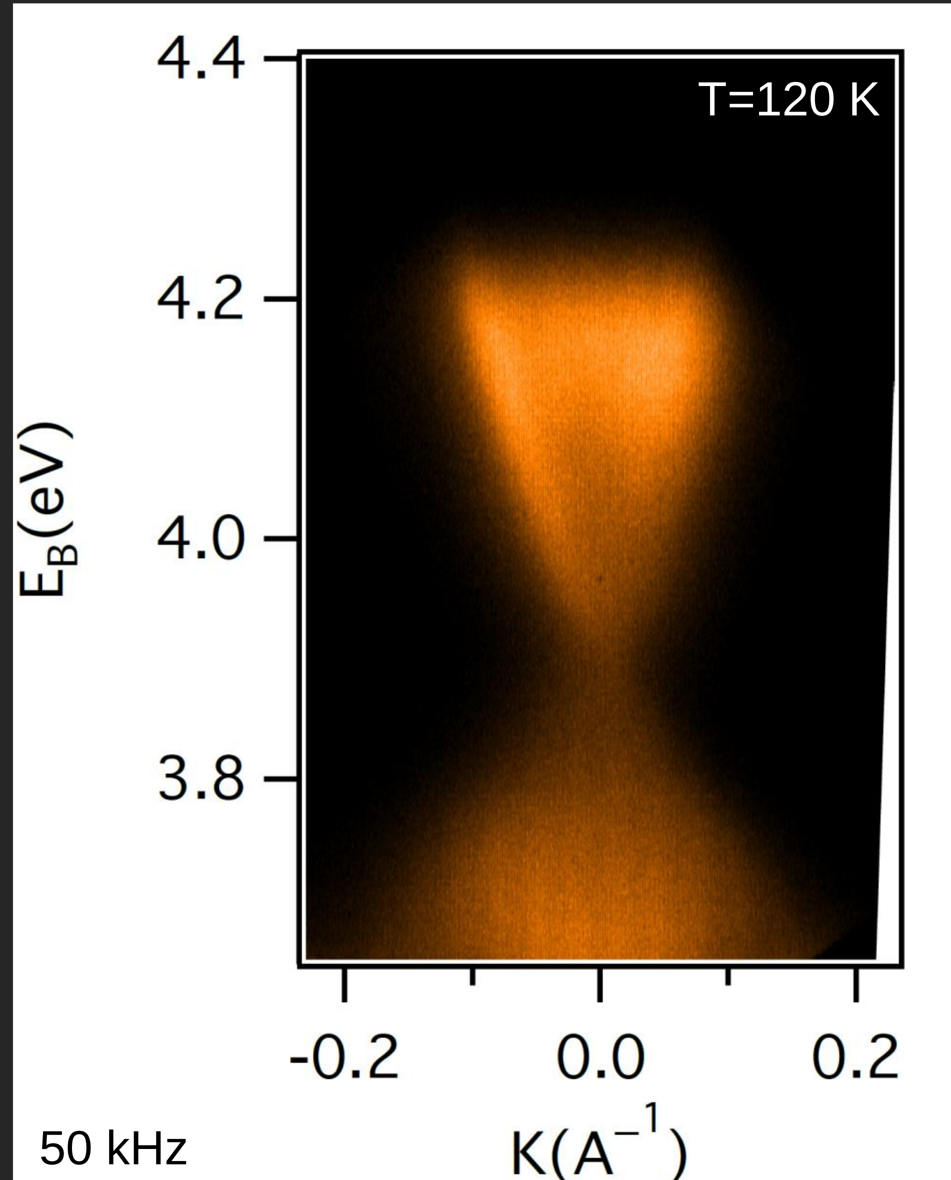
hν=9.3 eV



250 kHz



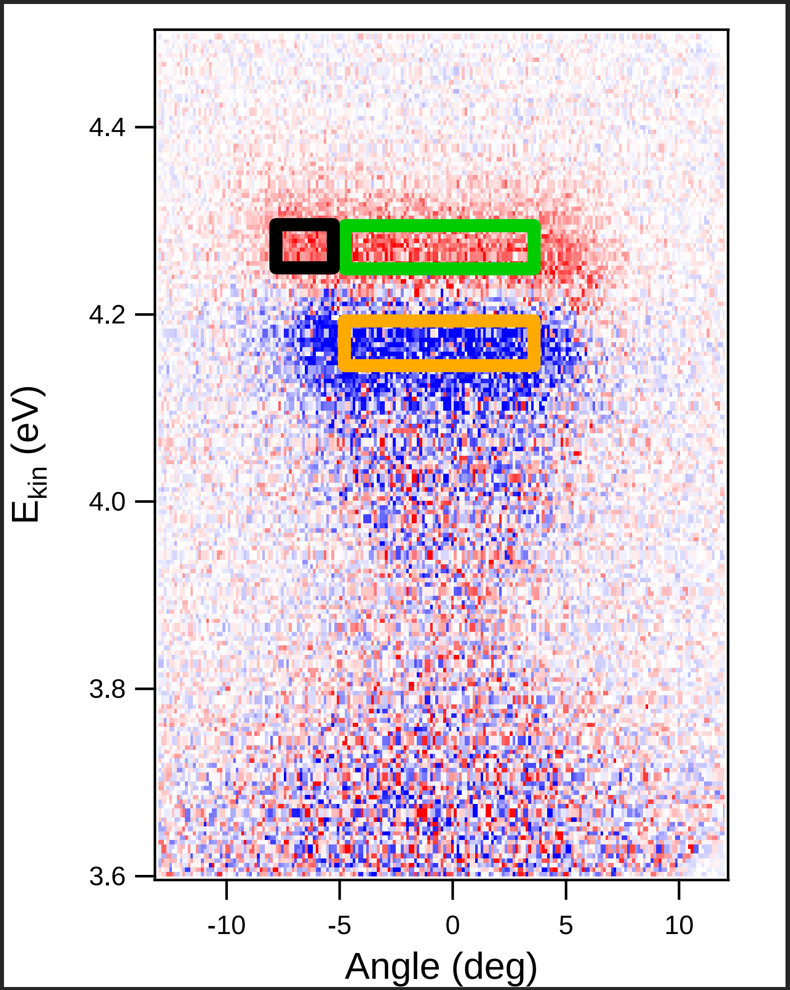
hν=8.5 eV

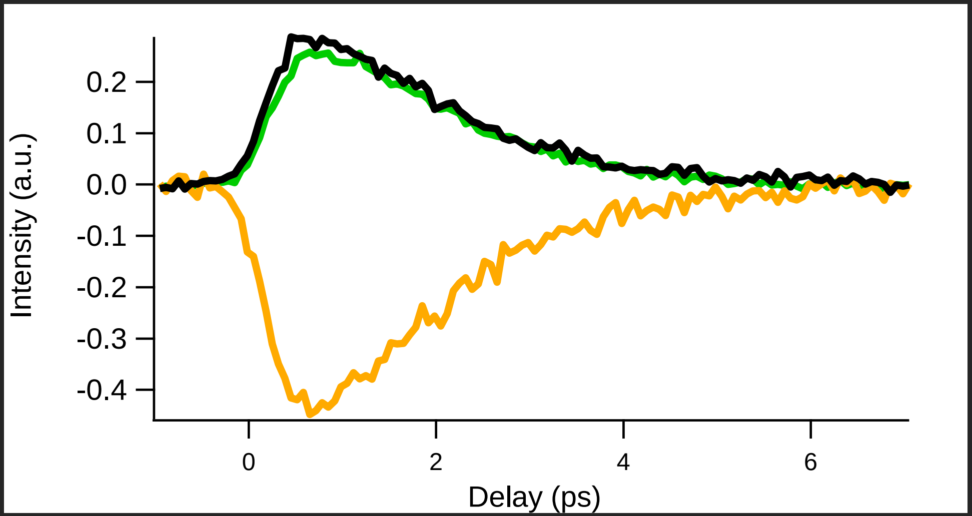


50 kHz

2) Harmonics in Gases (8-11 eV): results on Bi_2Se_3, pump-probe

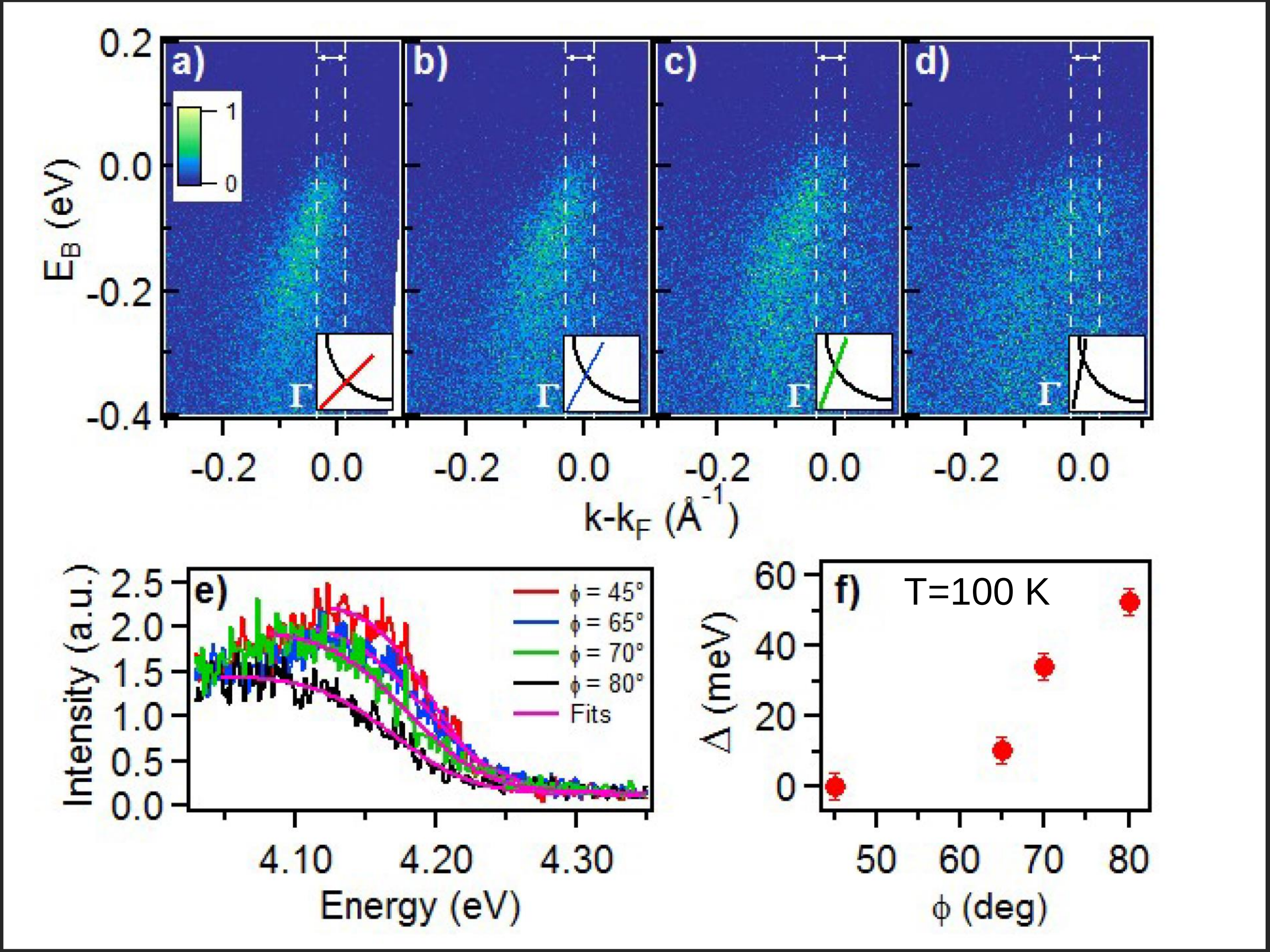
Pump-Probe at 8.5 eV – Bi_2Se_3



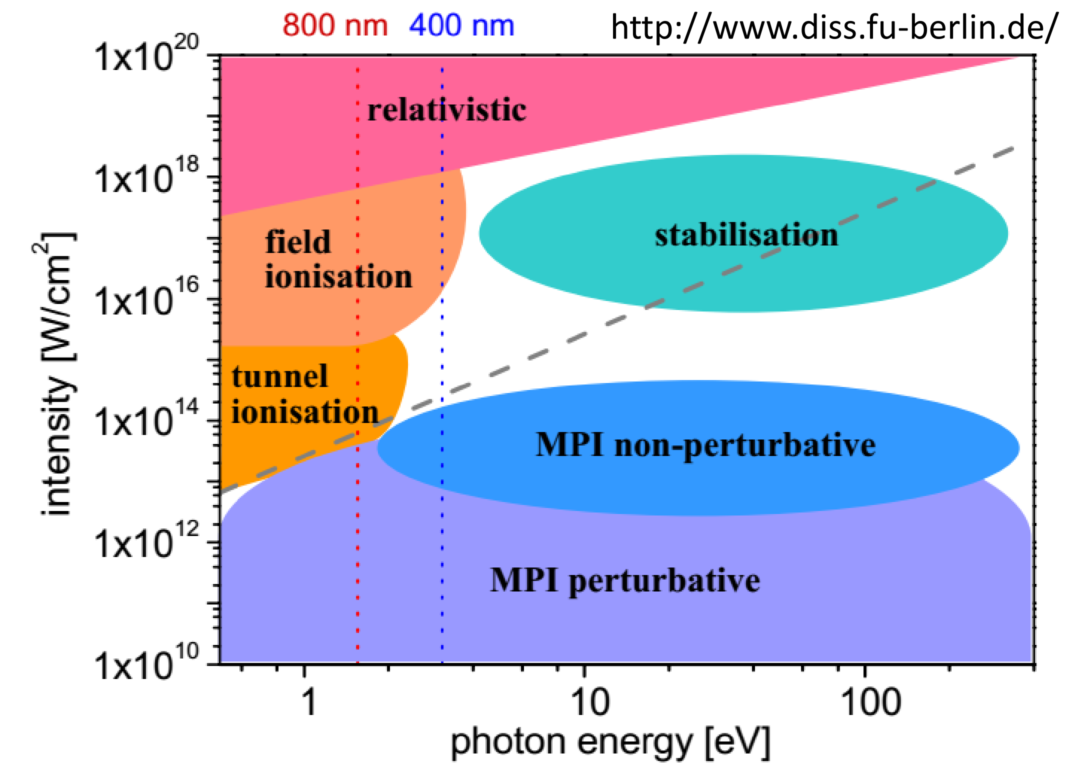


<<1 hour acquisition time!!

2) Harmonics in Gases (8-11 eV): results on Bi2212, Full BZ mapping



3) HHG: generation, intensity regime

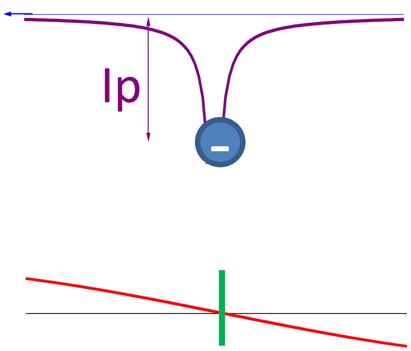


Two ways to obtain the required intensity

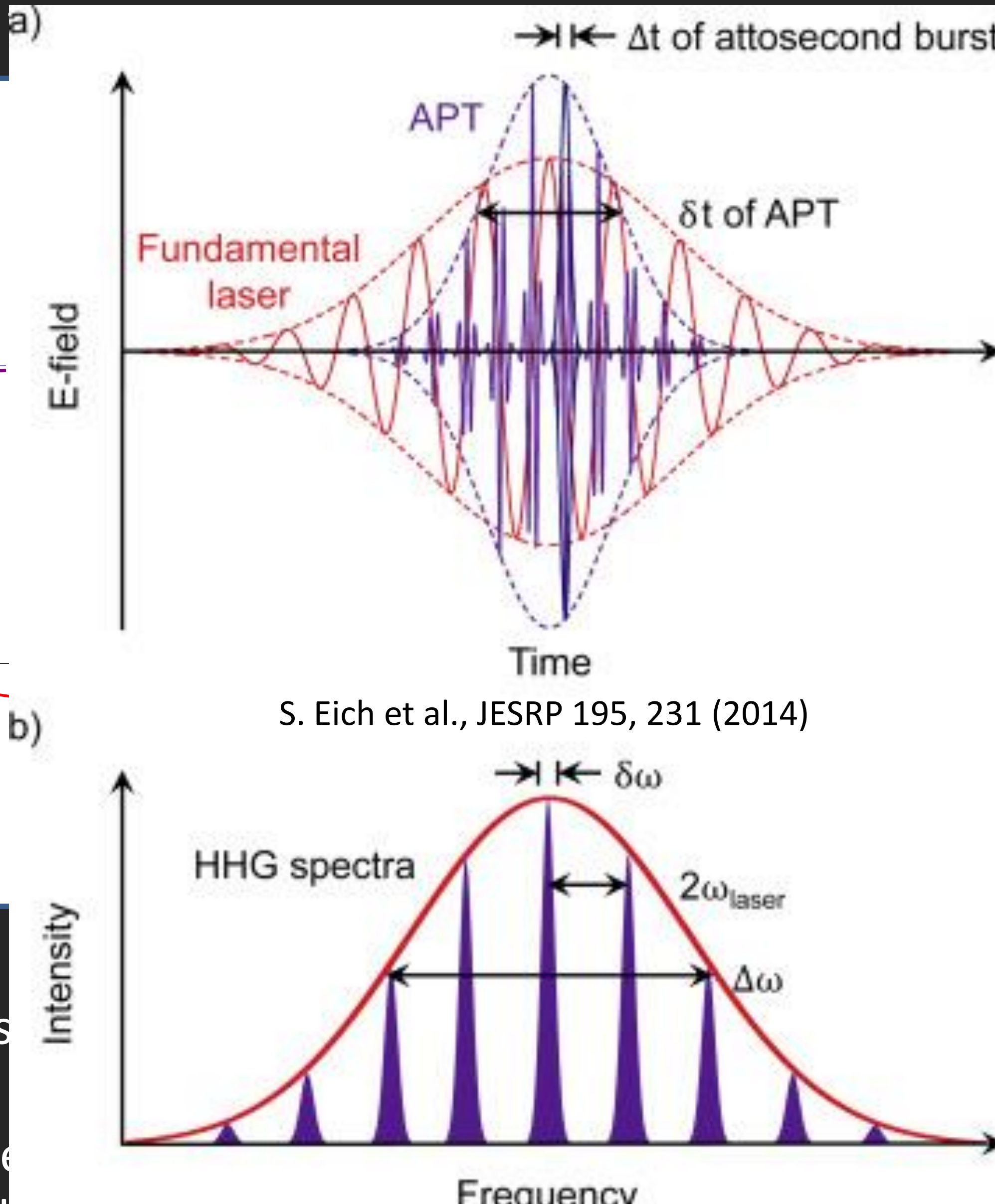
- high energy/pulse ~ mJ
- low repetition rate ~ 1 kHz
- loose focusing

- moderate energy/pulse ~ 10*s* μJ
- high repetition rate ~ 50 kHz
- tight focusing

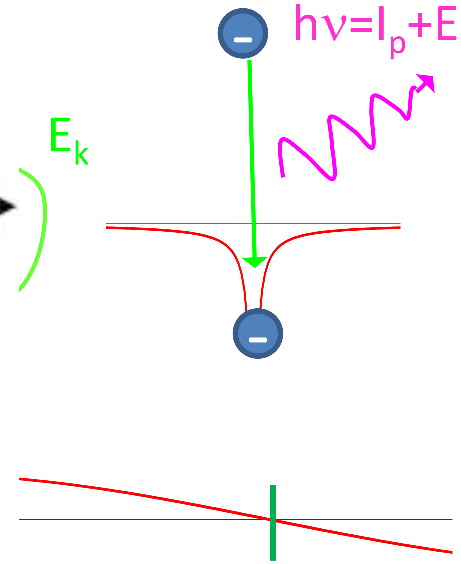
3) HHG: generation, three step model



fundamental state



S. Eich et al., JESRP 195, 231 (2014)

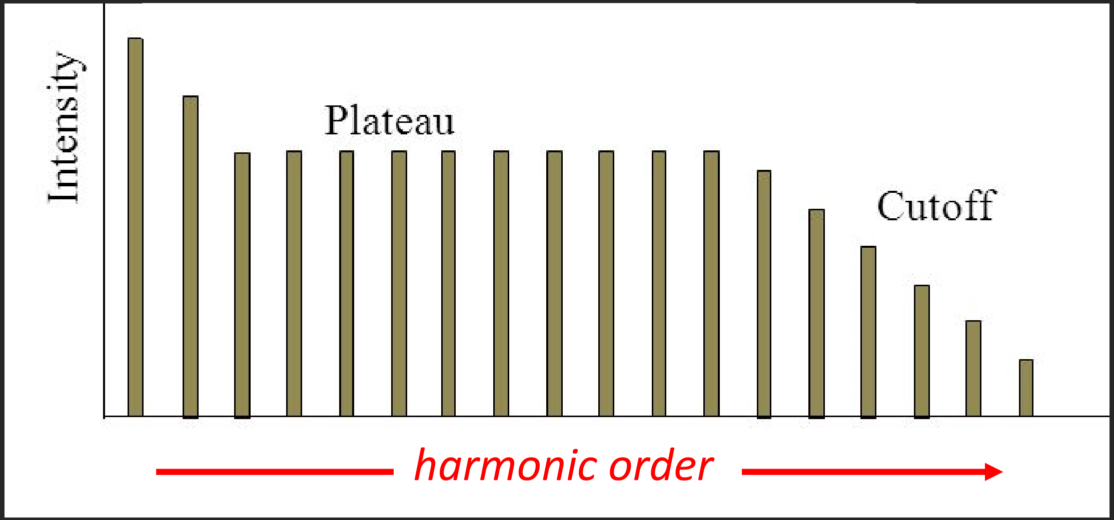


on recombination to fundamental state

P. B. Corkum PRL 71, 1994 (1993)

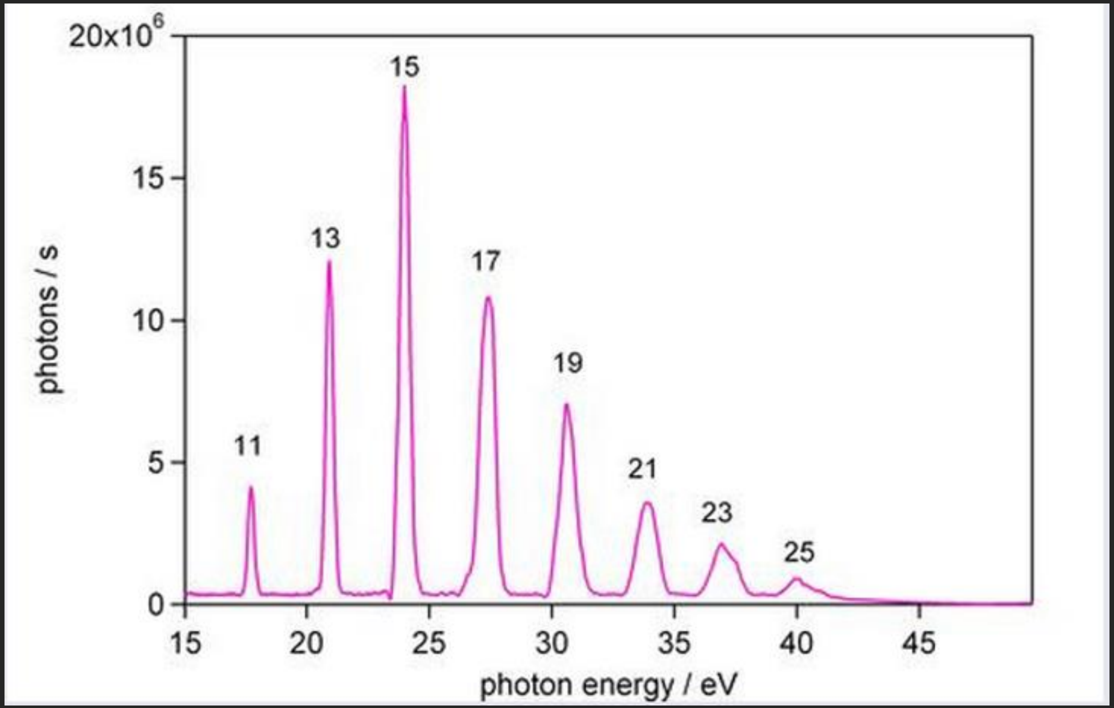
- The periodicity is … duced
- The energy of the … ion potential of the gas plus the kinetic energy of the electron.

3) HHG: generation, qualitative aspects



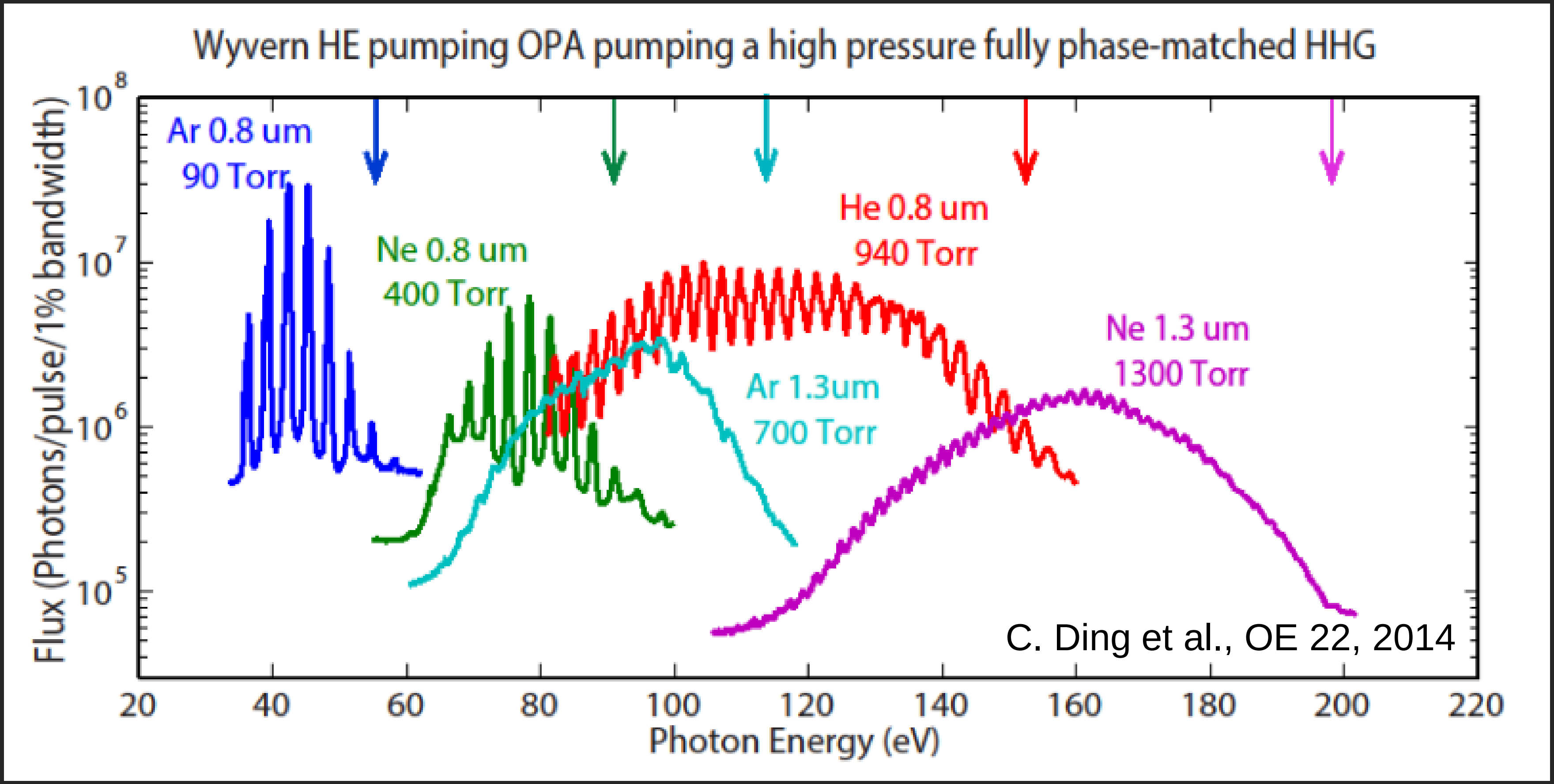
$$E_{cutoff} = I_P + 3.17\,U_P$$
$$\propto I_P + \lambda^2$$

$$Intensity \propto \lambda^{-5} .. \ \lambda^{-6}$$



Measured HHG spectrum, driven by λ=800 nm (1.55 eV)

3) HHG: generation, tunable seed



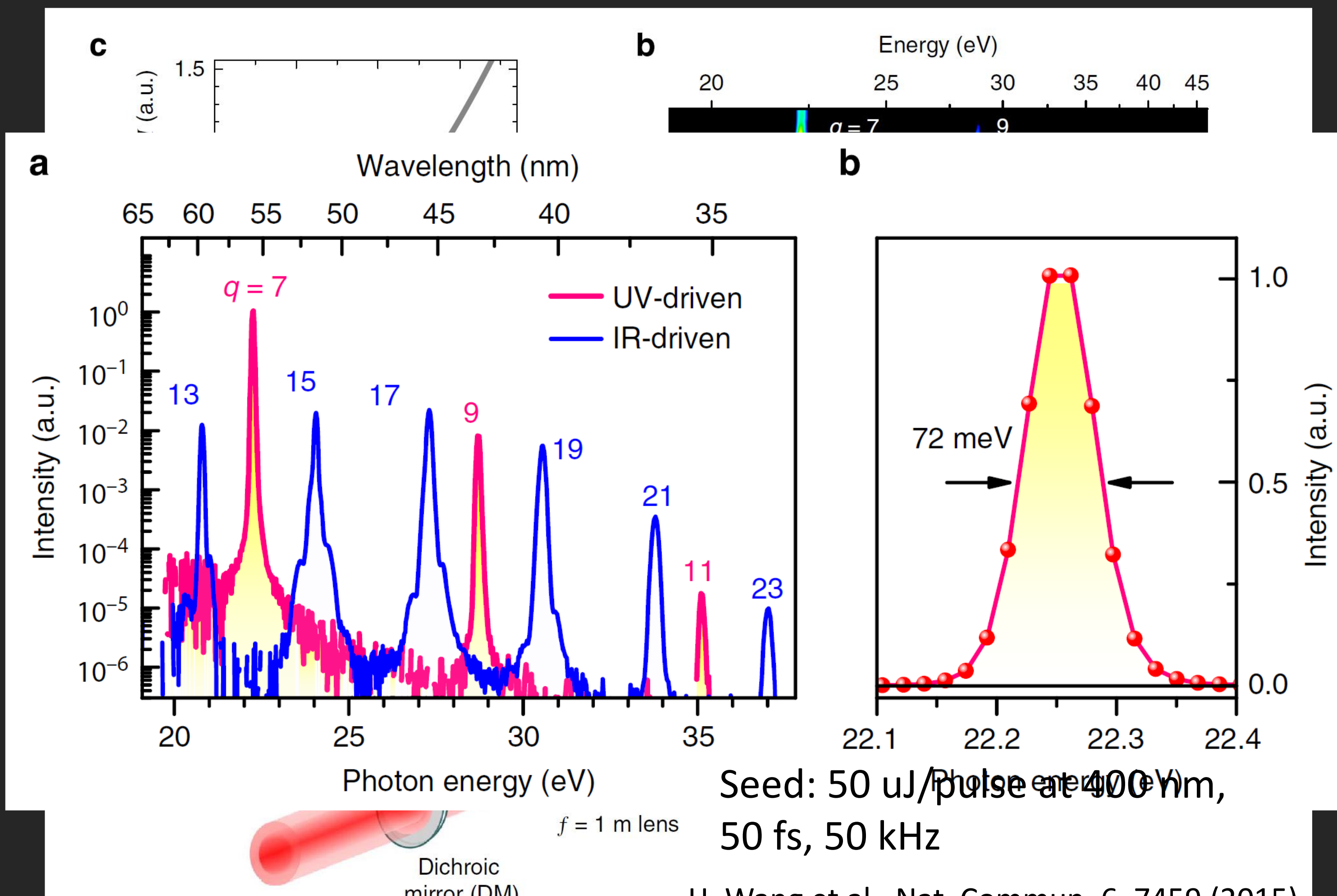
$$E_{cutoff} = I_P + \ 3.17\ U_P \propto I_P + \ \lambda^2$$

Ionization potential: He=24.6 eV, Ne=21.6 eV, Ar=15.8 eV, Kr=14 eV, Xe=12.1 eV

3) HHG: latest developments

Most recent HHG setups / beamlines have been developed to obtain two goals:

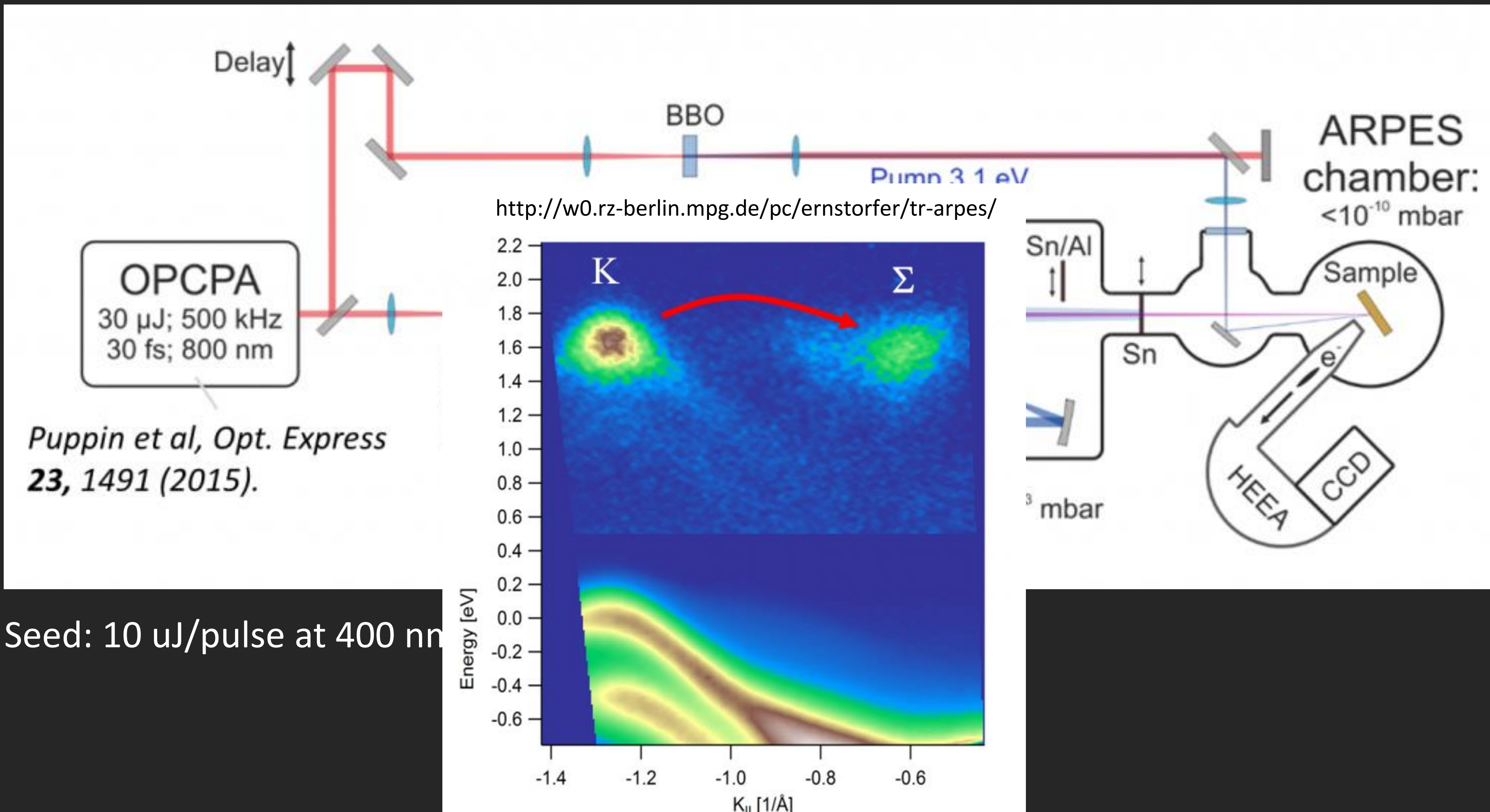
1) high rep. rate operation (>50 kHz, up to 500 kHz – 1 MHz)
2) high photon energies (100-400 eV, ‘supercontinuum’)



Seed: 50 uJ/pulse at 400 nm, 50 fs, 50 kHz

H. Wang et al., Nat. Commun. 6, 7459 (2015)

3) HHG: latest developments

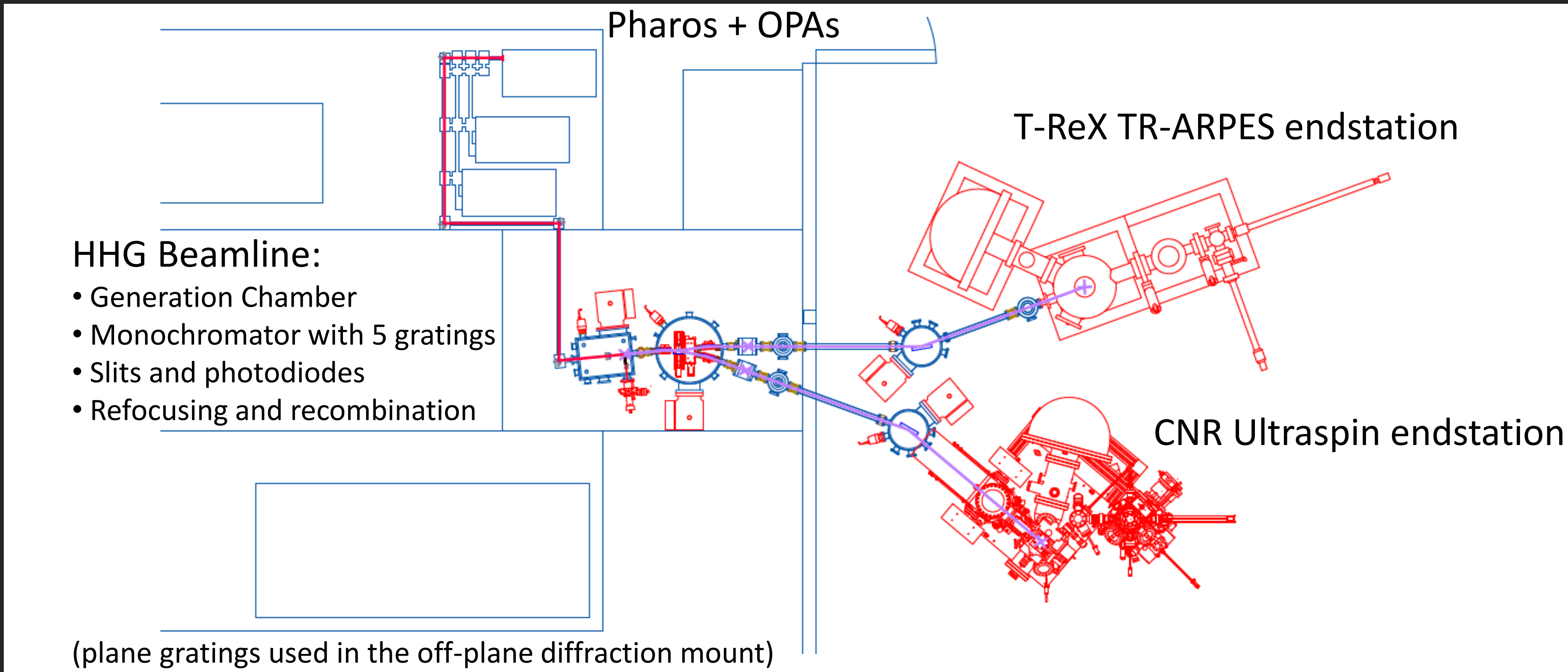


Puppin et al, Opt. Express ***23,*** *1491 (2015).*

Seed: 10 uJ/pulse at 400 nm

First measurements by this setup on In/Si(111), reported on arXiv:1803.11022

3) HHG: latest developments, T-ReX @ FERMI

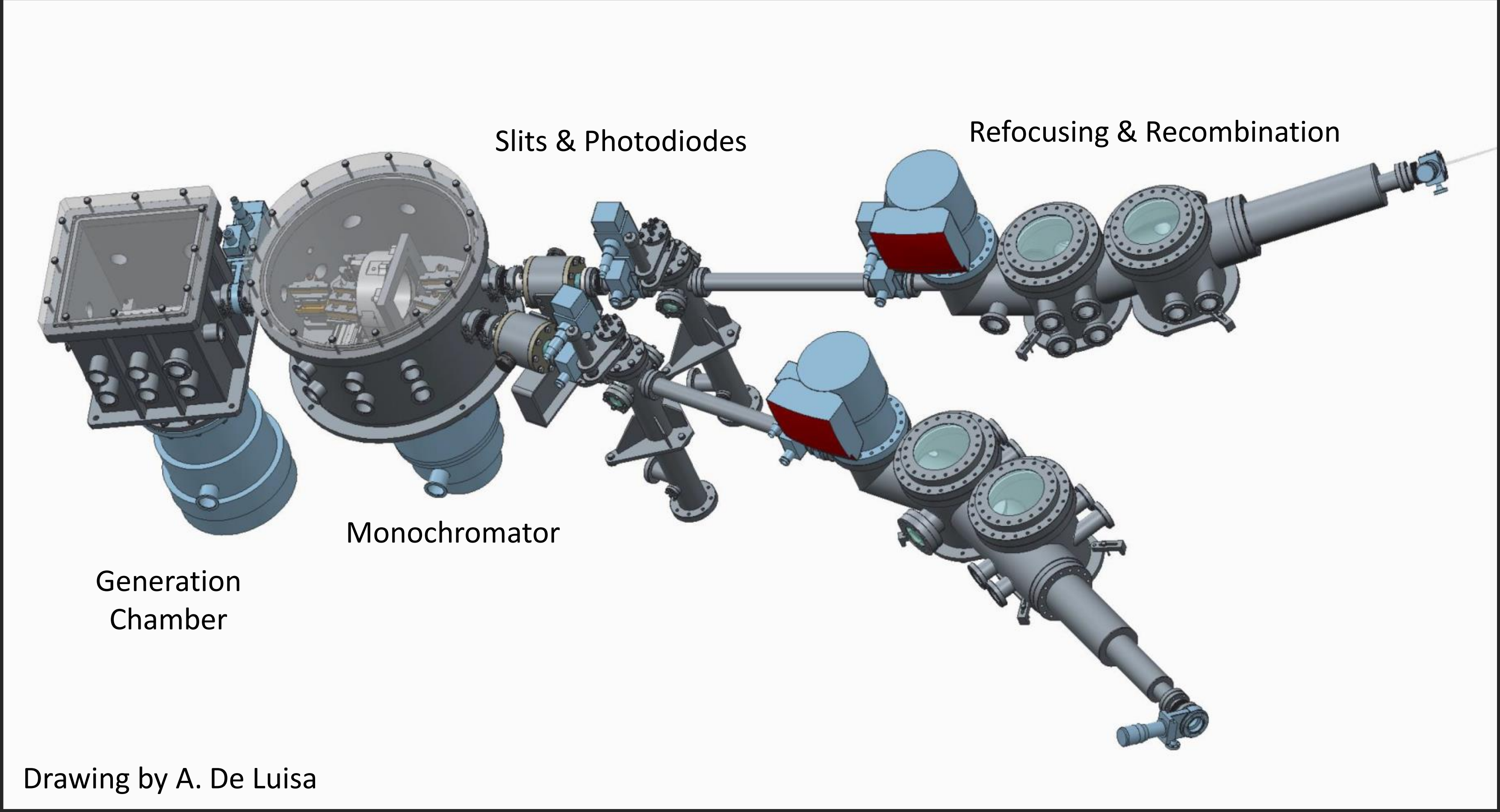


New HHG beamline at high repetition rate (>50 kHz) is under commissioning

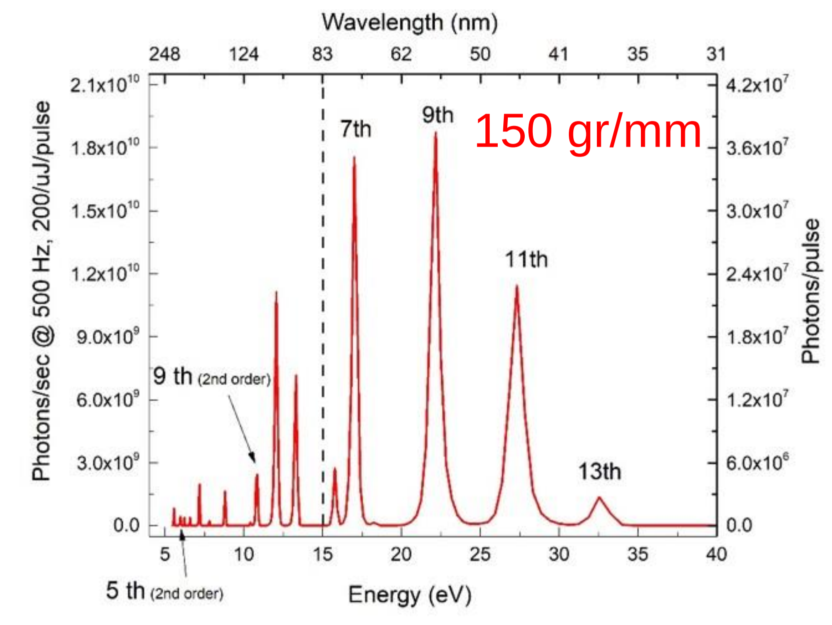
- Seeded by LC Pharos laser (400 uJ/pulse at 1030 nm, 50 kHz, 300 fs)
- Will provide EUV pulses to:
  - the T-ReX TR-ARPES chamber (15-35 eV or 30-80 nm)
  - the Ultraspin Endstation (10-70 eV or 18-90 nm)
- A flux of ~10^{10} photons/s on the sample (at 20 eV) is estimated.
- Estimated energy resolution: 40 meV at 20 eV

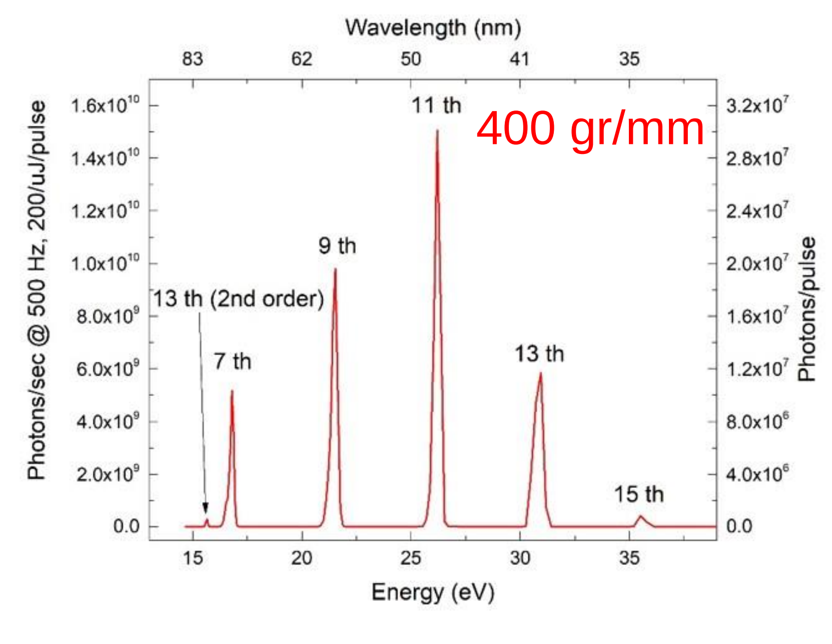
3) HHG: latest developments, T-ReX @ FERMI

The two-brench HHG beamline

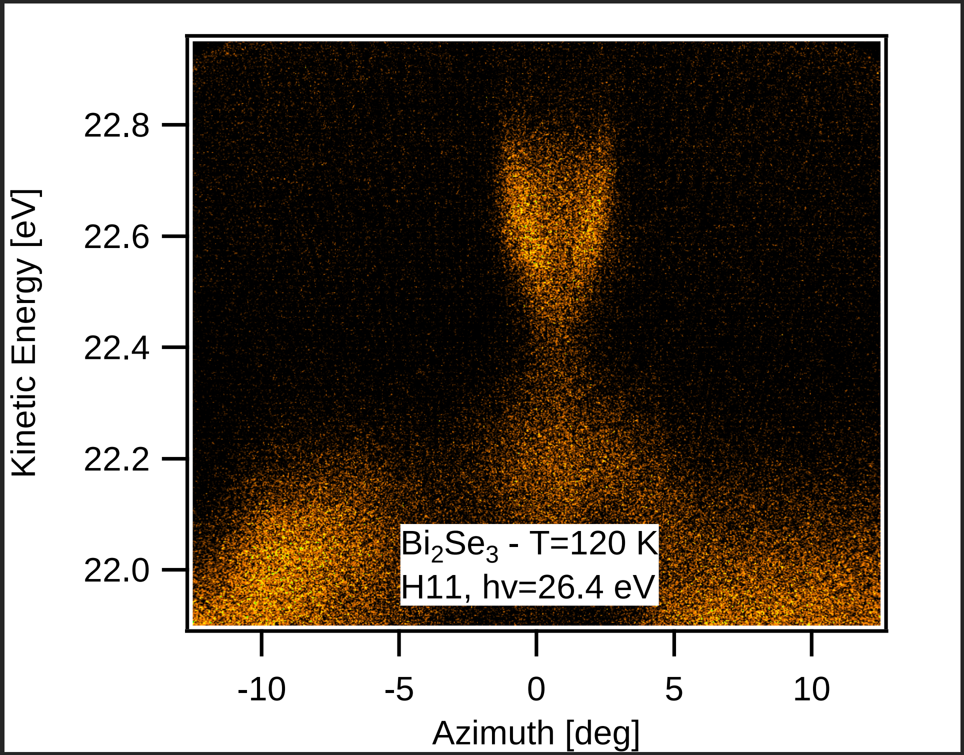


3) HHG: latest developments, T-ReX @ FERMI, first results



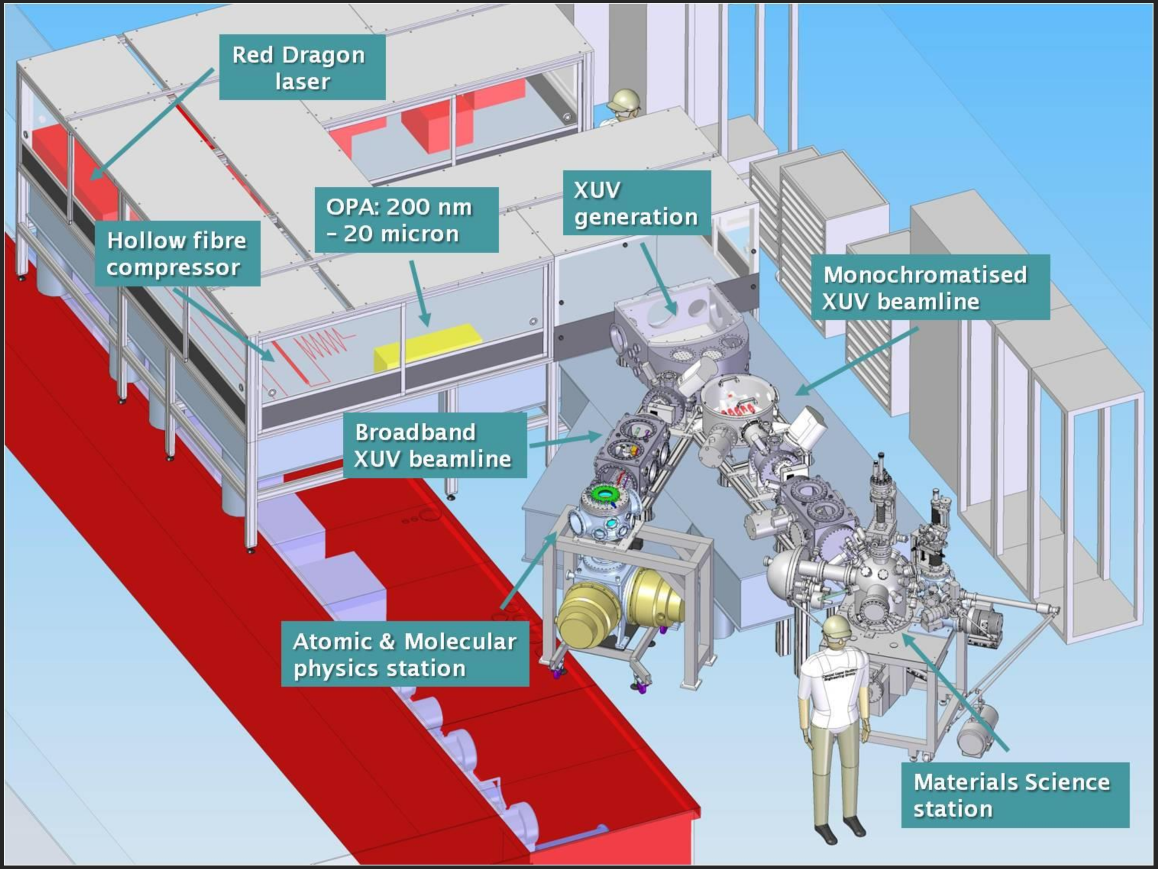


First ARPES map on a TI



At 50 kHz, we produce >10^{12} photons/s

3) HHG: The ARTEMIS facility



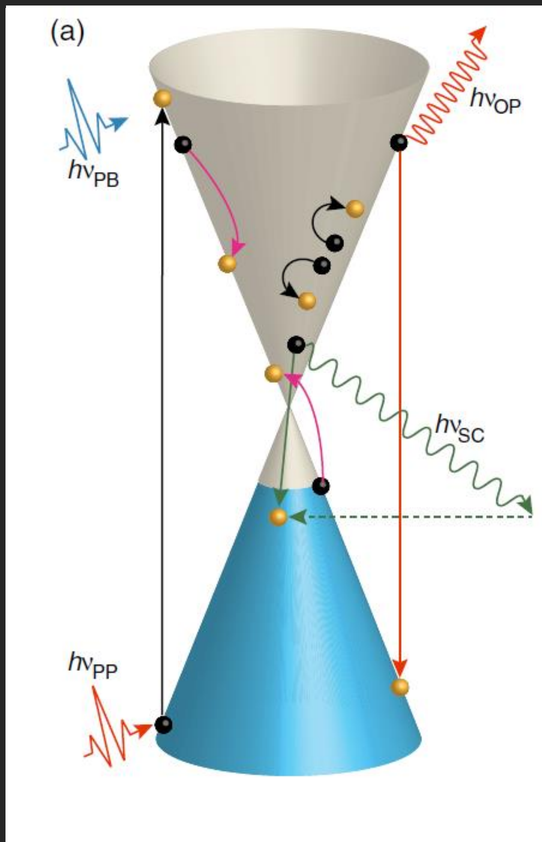
F. Frassetto et al., OE 19, 19169 (2011)

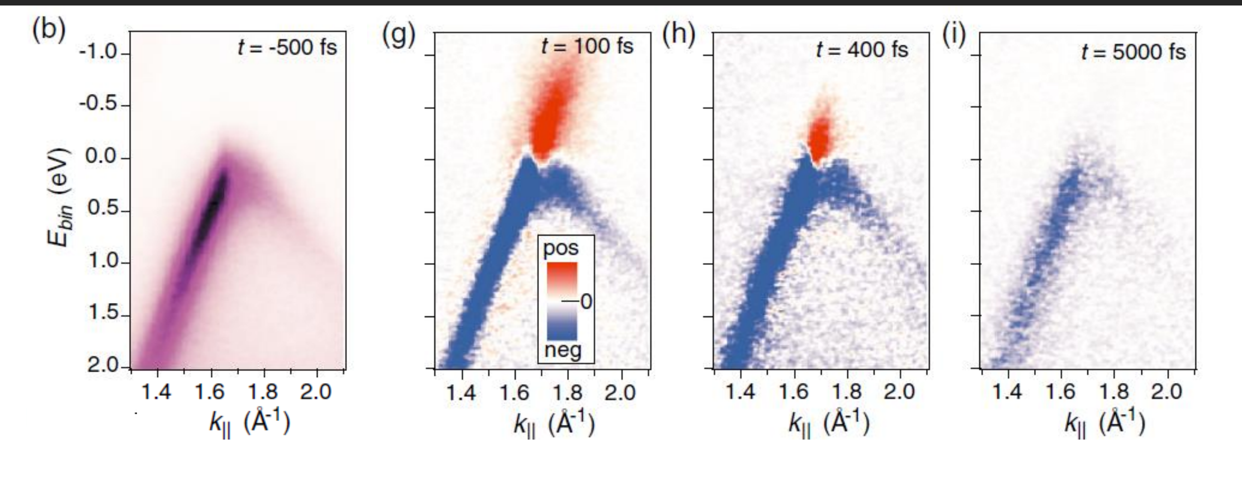
- XUV probe in the 15-50 eV range: no constraints on $k_{||}$, E_B
- Energy resolution (XUV bandwidth): ~150-300 meV; time res.: ~40 fs
- Fully tunable pump pulse (190-20000 nm)
- 1 kHz repetition rate (~1000 e^- counts/s)

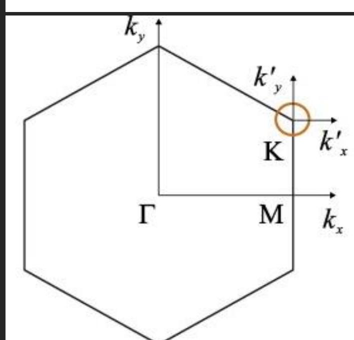
3) HHG TR-ARPES on Graphene

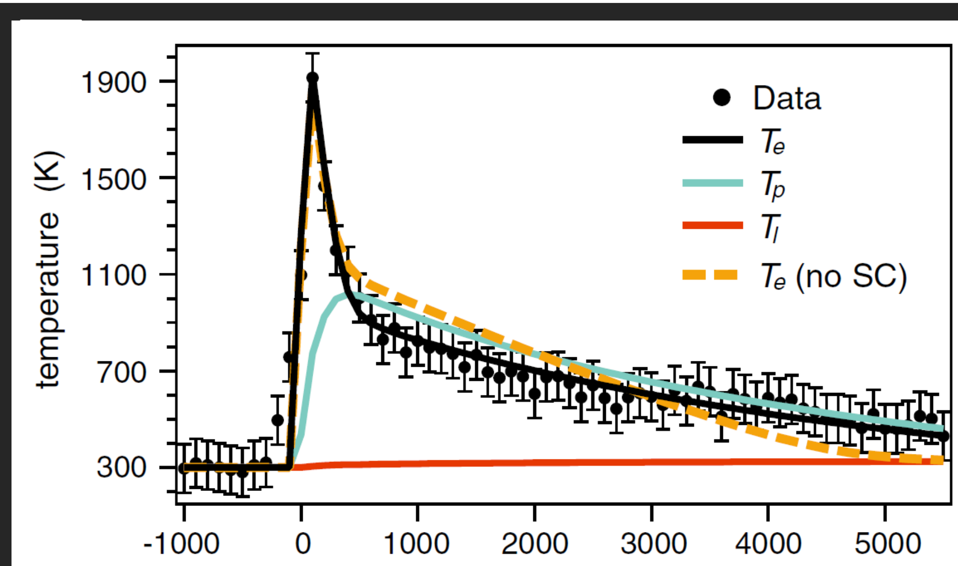
Results on <u>Monolayer</u> Graphene

hν=31.5 eV





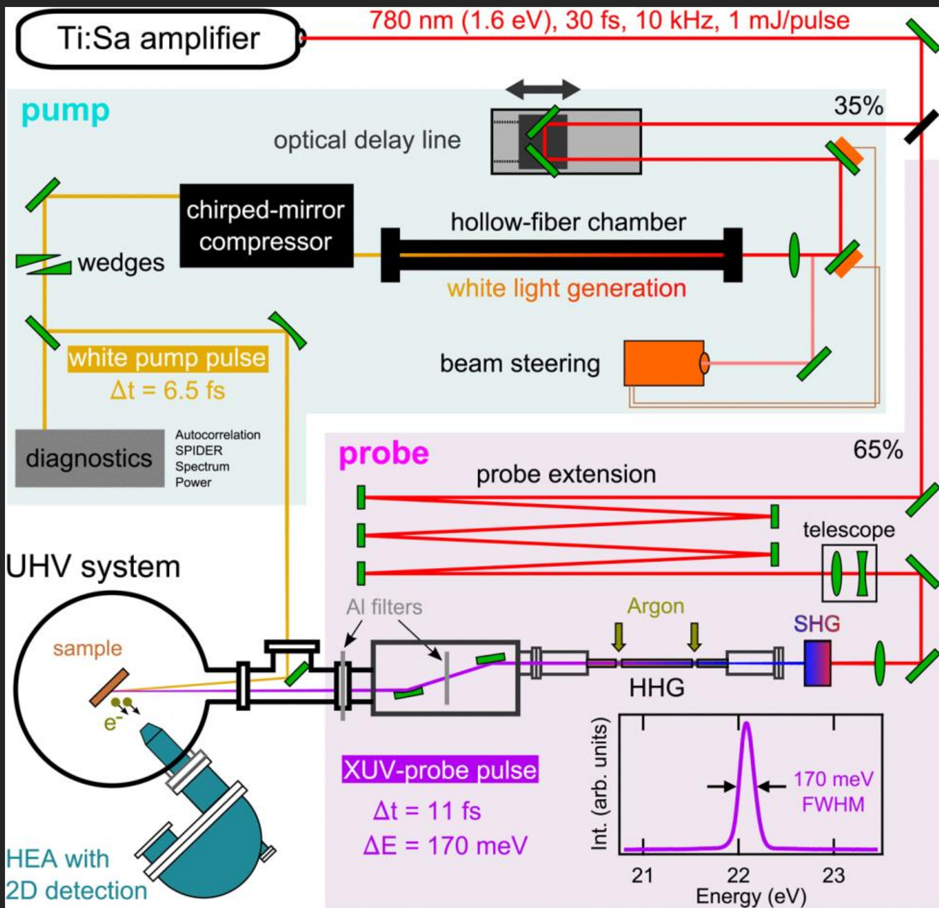




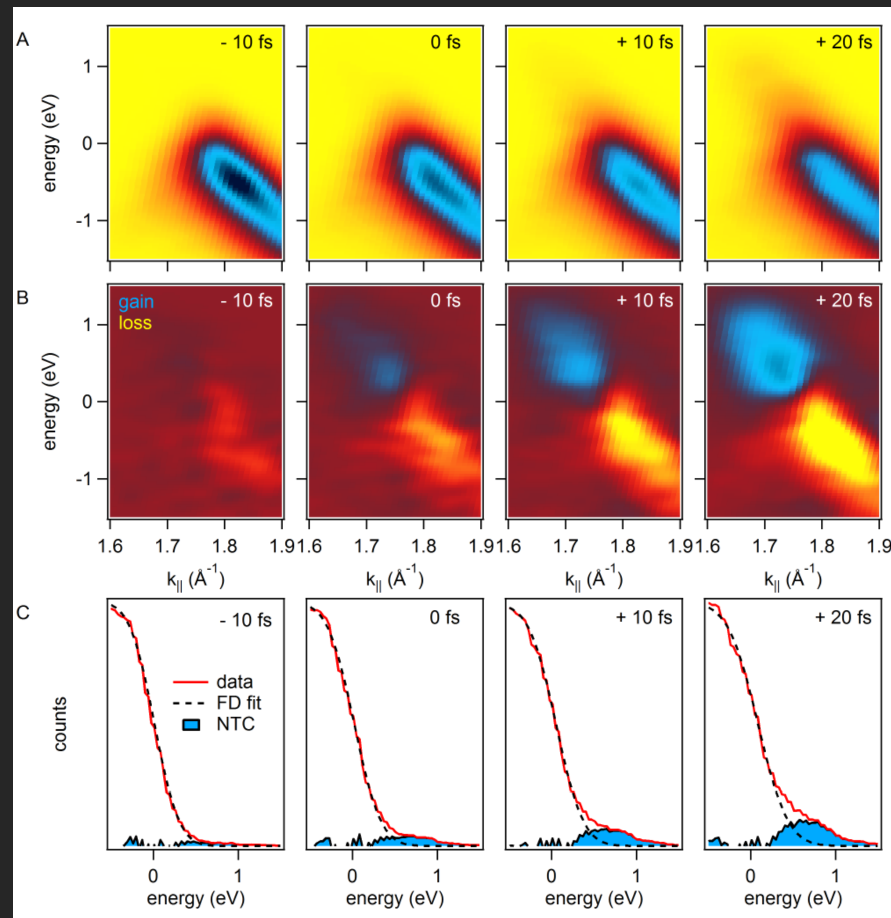
Studies on Graphene at ARTEMIS:

<u>Monolayer</u>: PRL 111, 027403 (2013), <u>Bilayer</u>: PRL 112, 257401 (2014), <u>n- vs p-dop</u>: NanoLett. 15, 326 (2015)

3) HHG TR-ARPES on Graphene, ultrashort timescales



G. Rhode et al., RSI **87**, 103102 (2016)

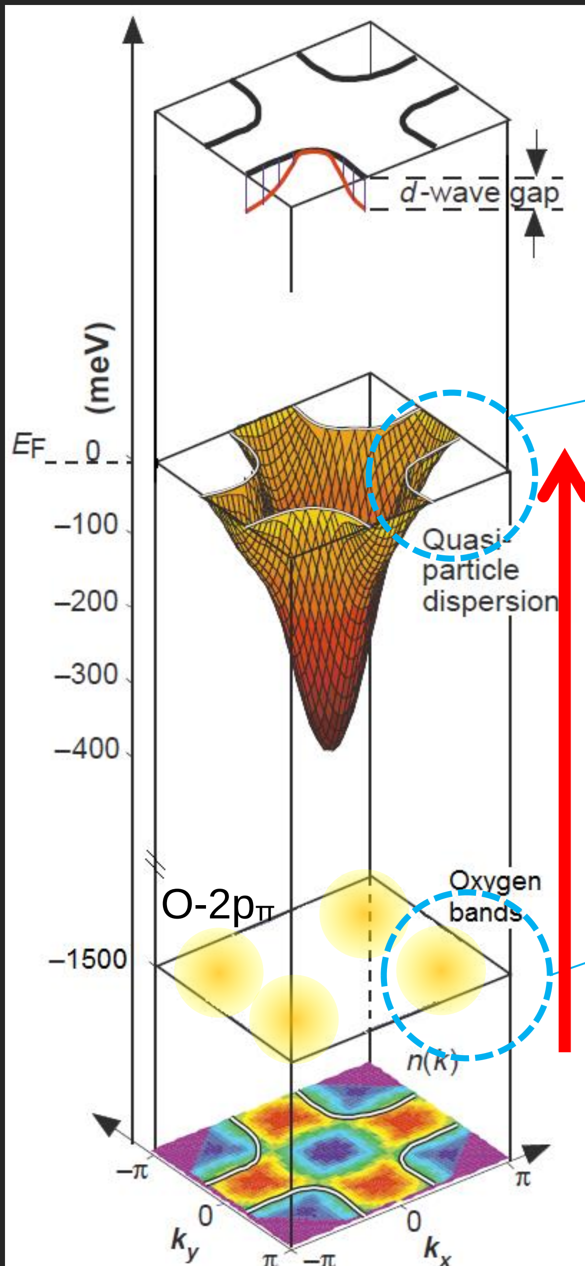


I. Gierz et al., PRL **115**, 086803 (2015)

PP cross-correlation: 13 fs
HHG: 11 fs duration, BW≈170 meV: very close to transform limit.

Dynamics of Graphene on <10 fs timescale.

3) HHG TR-ARPES on Cuprate Superconductors



Adapted from F. Ronning et al., Science 282, 2067 (1998)

i) Probe of the Fermi surface

ii) Probe of the Oxygen bands

<u>Pump</u>: 1.65 eV, 400 μJ/cm^2

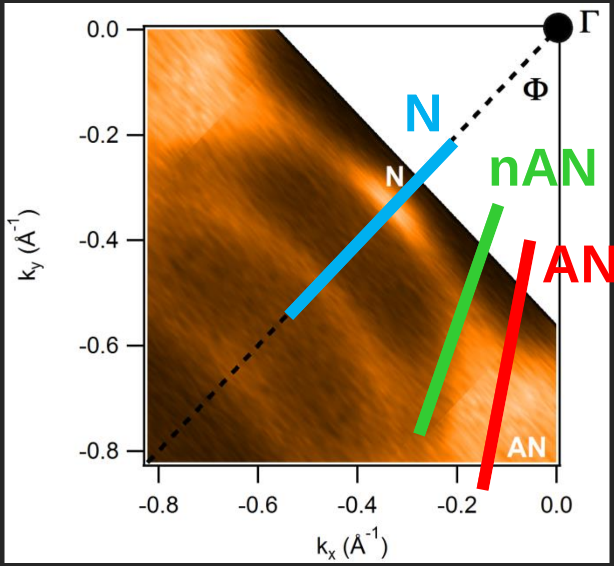
<u>Probe</u>: XUV ≈18 eV, s-pol

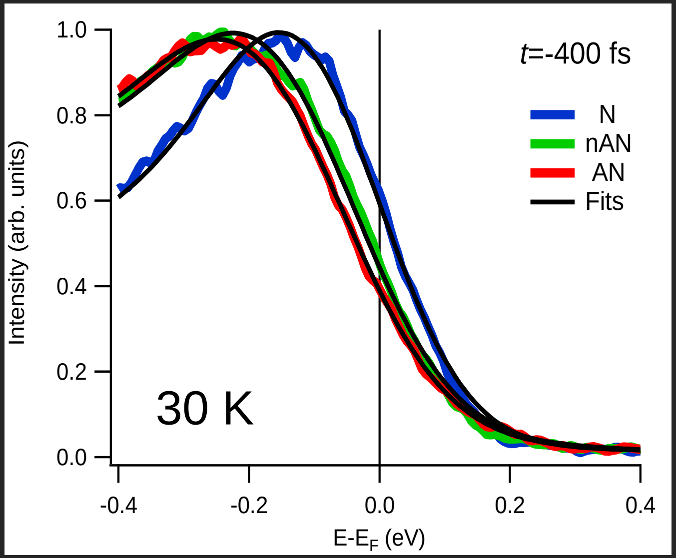
3) HHG TR-ARPES on Cuprate Superconductors

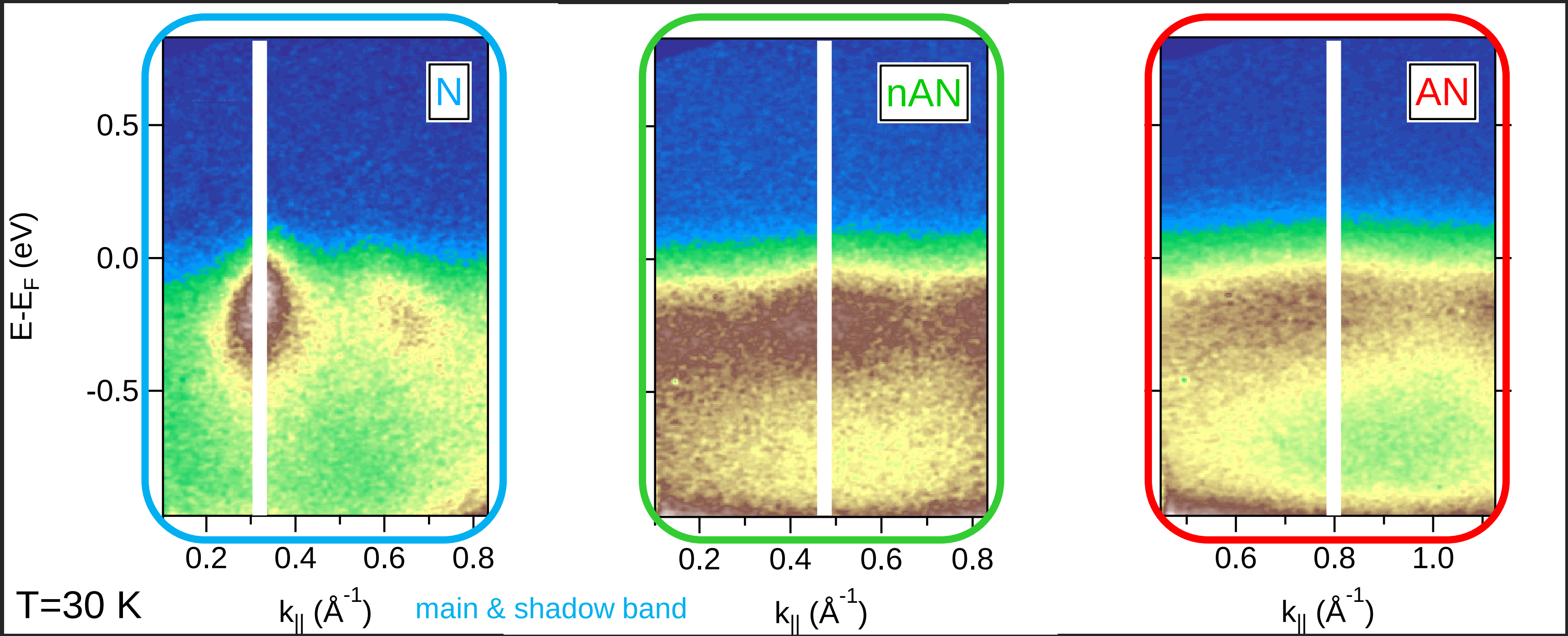
$Bi_2Sr_2Ca_{0.92}Y_{0.08}Cu_2O_{8+\delta}$
(T_c=96 K)

Three k-space directions:
* Nodal (N)
* nearAntiNodal (nAN)
* AntiNodal (AN)

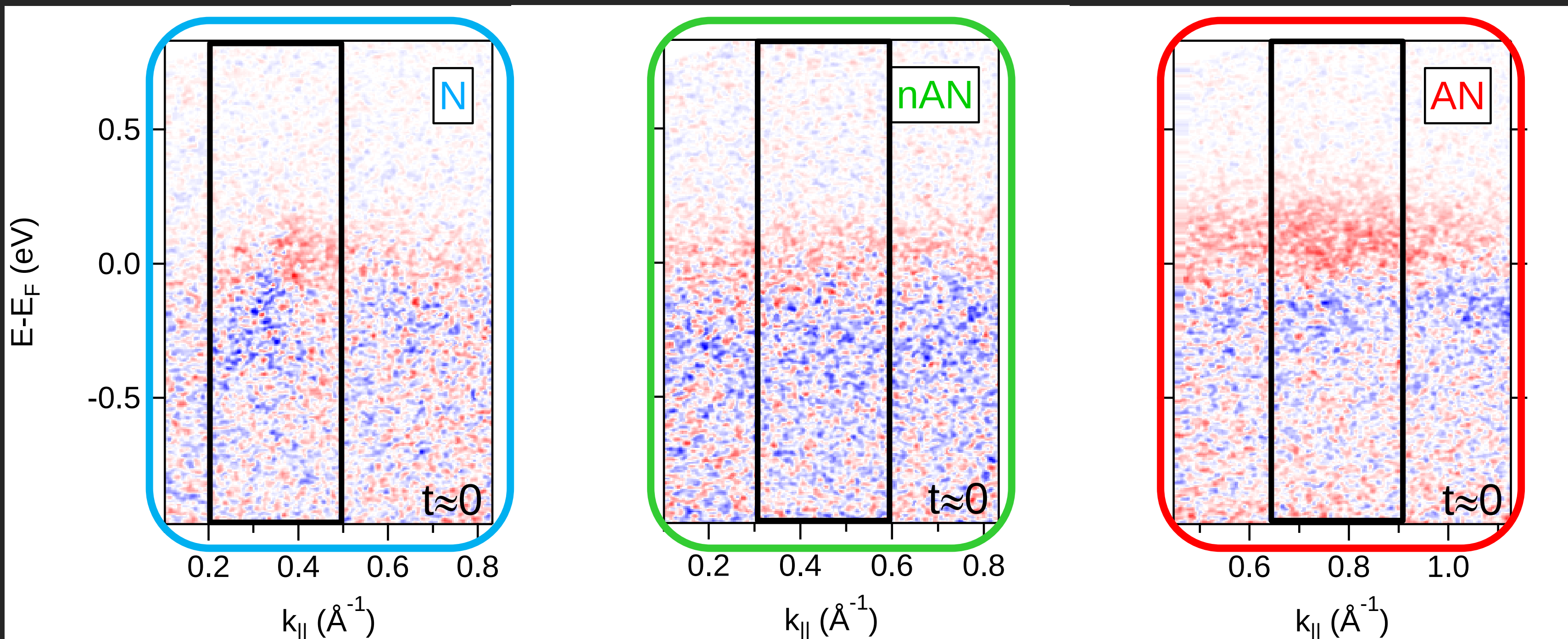
F. Cilento et al., Science Advances 4, eaar1998 (2018)

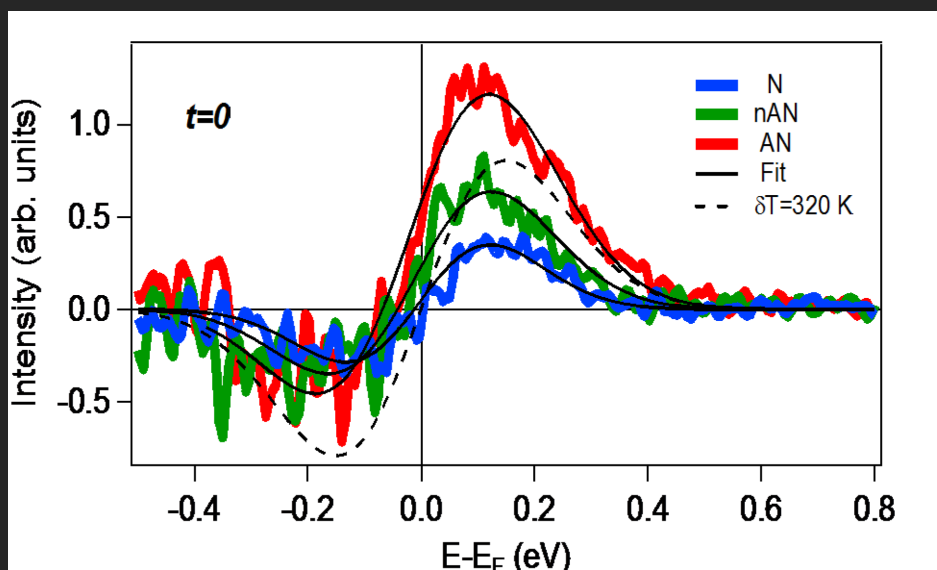






3) HHG TR-ARPES on Cuprate Superconductors

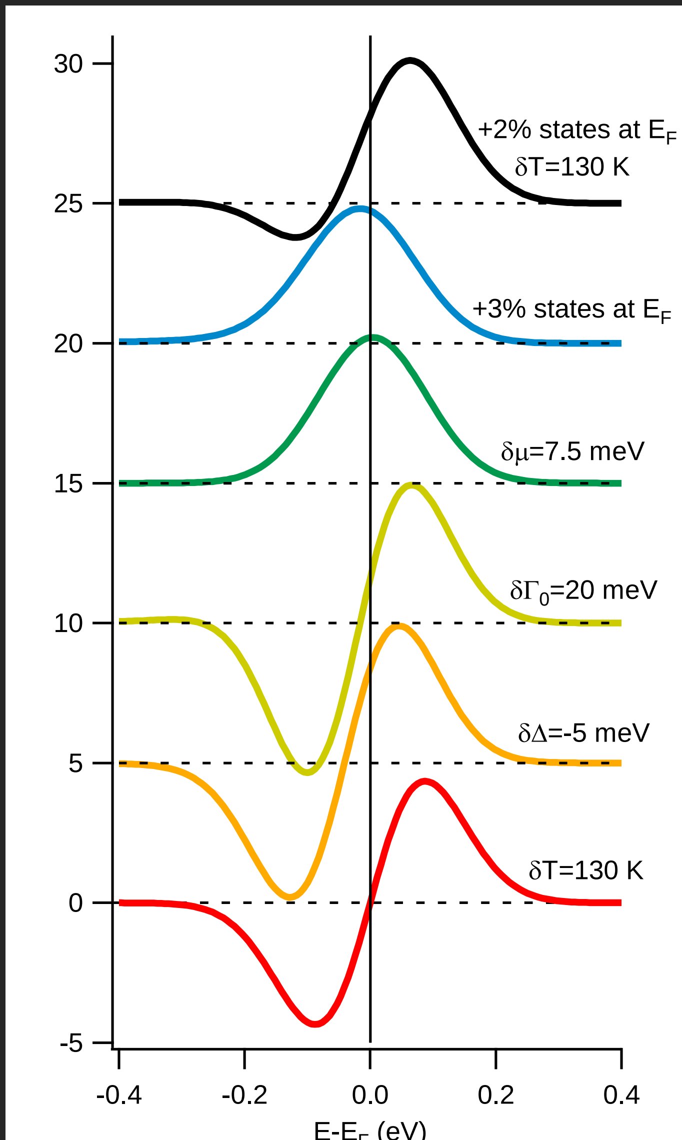


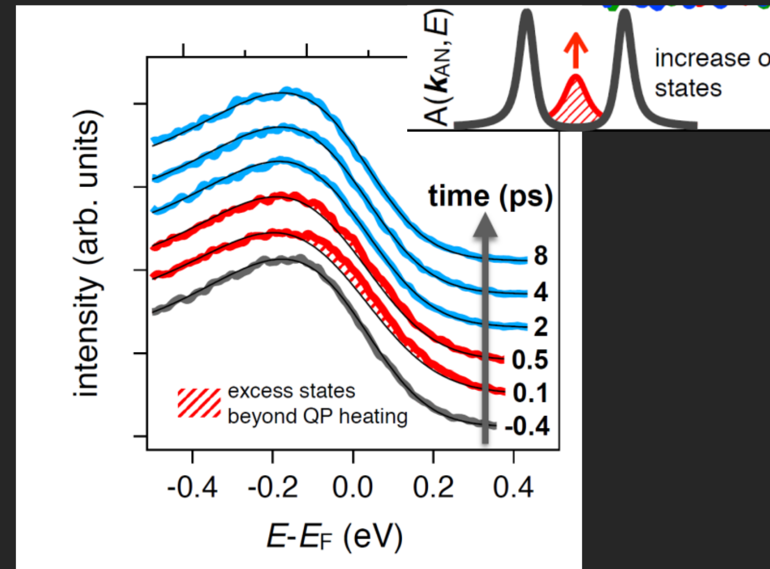


3) HHG TR-ARPES on Cuprate Superconductors

Simulation of differential EDCs

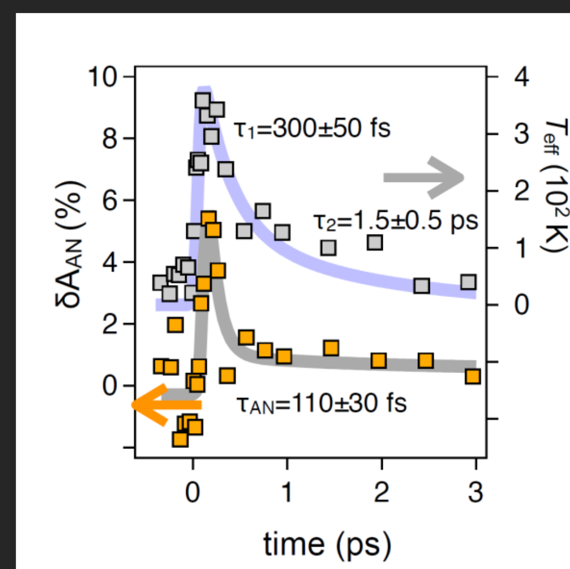
Σ by Norman (et al., PRB 57, 11093, 1998)





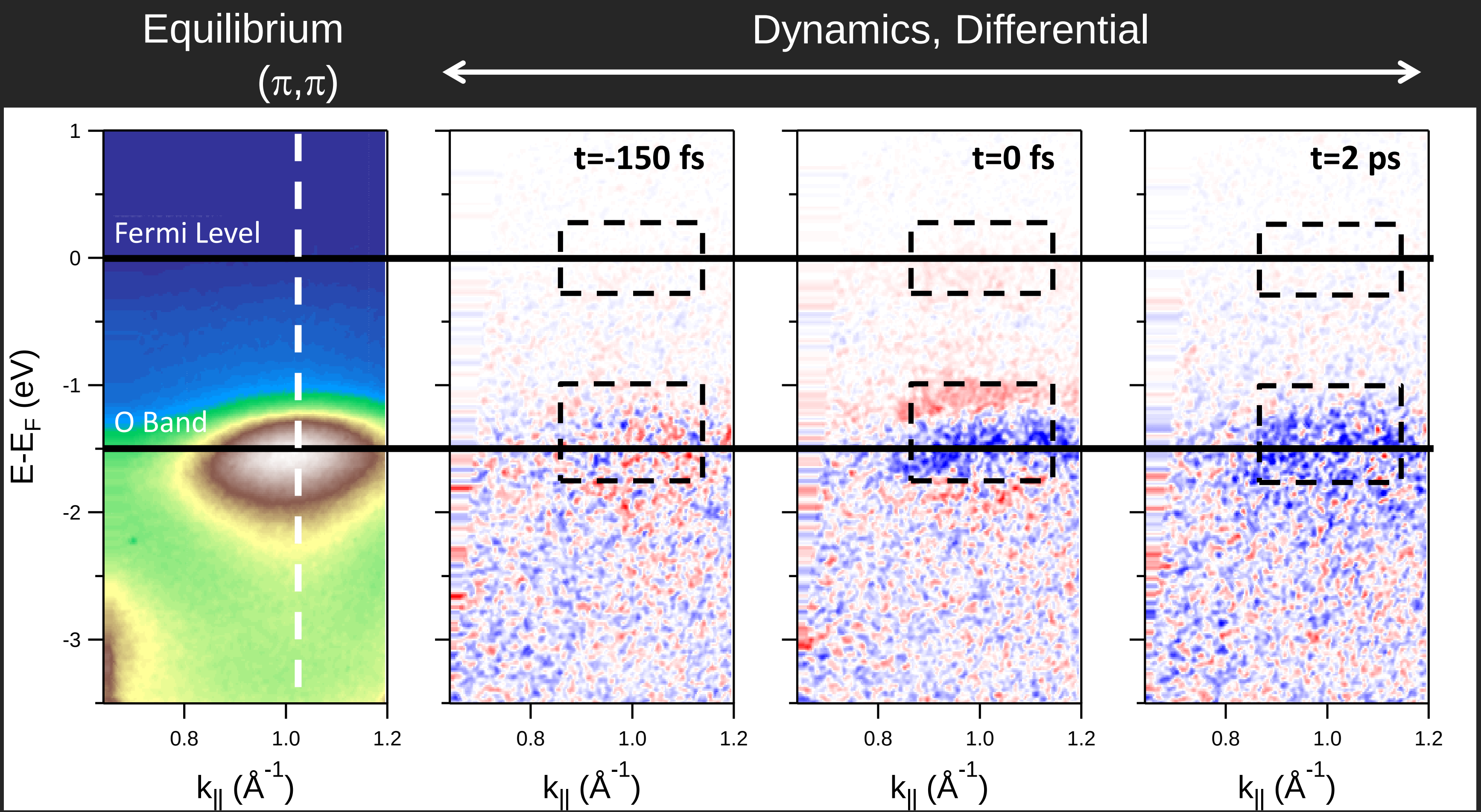
EDCs at AntiNode

Excess AN states, %



T_e at Node

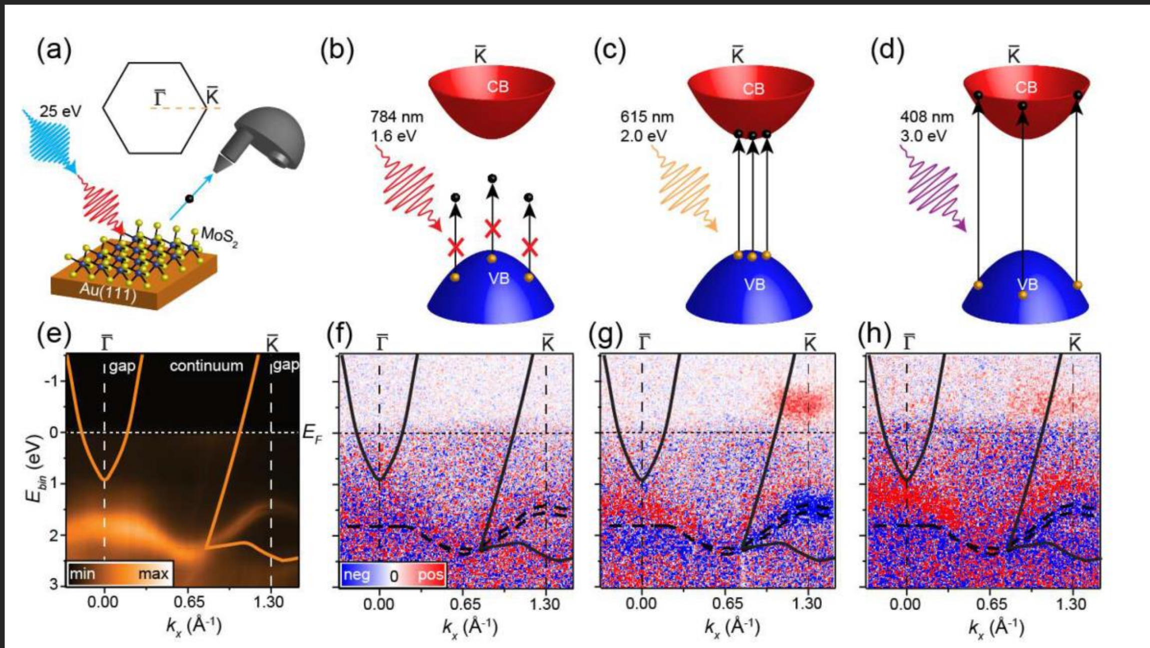
3) HHG TR-ARPES on Cuprate Superconductors



Two effects are found, showing different dynamics:

* t≈0: filling of the empty states at Fermi and broadening of the 1.5 eV peak
* t>1 ps: change of the spectral weight of the 1.5 eV peak

3) HHG TR-ARPES on TMDCs



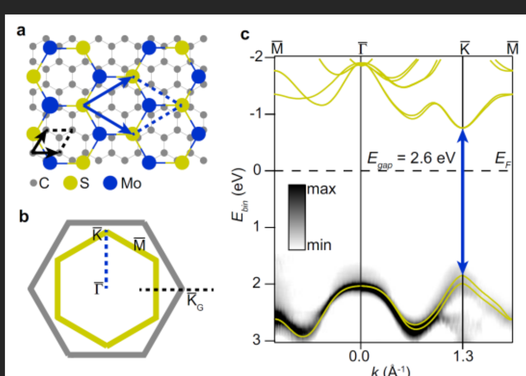
A. G. Čabo et al., Nano Lett. 15, 5883 (2015)

<u>TR-ARPES on MoS_2 single-layer</u>

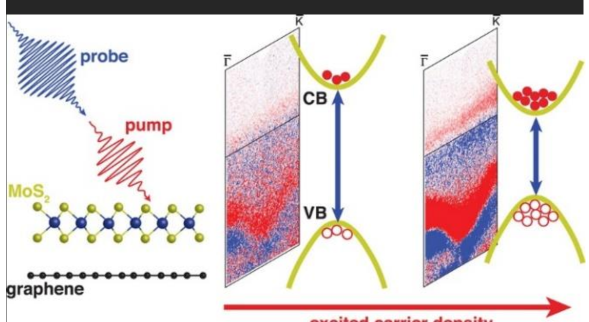
Use of a IR-VIS-UV tunable-pump to populate and visualize the gap.

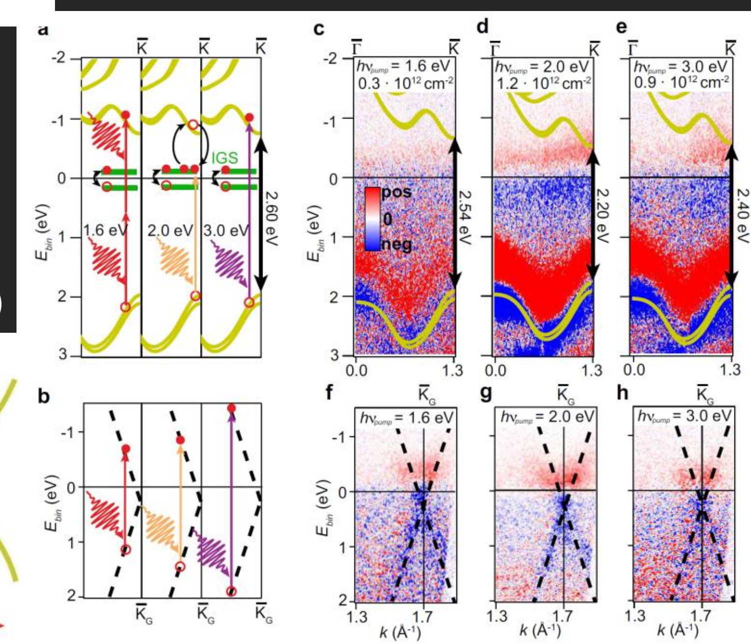
A direct quasiparticle band gap of 1.95 eV has been revealed.

MoS_2/Graphene heterostructure: band structure can be significantly altered by screening effects. A 0.4 eV renormalization of the gap can be induced.



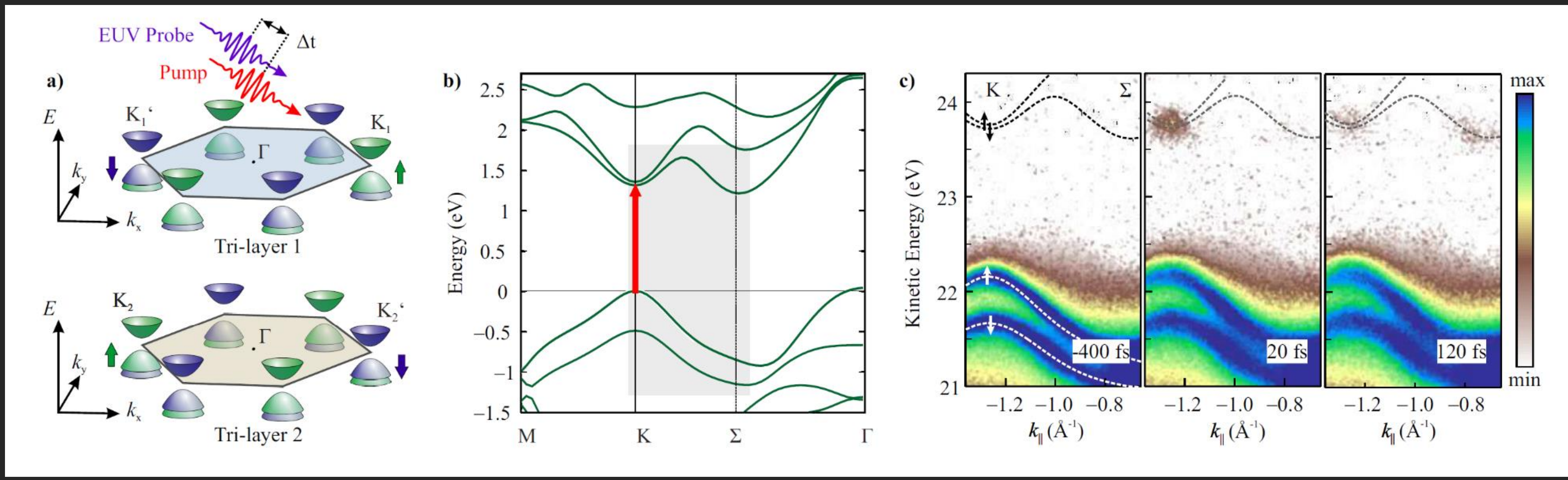
S. Ulstrup et al., ACS Nano 10, 6315 (2016)

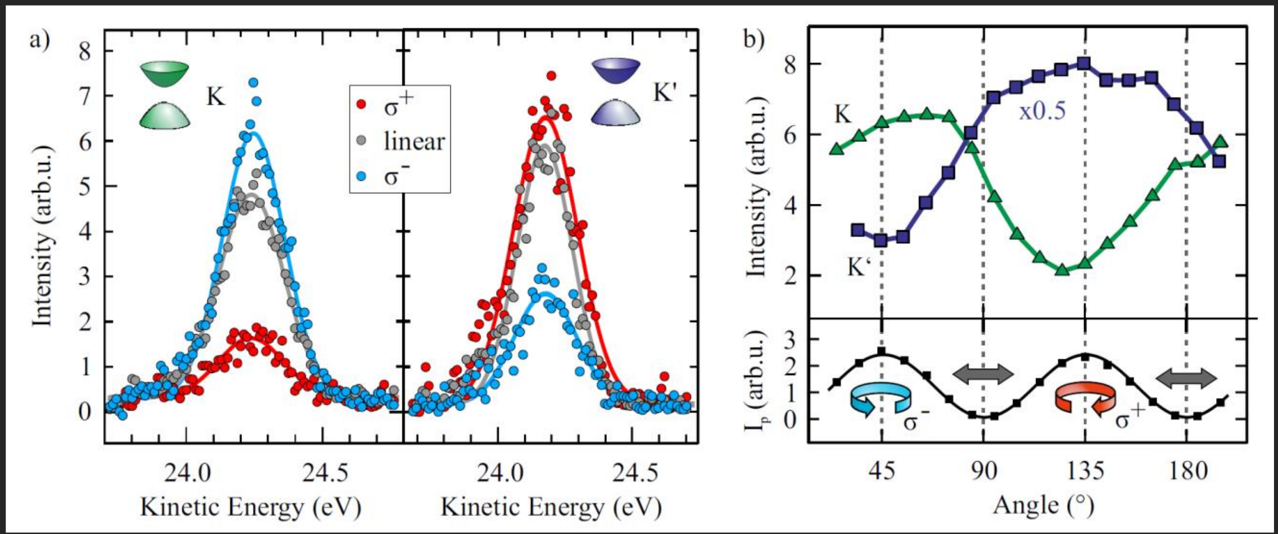




3) HHG TR-ARPES on TMDCs

WSe_2 TMDC measured at hν=23 eV. Pump: 1.5 eV, *few* mJ/cm^2



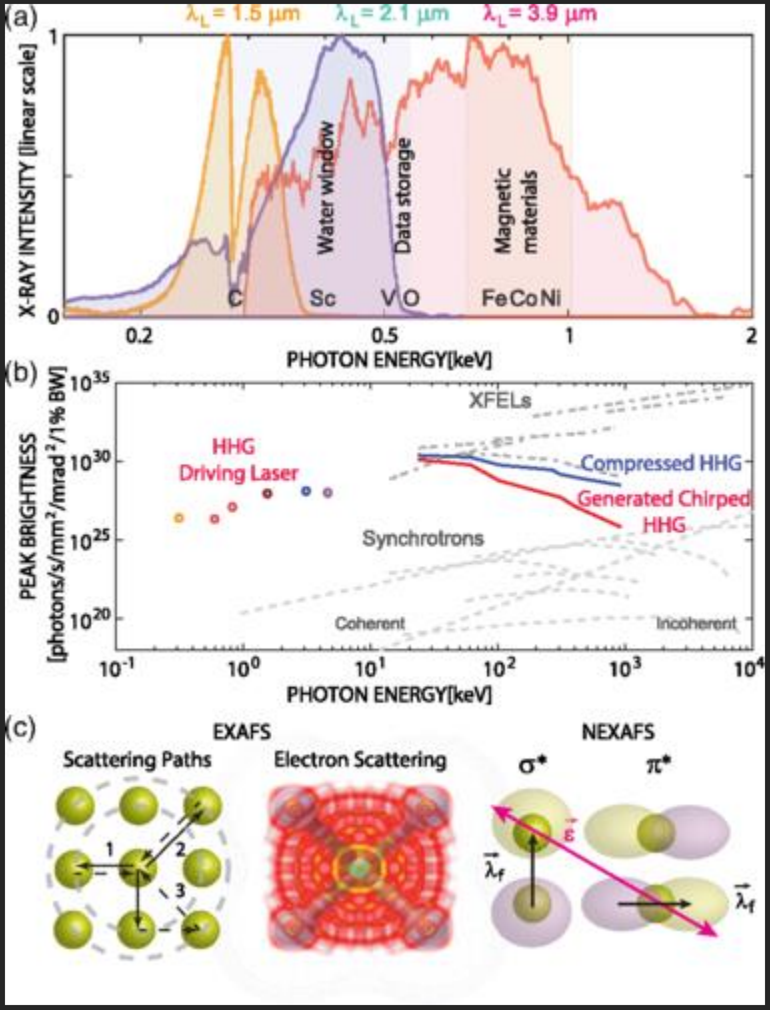


R. Bertoni et al., PRL **117**, 277201 (2016)

Selectivity of excitation of K, K' valleys obtained by pumping with CL, CR light

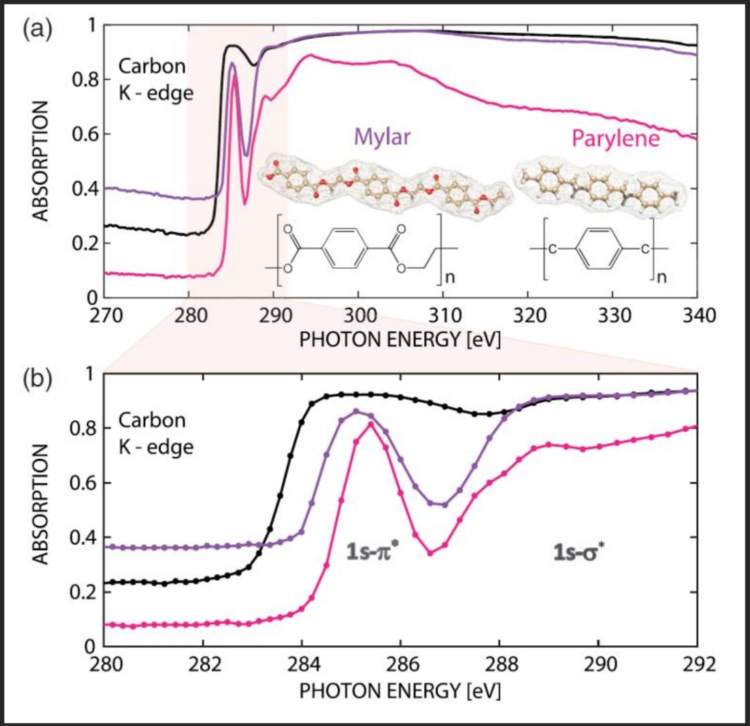
3) HHG: latest developments

After showing the latest developments in the direction of high repetition rate beamlines, we consider developments where high photon energies are produced

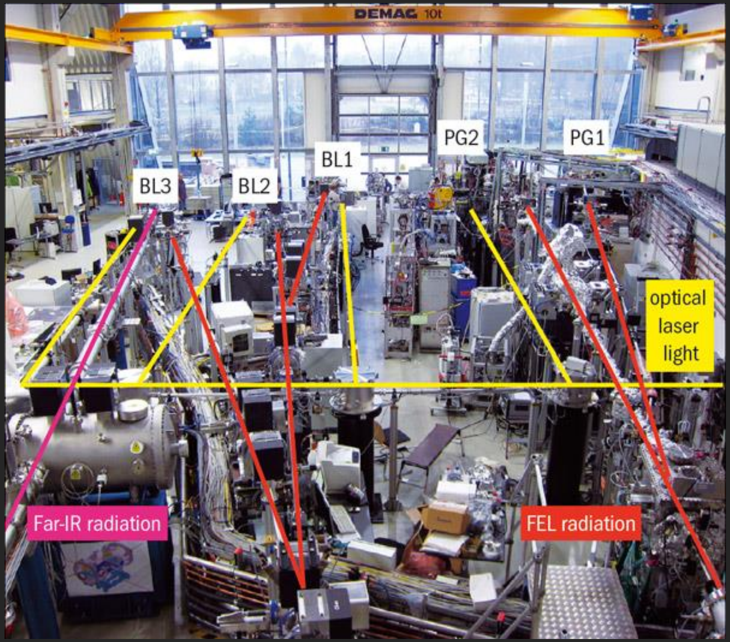


D. Popmintchev et al., PRL 120, 093002 (2018)

First demonstration of a table top setup producing coherent beams with bandwidth spanning from the ultraviolet up to x-ray photon energies >1.6 keV. This light has been used for x-ray absorption spectroscopy at the K- and L-absorption edges of solids.

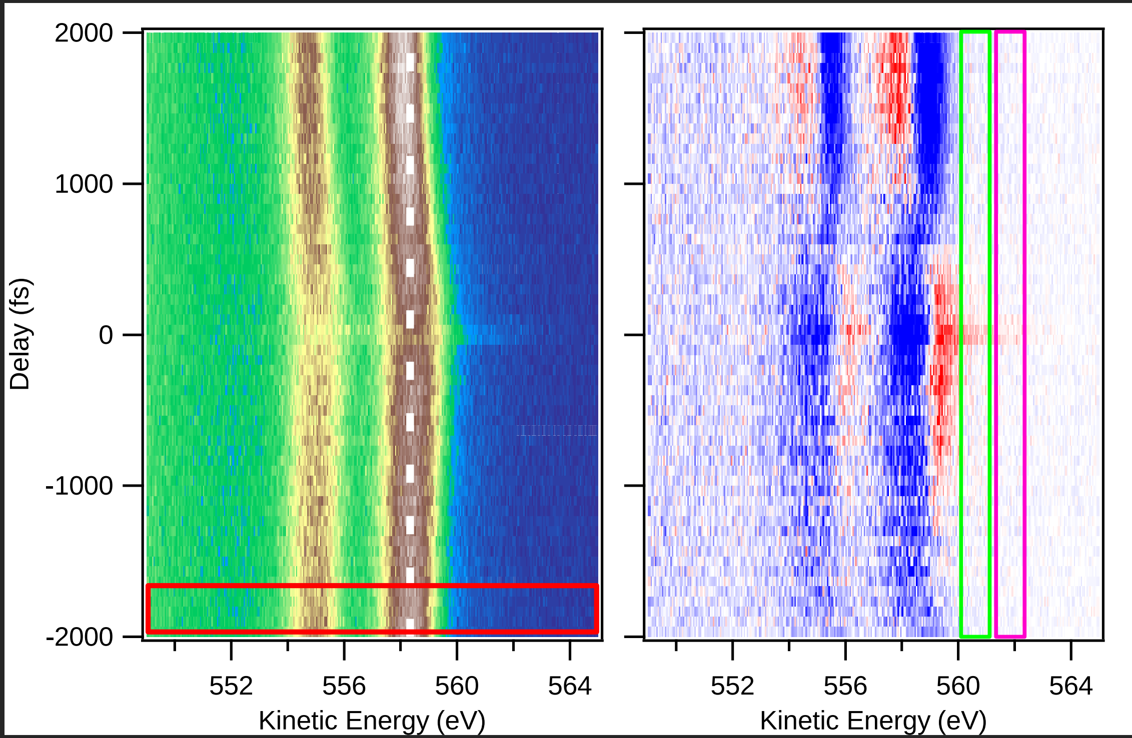


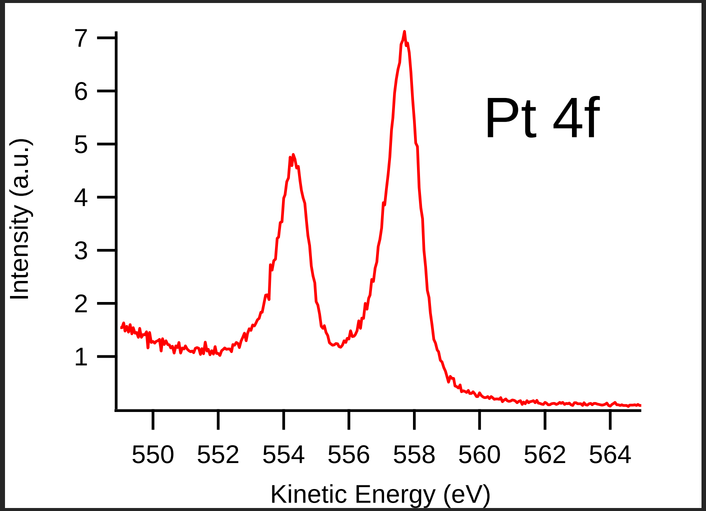
TR-PES at a FEL: results from FLASH

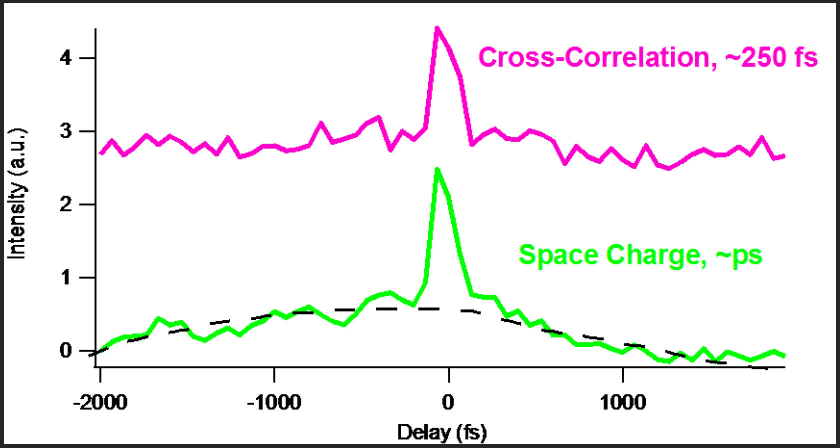


Time-resolved PES on Pt 4f core levels:

finding t0 and determine cross-correlation

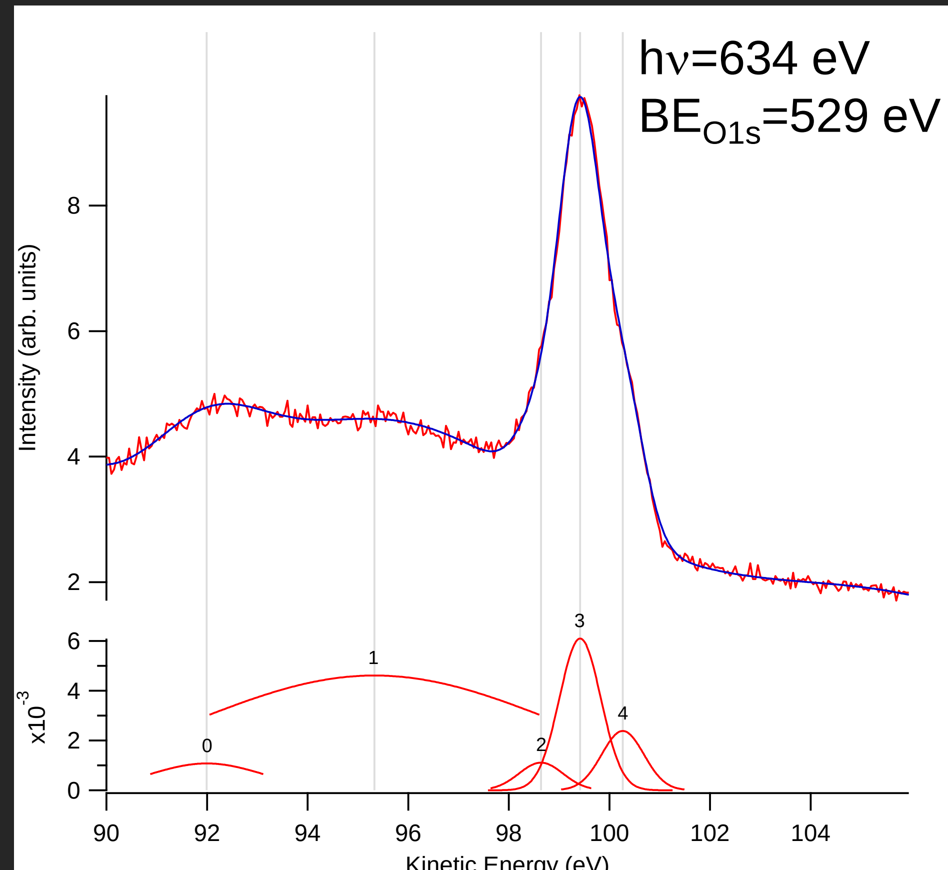


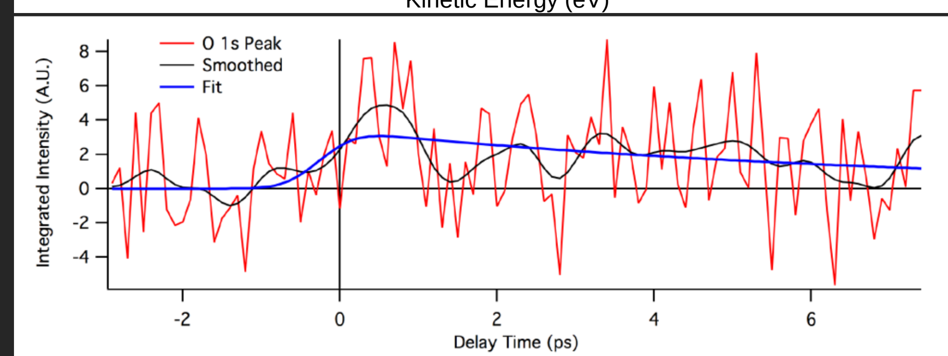


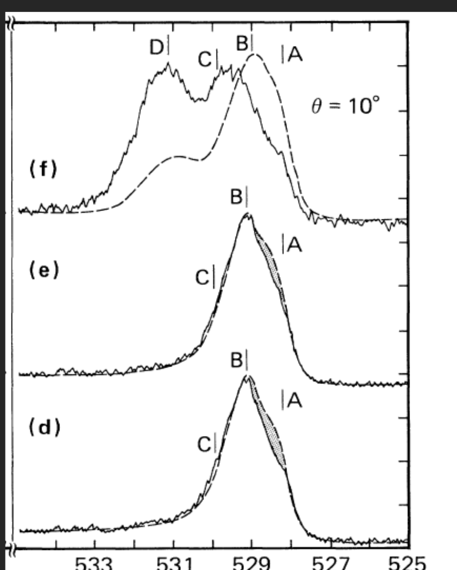


TR-PES at a FEL: results from FLASH

Measurement of O 1s core level in Bi2212







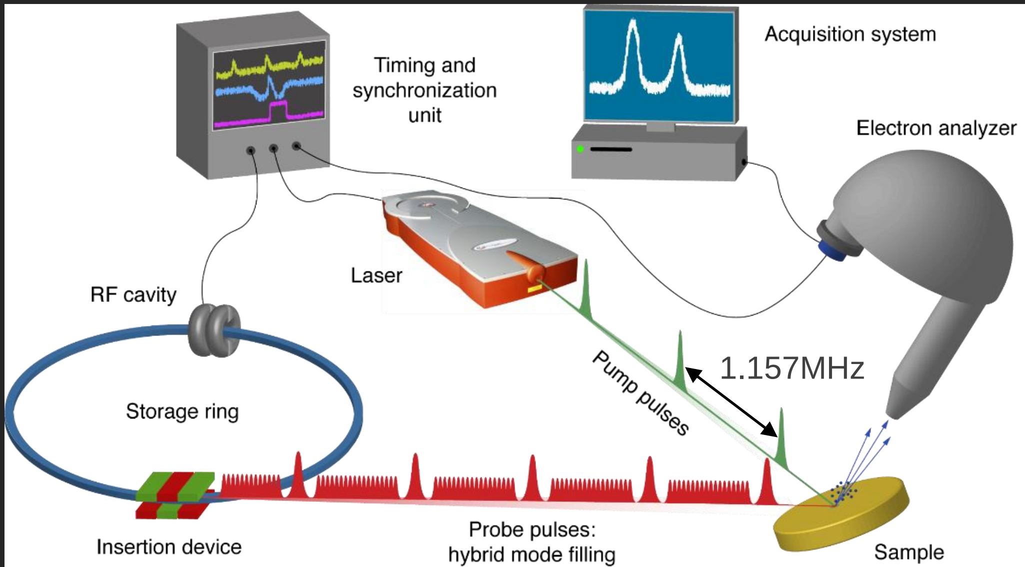
PRB 83, 3085 (1991)

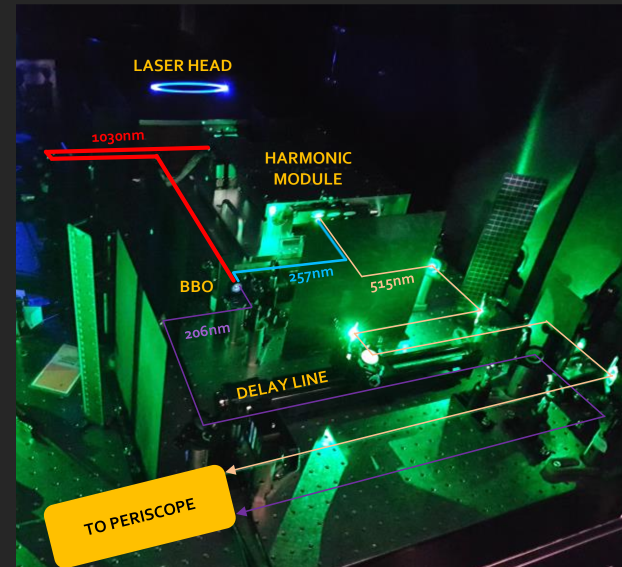
4 kHz effective rep. rate

Pump: 1.5 eV, 1 mJ/cm^2

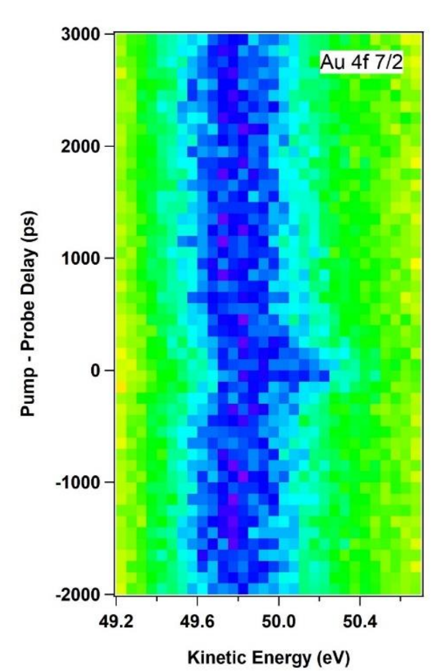
Resolution: 250 fs

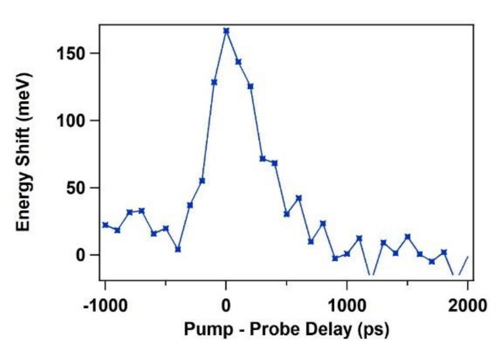
TR-PES at a Synchrotron: results from ALOISA BL, Sundyn endstation



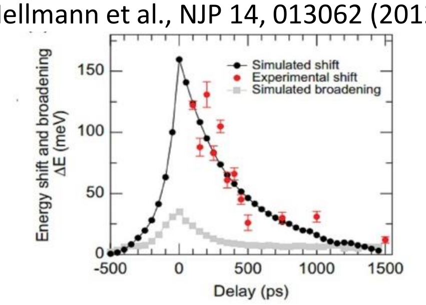


Pictures are a courtesy of M. Dell'Angela

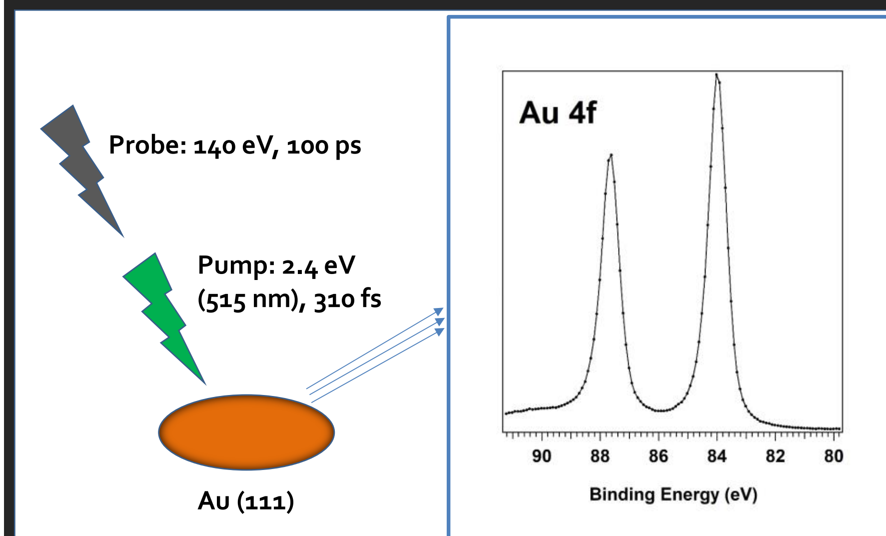




S. Hellmann et al., NJP 14, 013062 (2012)



Find t0 with space charge effect



Conclusions

Table-top spectroscopies are assuming a fundamental role for the study of the non-equilibrium dynamics of materials.

They are complementary to large-scale facilities like Synchrotrons and FELs, and are competitive when:

- Low intensities/pulse are needed
- High average flux is needed
- Ultrashort timescales are needed

HHG methods are pushing the flux (rep. rate), the photon energy and the timescales to extreme values (MHz, keV, as), and will constitute a breakthrough in research.

Conventional, laser based spectroscopies (optical and photoelectronic) are important to investigate the ground state properties of material and to achieve the control of matter on ultrashort timescales.