Elettra Sincrotrone Trieste

Drafting the Science Case for Ultrafast Laser-based experiments with EUV Soft X-Ray Photons

Fulvio Parmigiani

University of Trieste
Elettra Sincrotrone Trieste

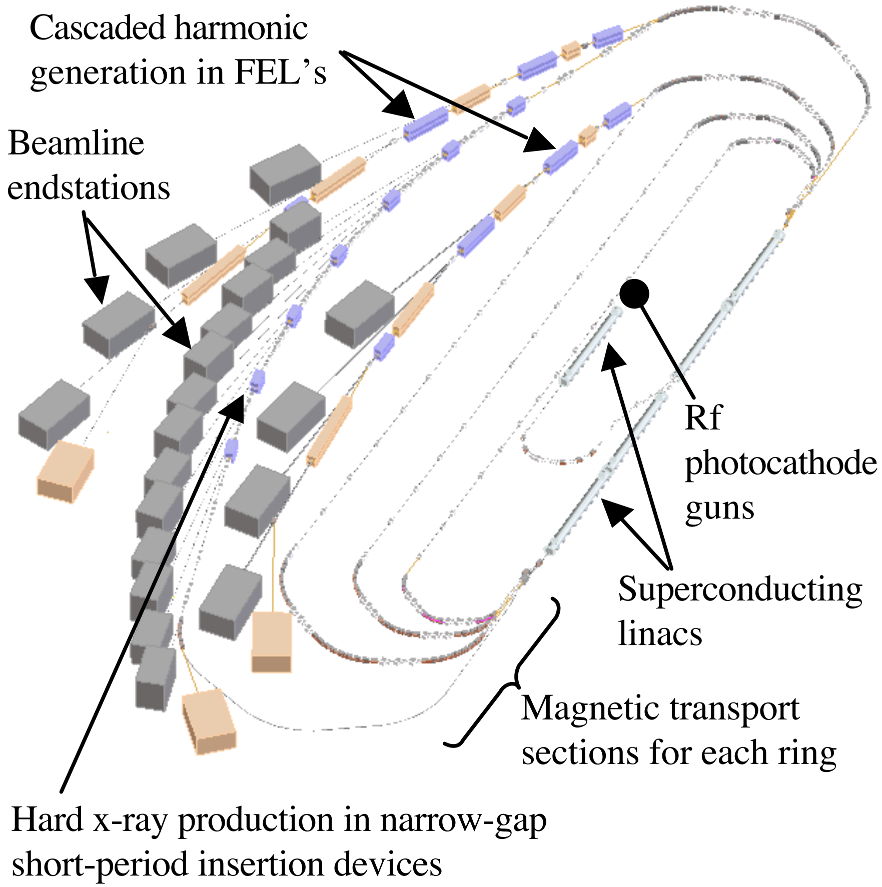
THE EARLY TIMES

LUX

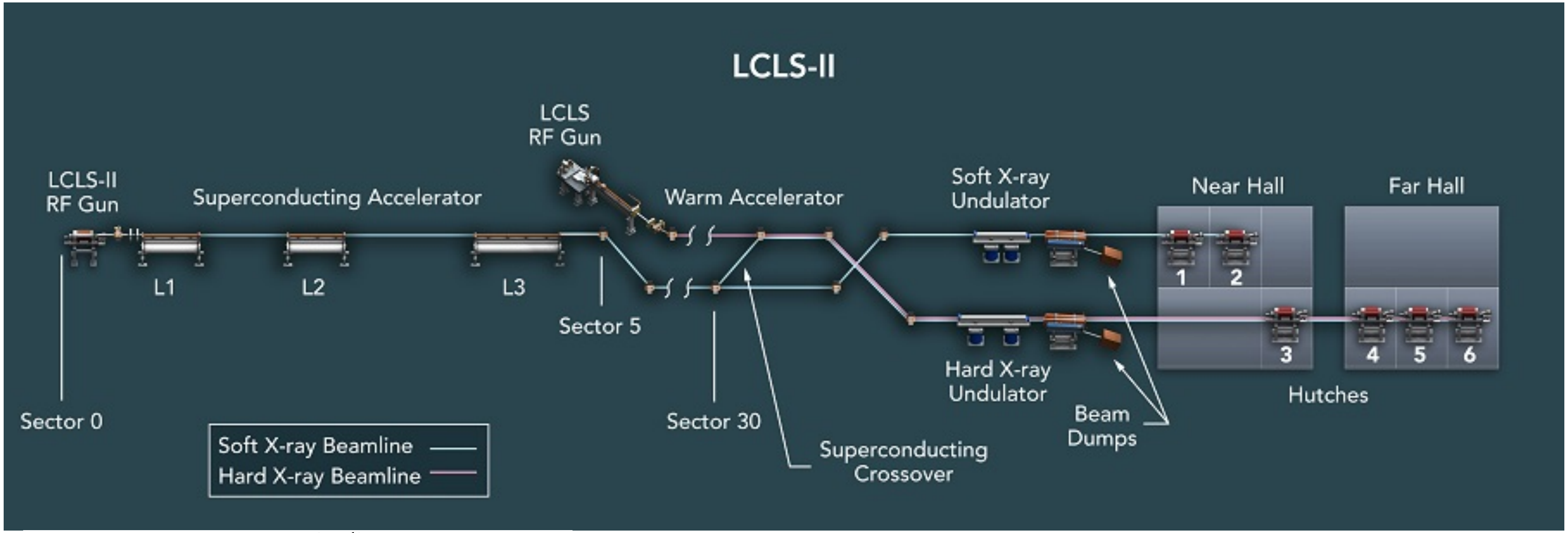
Proceedings of the 2003 Particle Accelerator Conference

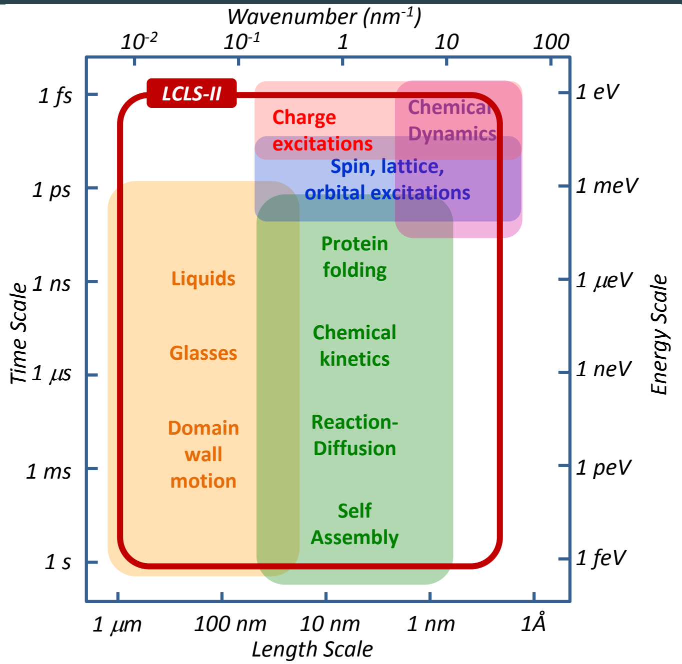
A RECIRCULATING LINAC-BASED FACILITY FOR ULTRAFAST X-RAY SCIENCE*

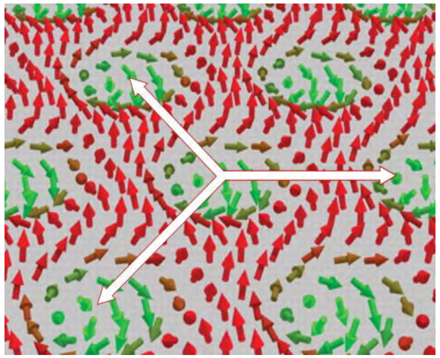
J. N. Corlett, W. A. Barletta, S. DeSantis, L. Doolittle, W. M. Fawley, M.A. Green, P. Heimann, S. Leone, S. Lidia, D. Li, A. Ratti, K. Robinson, R. Schoenlein, J. Staples, W. Wan, R. Wells, A.Wolski, and A. Zholents, LBNL, Berkeley, California, USA; F. Parmigiani, UCSC, Brecia, Italy; M. Placidi, W. Pirkl, Geneva, Switzerland; R. A. Rimmer, Thomas Jefferson National Accelerator Facility, Virginia, U.S.A., S. Wang, Indiana University, U.S.A.

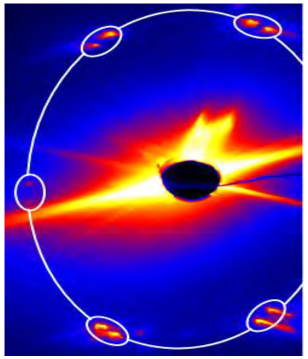


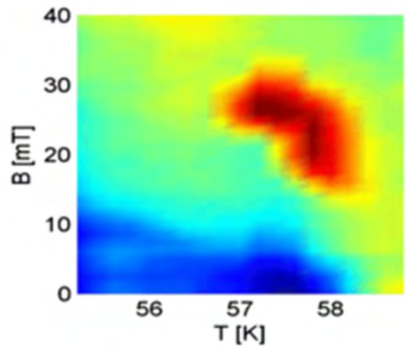
LCLS II


LCLS-II
LCLS RF Gun
LCLS-II RF Gun
Superconducting Accelerator
Warm Accelerator
Soft X-ray Undulator
Near Hall
Far Hall
1
2
L1
L2
L3
Sector 5
3
4
5
6
Hard X-ray Undulator
Hutches
Beam Dumps
Sector 0
Sector 30
Soft X-ray Beamline
Hard X-ray Beamline
Superconducting Crossover

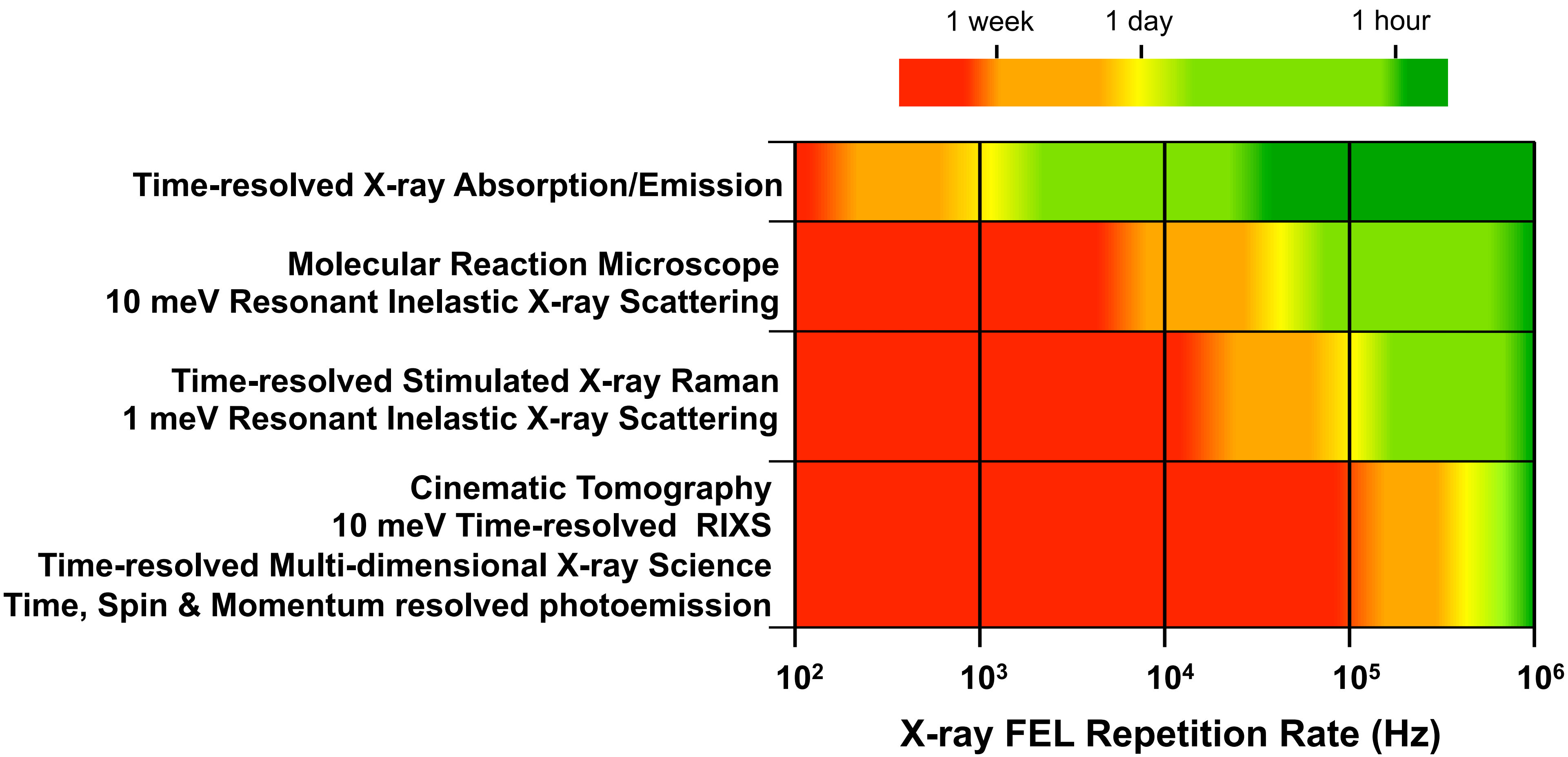

Wavenumber (nm⁻¹)
10⁻²
10⁻¹
1
10
100
LCLS-II
Charge excitations
Chemical Dynamics
Spin, lattice, orbital excitations
Protein folding
Liquids
Glasses
Chemical kinetics
Domain wall motion
Reaction-Diffusion
Self Assembly
1 fs
1 ps
1 ns
1 μs
1 ms
1 s
Time Scale
1 eV
1 meV
1 μeV
1 neV
1 peV
1 feV
Energy Scale
1 μm
100 nm
10 nm
1 nm
1Å
Length Scale





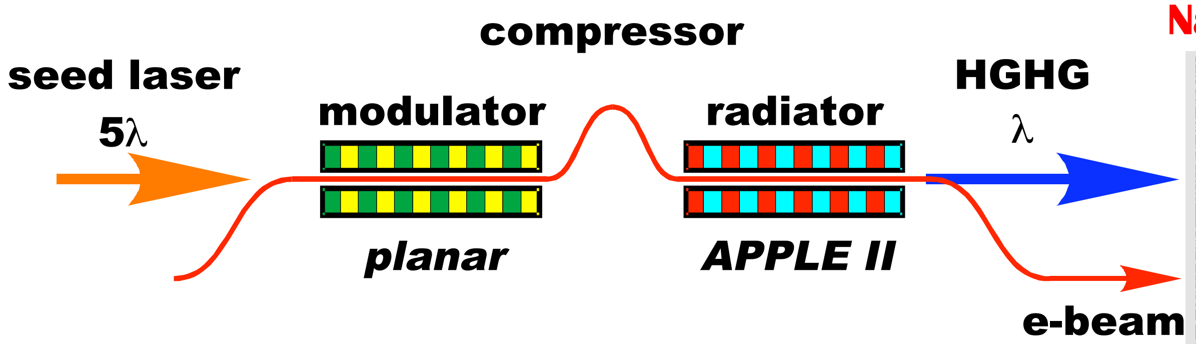

40
30
20
10
0
B [mT]
56
57
58
T [K]

THE PHOTON PULSE ISSUE

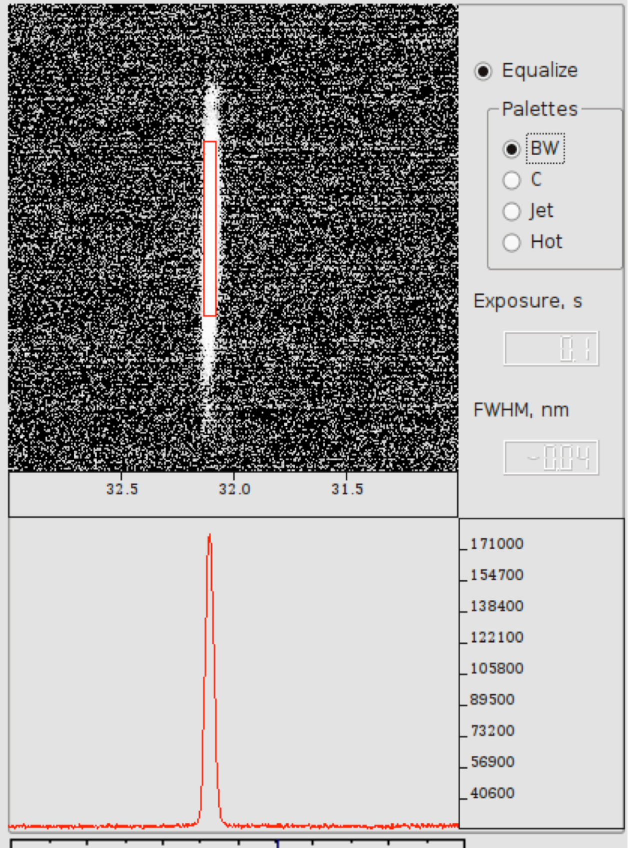


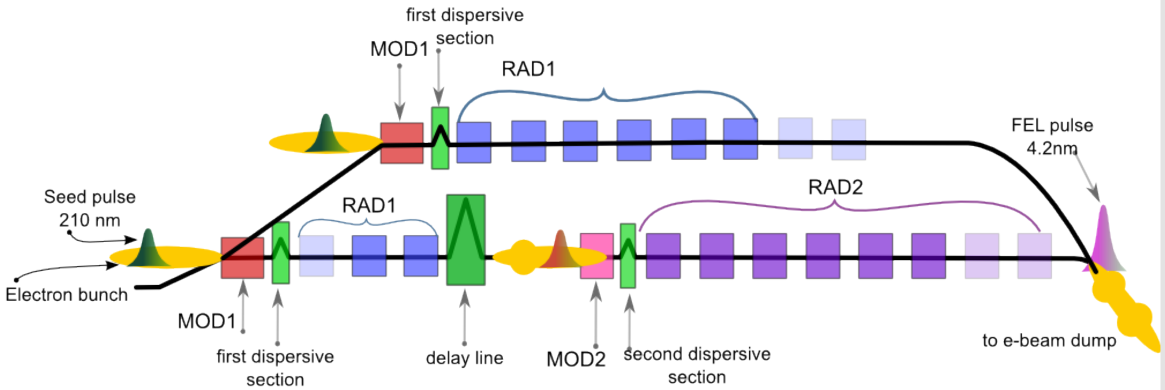
FROM NGLS STUDIES

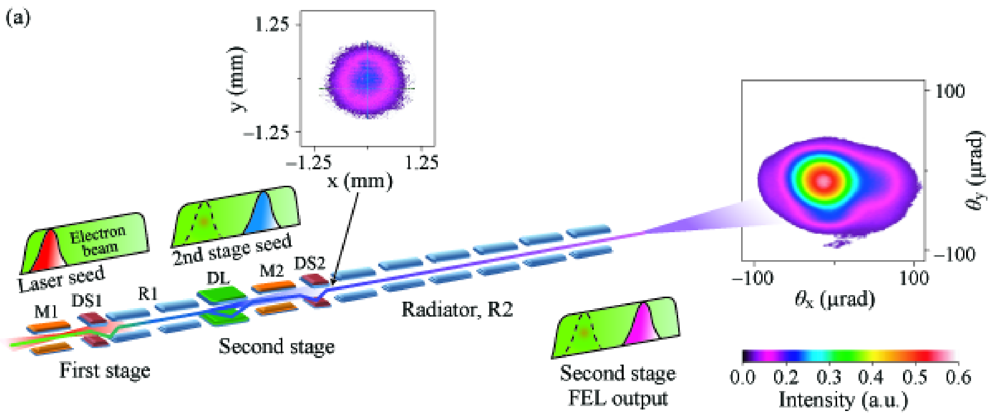
LASER FROM LASER: EXTERNAL COHERENT SEEDING

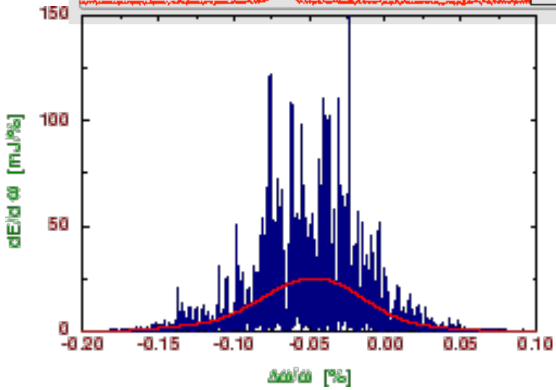


Nature Photonics 6, 699–704 (2012)









FEL LIGHT HANBURY BROWN AND TWISS (HBT) INTERFEROMETRY

nature COMMUNICATIONS

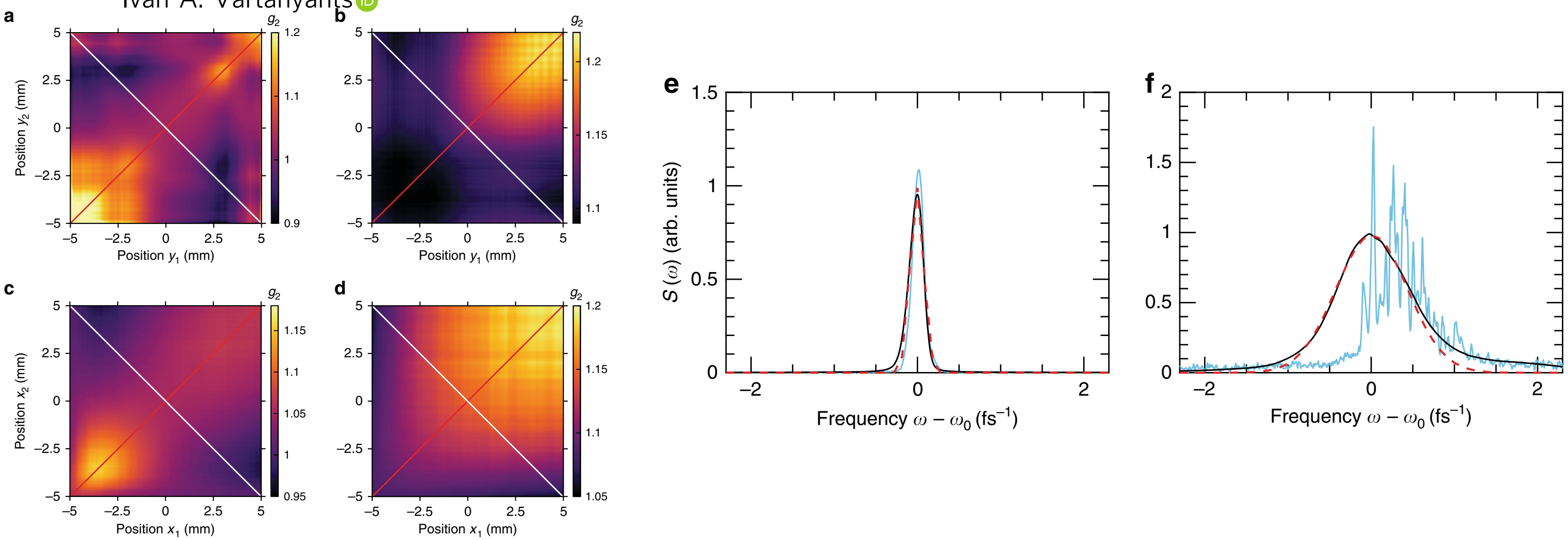
2018 FEL Hanbury Brown and Twiss (HBT) interferometry

ARTICLE

DOI: 10.1038/s41467-018-06743-8 OPEN

Seeded X-ray free-electron laser generating radiation with laser statistical properties

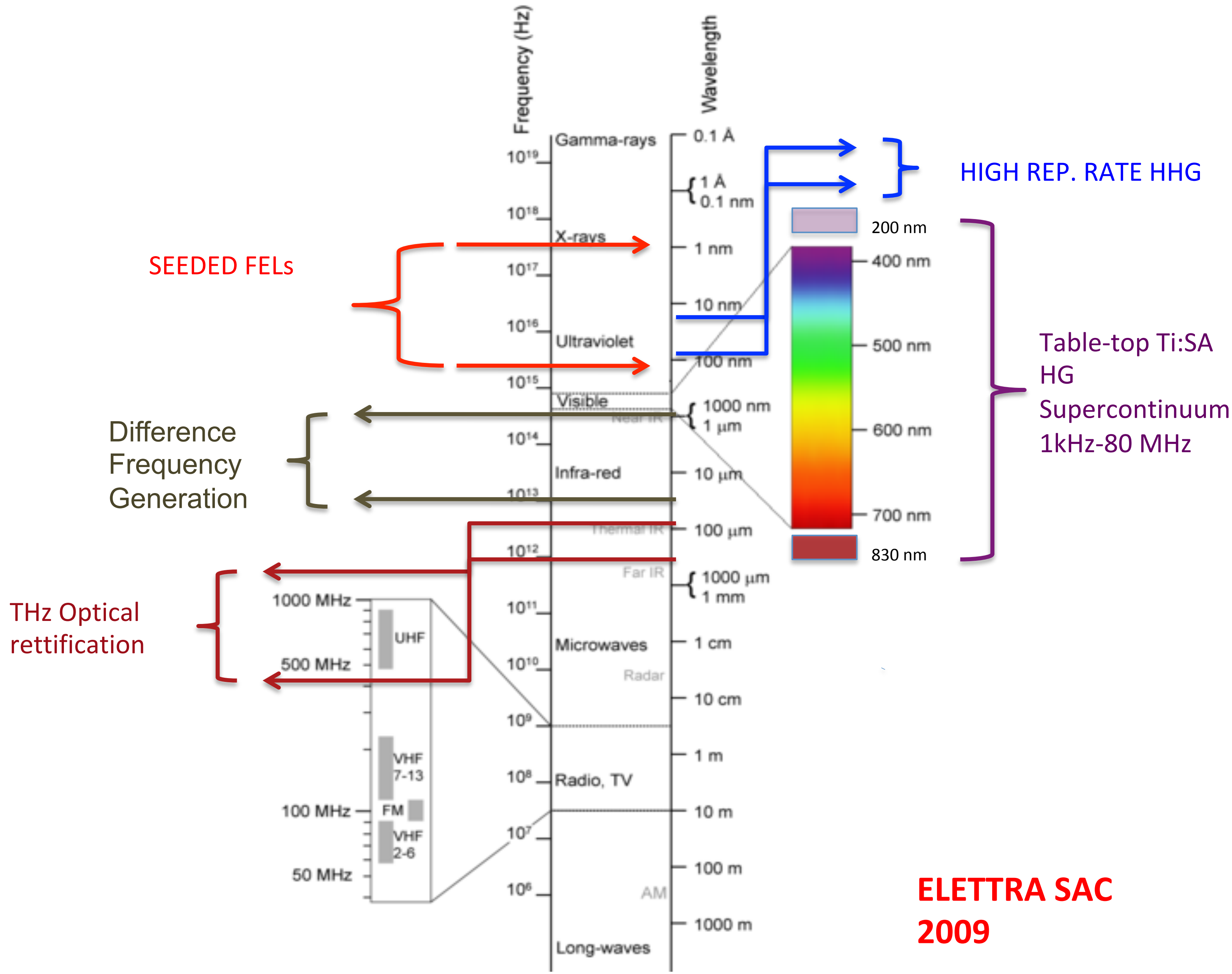
Oleg Yu. Gorobtsov[1,9], Giuseppe Mercurio[2,10], Flavio Capotondi[3], Petr Skopintsev[1,11], Sergey Lazarev[1,4], Ivan A. Zaluzhnyy[1,5,12], Miltcho B. Danailov[3], Martina Dell'Angela[6], Michele Manfredda[3], Emanuele Pedersoli[3], Luca Giannessi[3,7], Maya Kiskinova[3], Kevin C. Prince[3,8], Wilfried Wurth[1,2] & Ivan A. Vartanyants[1,5]



PRINCIPAL FEL FACILITIES

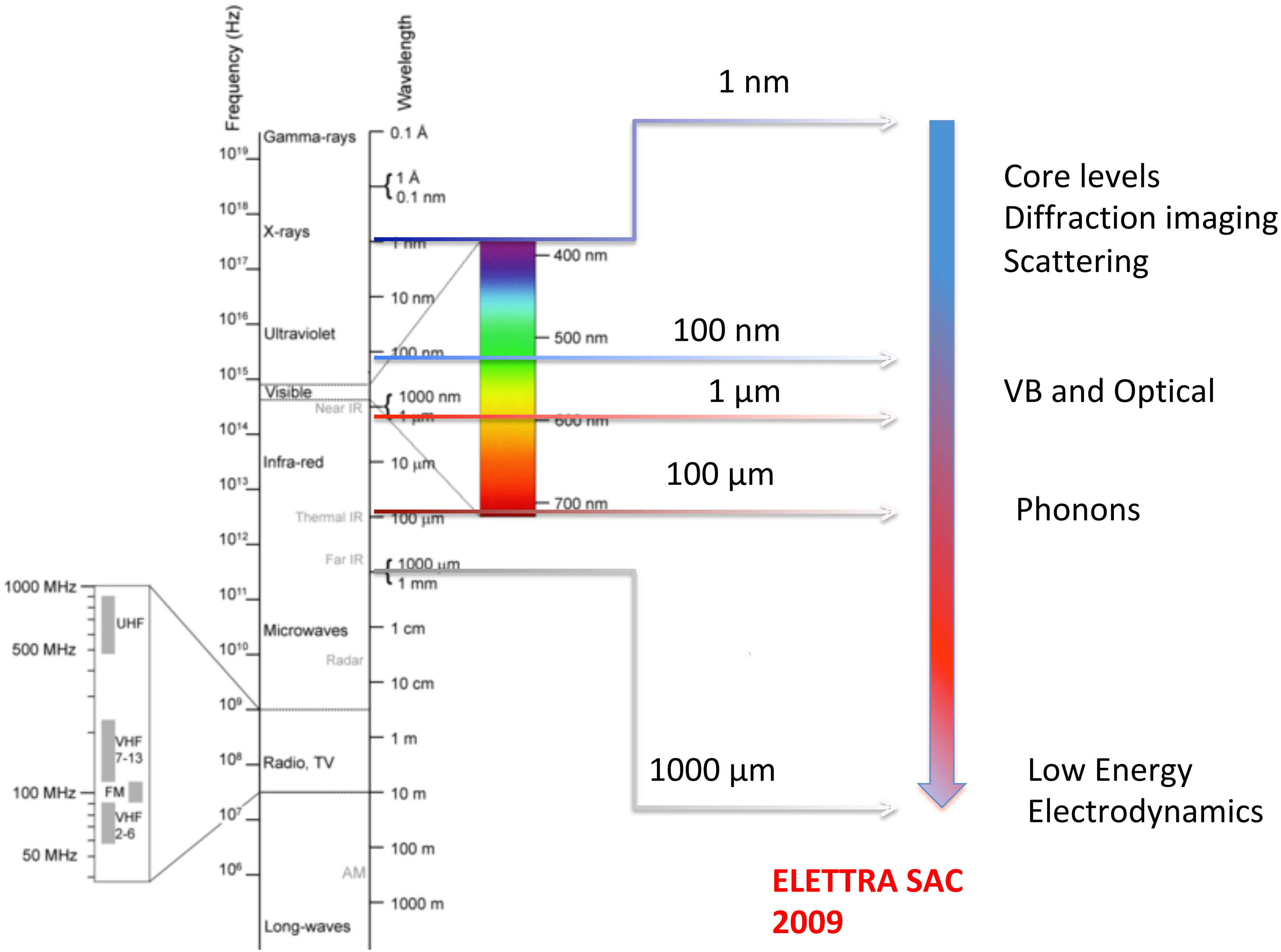
Source	Where	1st Light	Type	Notes
FLASH	Germany	2005	SC	UV FEL - 10 Hz / bunch trains
FLASH2		2013		Seeded UV FEL
LCLS	USA	2009	NC	HXR FEL - 120 Hz
LCLS-II		2018		
FERMI	Italy	2011	NC	UV FEL - 50 Hz
FEL2		2013		Seeded UV FEL
SACLA	Japan	2011	NC	HXR FEL - 60 Hz
PALFEL	Korea	2015	NC	HXR FEL - 120 Hz
EXFEL	Germany	2016	SC	HXR FEL - 10 Hz / bunch trains
SwissFEL	Switzerland	2017	NC	HXR FEL - 100 Hz
LCLS II	USA	2023	CW SC	SXR FEL - 1,000,000 Hz (CW)

TABLE-TOP AND SOFT X-RAY FELS



ELETTRA SAC 2009

TABLE-TOP AND SOFT X-RAY FELS



ELETTRA SAC 2009

A GLANCE TO THE SCIENCE

TABLE-TOP AND SOFT X-RAY FELs SCIENCE

1. Nonlinear & Quantum X-ray Optics

Multi-dimensional X-ray spectroscopy (2-color - fully tunable, synchronized) ! Stimulated X-ray Raman (local valence excitation)

Stimulated X-ray emission spectroscopy

2. High-resolution Spectroscopy at the Transform-limit

Time-resolved meV RIXS

Time & Spin Resolved ARPES

3. Heterogeneous Systems – Fluctuation Dynamics

Dynamics of mesoscale assembly (>1010 scattering snapshots)

Biological macromolecular function & interaction

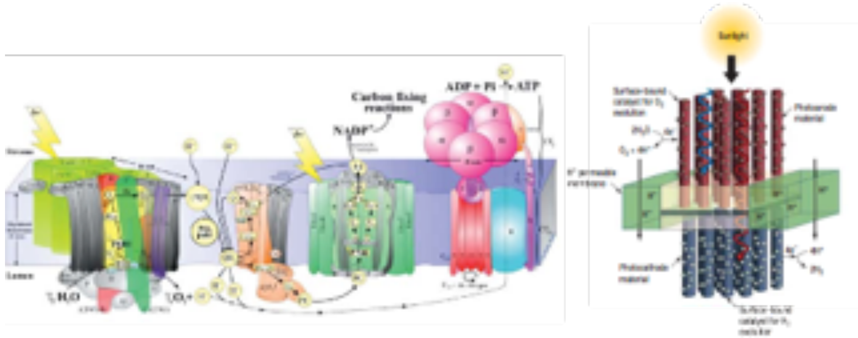
X-ray Pulses on demand for synchronization to droplets.

4. Ultrafast Molecular Reaction Microscope.

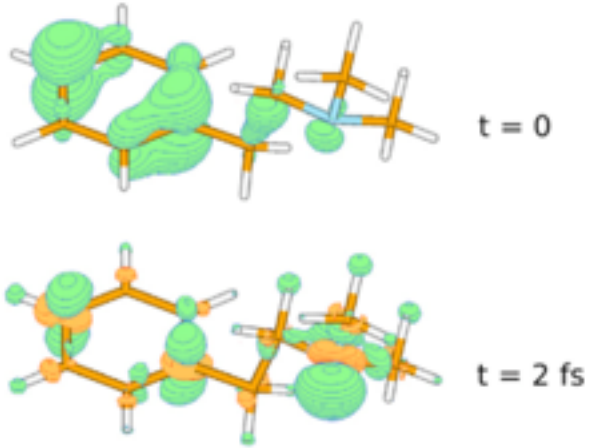
5. Cinematic Tomography with chemical selectivity

TABLE-TOP AND SOFT X-RAY FELs SCIENCE

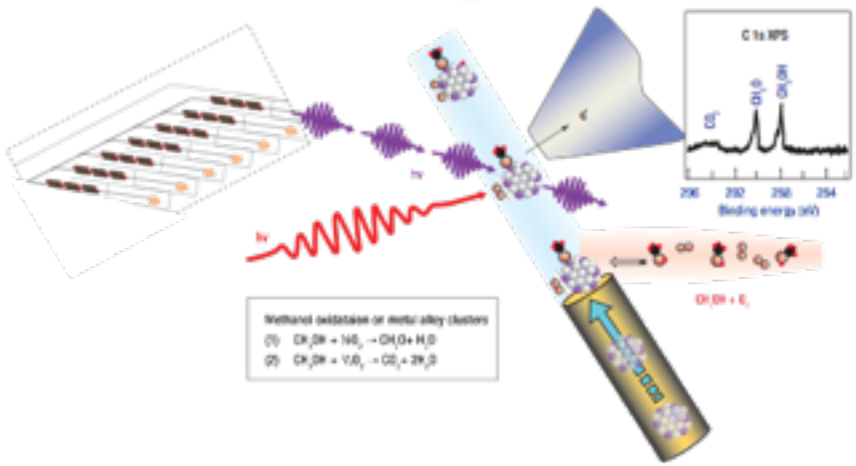
Natural and Artificial Photosynthesis



Fundamental Charge Dynamics



Catalysis



Advanced Combustion Science

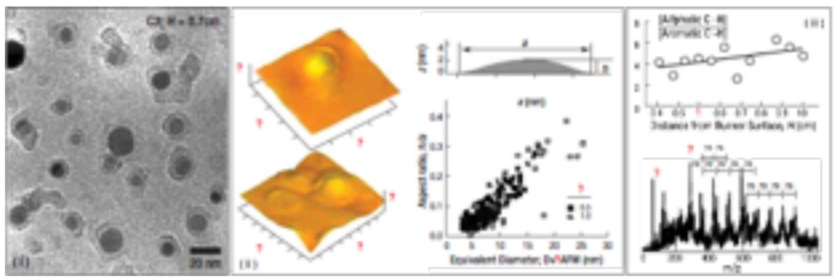
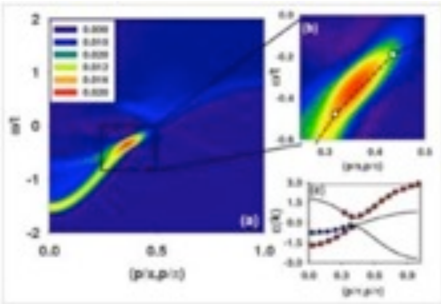
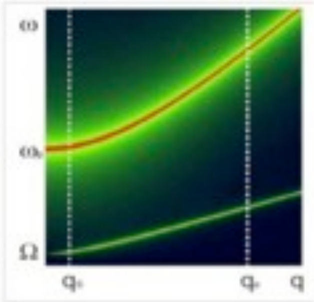


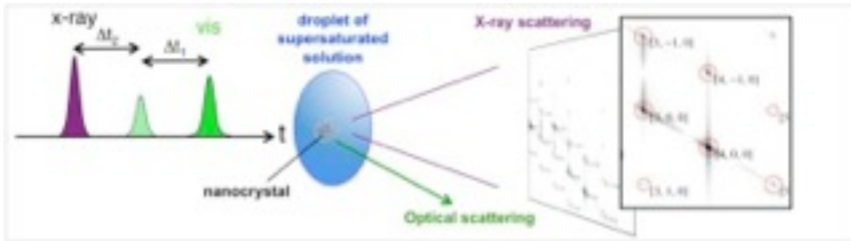
TABLE-TOP AND SOFT X-RAY FELs SCIENCE

Quantum Materials

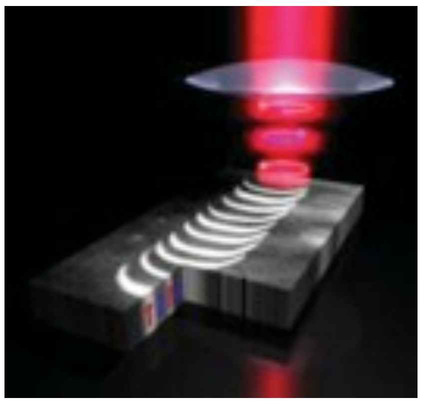
Nanoscale Materials Nucleation

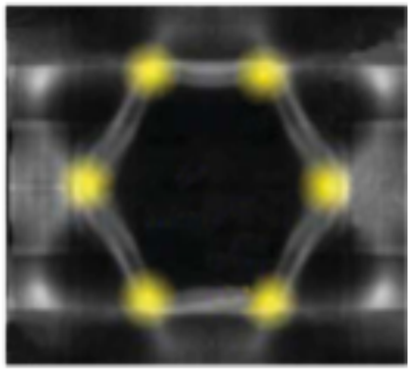




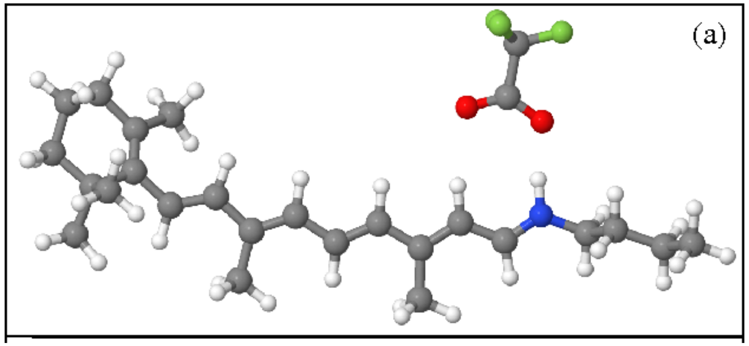


Nanoscale Spin and Magnetization

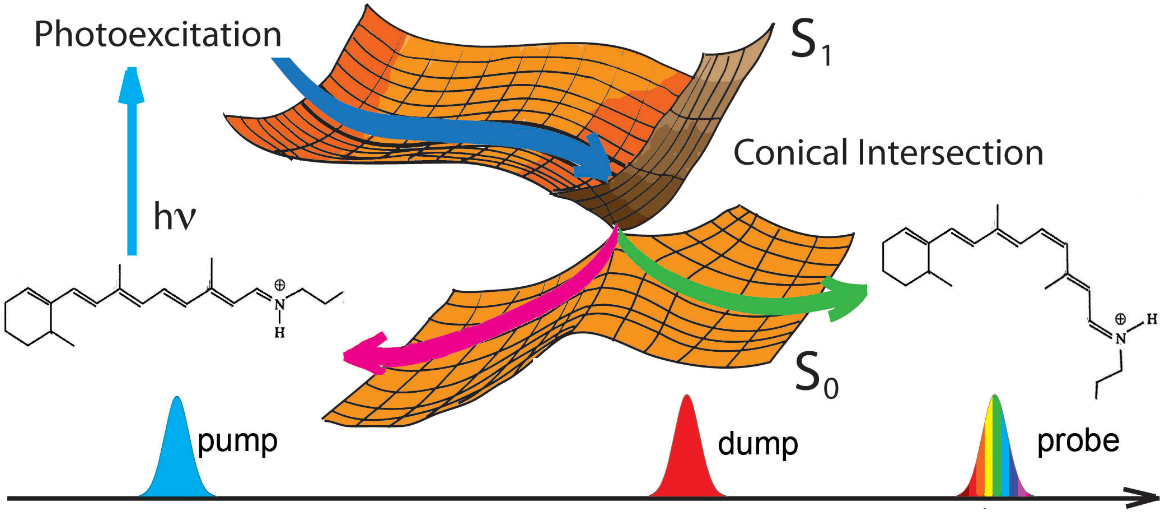




SOLAR ENERGY CONVERSION STARTS WITH CHROMOPHORES



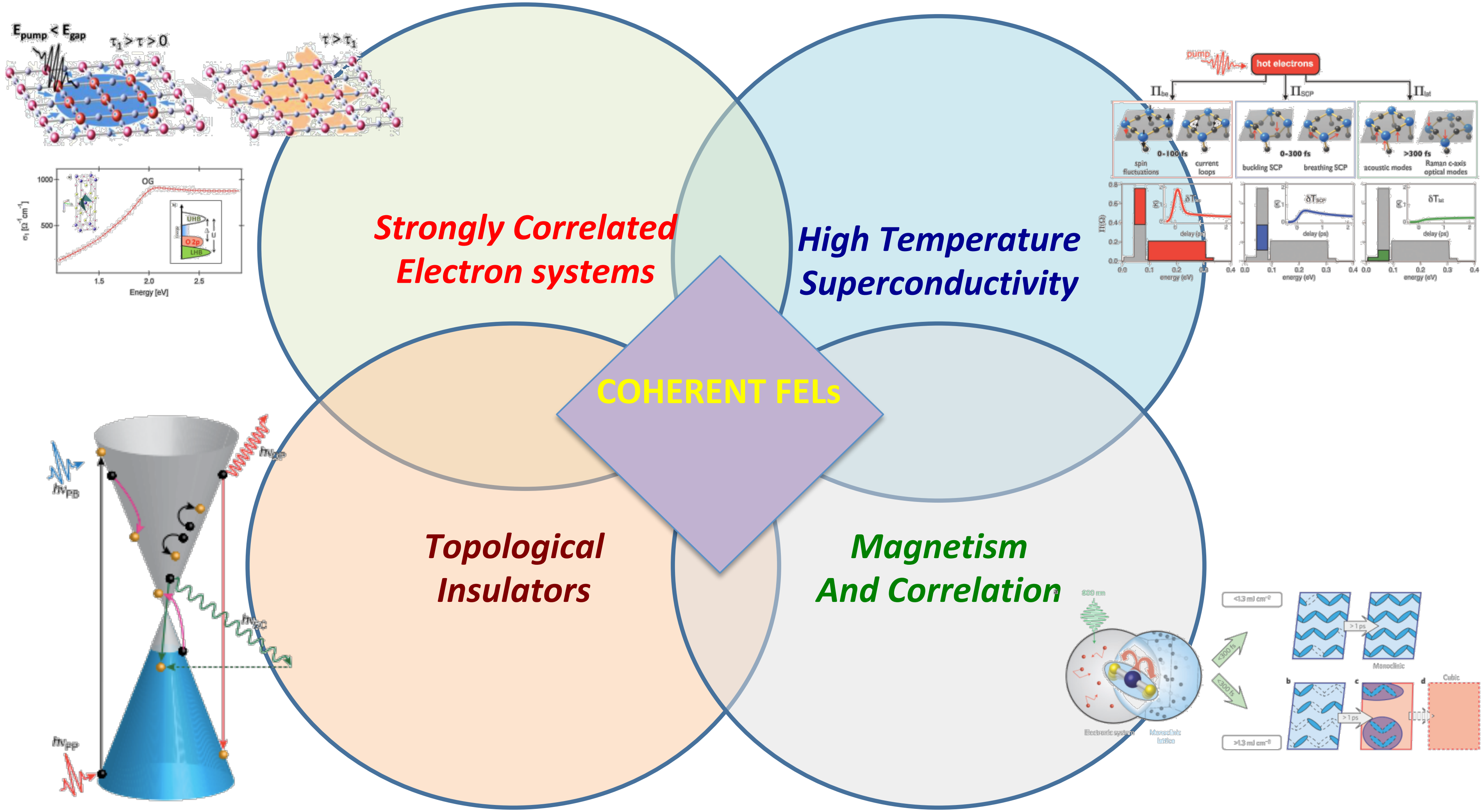
Workhorse:
The retinal chromophore
in solution



Zgrablić, Novello, Parmigiani, JACS, **134** (2) , 955-961 (2012)

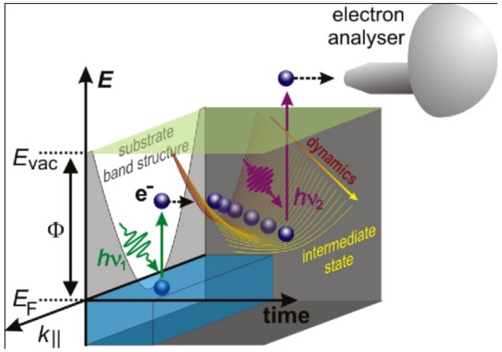
→ solvent polarity changes branching in the conical intersection

MATERIALS

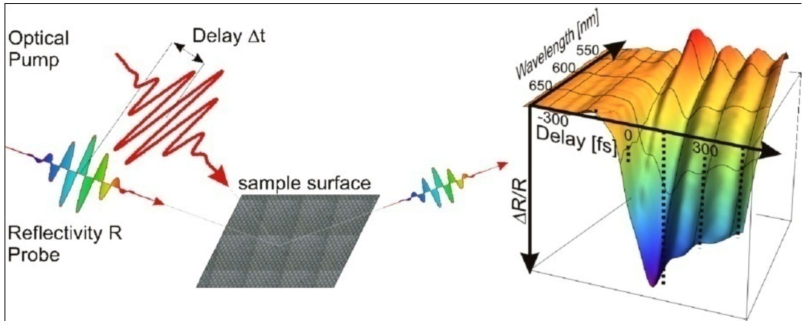

Strongly Correlated
Electron systems
High Temperature
Superconductivity
COHERENT FELs
Topological
Insulators
Magnetism
And Correlation
hot electrons
Energy [eV]

OPTICAL – PHOTOELECTRONIC - MAGNETO-OPTICAL

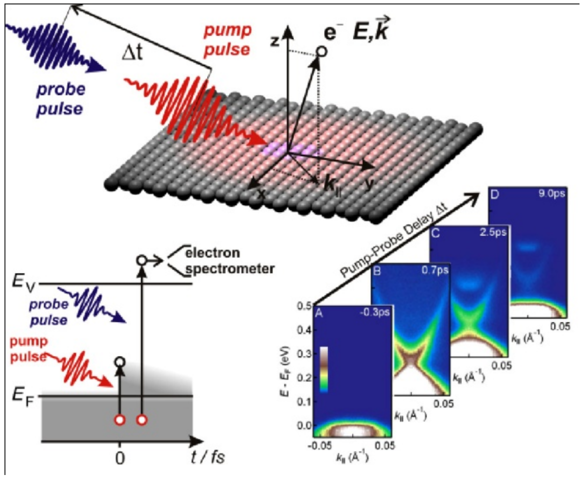
TR-2PPE



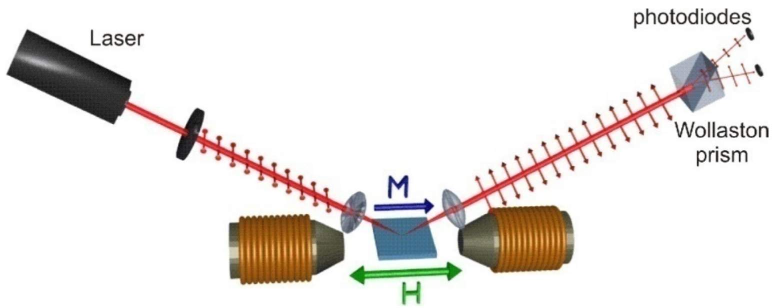
TR-OS



TR-ARPES

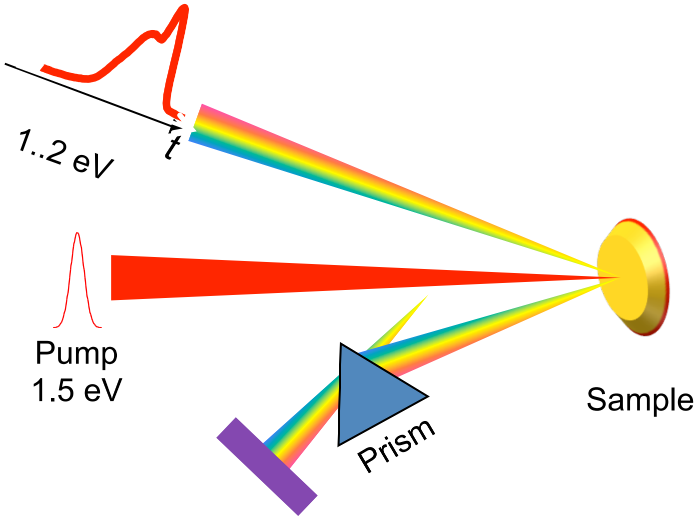


TR-MAGNETO-OPTICS



TR-OPTICAL BROADBAND SPECTROSCOPY

THE TIME RESOLVED OPTICAL SPECTROSCOPY (TROS)

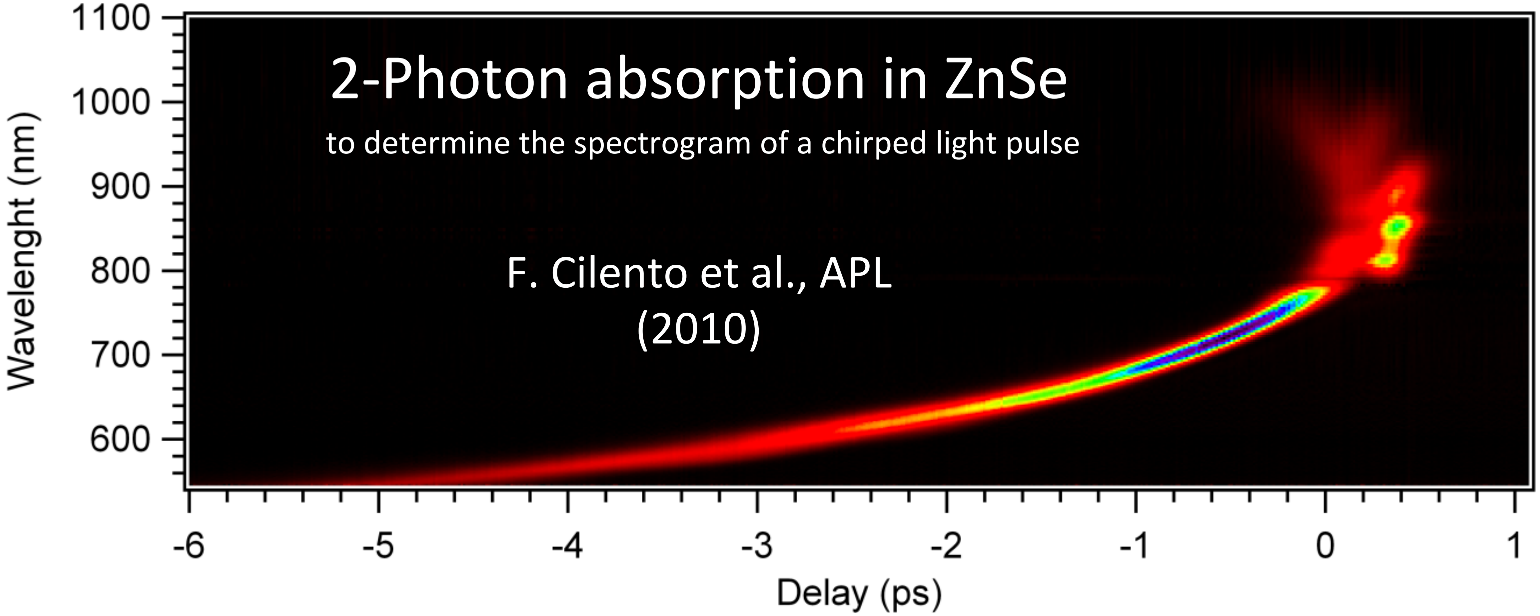


* True spectroscopy with temporal resolution
* Disentangle effects by their spectral fingerprint AND timescale
* Study the effect of a small non-equilibrium distribution

Resolved Optical Spectroscopy at T-ReX:

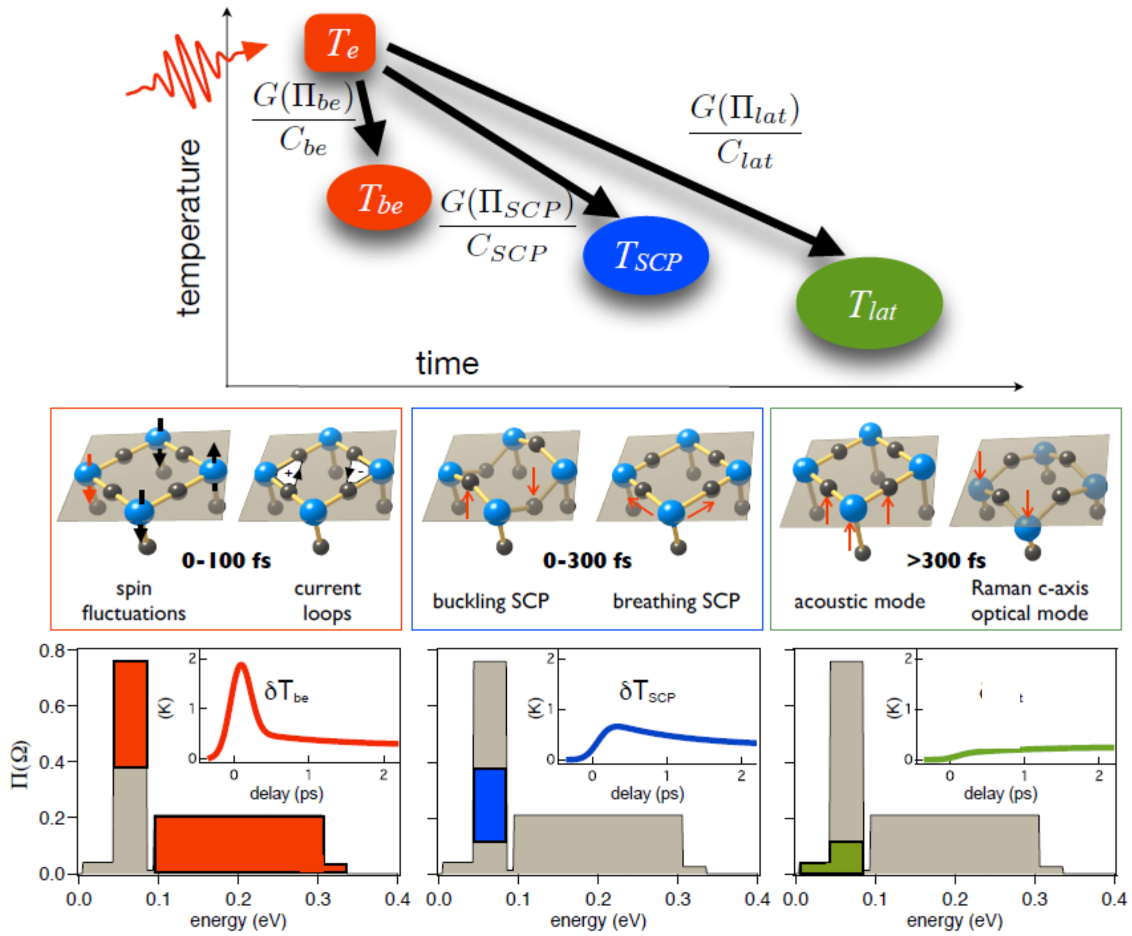
any combination is currently possible:

Probe: VIS and IR Supercontinuum (400-1100 nm and 500-1700 nm), OPA (1100-2600 nm)
Pump: 800 nm, 400 nm, OPA (1100-2600) nm)



THE NORMAL METAL STATE - SUPERCONDUCTING STATE

Unveiling the pairing mechanism
in Cuprate High-Tc superconductors



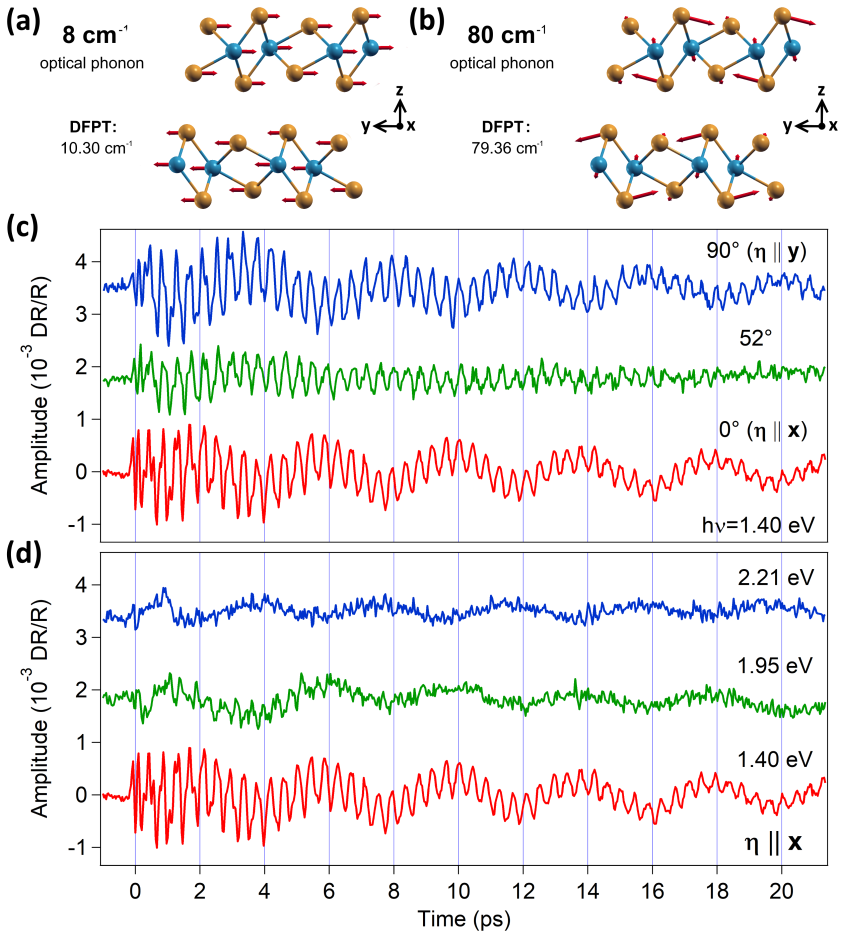
S. Dal Conte, C. Giannetti, F. Cilento et al., Science 335, 1600 (2012)

C. Giannetti, F. Cilento et al., Nature Communications 2, 353 (2011)

Sub-pm ATOMIC DISPLACEMENTS

Ultrafast broadband optical spectroscopy for quantifying subpicometric coherent atomic displacements in WTe_2

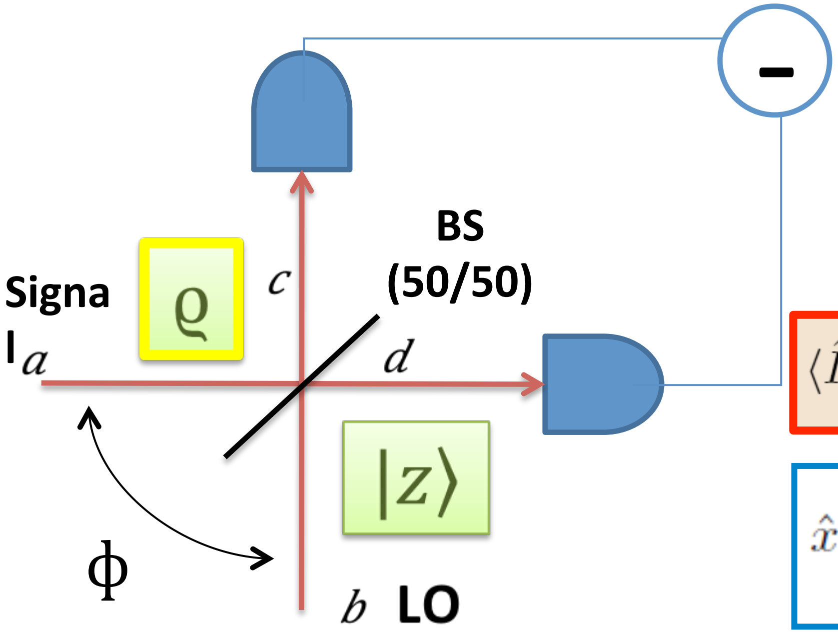
Davide Soranzio,[1] Maria Peressi,[1] Robert J. Cava,[2] Fulvio Parmigiani,[1,3,4] and Federico Cilento[4,*]



In press
Phy. Rev. Research.2019

QUANTUM OPTICS TR-SPECTROSCOPY

BALANCED HOMODYNE DETECTION



$$\hat{I} = \hat{c}^\dagger \hat{c} - \hat{d}^\dagger \hat{d} = \hat{a}^\dagger \hat{b} + \hat{b}^\dagger \hat{a}$$

$$|z|^2 \gg \langle \hat{a}^\dagger \hat{a} \rangle$$

$$\langle \hat{I} \rangle = |z| \, \langle (\hat{a}^\dagger e^{i\Phi} + \hat{a} e^{-i\Phi}) \rangle = (\sqrt{2}\,|z|) \, \langle \hat{x}_\Phi \rangle$$

$$\hat{x}_\Phi = \frac{1}{\sqrt{2}} (\hat{a} e^{-i\Phi} + \hat{a}^\dagger e^{i\Phi})$$

$$\Phi \in [0; 2\pi]$$

$$z = |z| e^{i\Phi}$$

$$\hat{b}|z\rangle = z|z\rangle$$

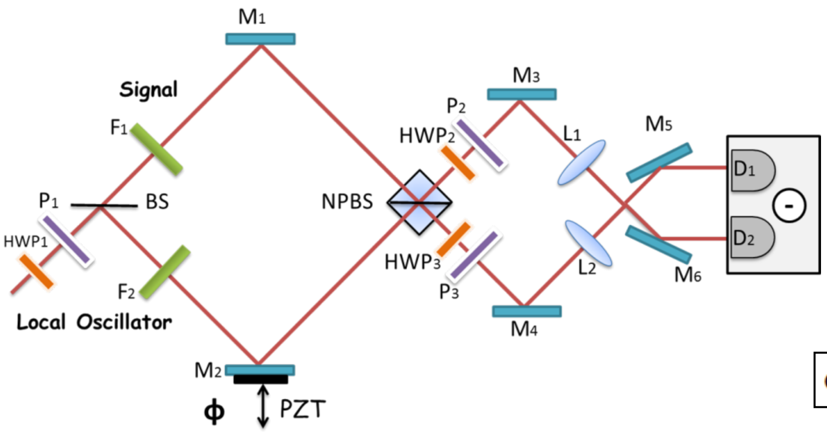
$$\mathrm{P}(x, \phi) = Tr[\varrho |x_\phi\rangle\langle x_\phi|] = \langle x_\phi | \varrho | x_\phi \rangle$$

Determination of the state ϱ!

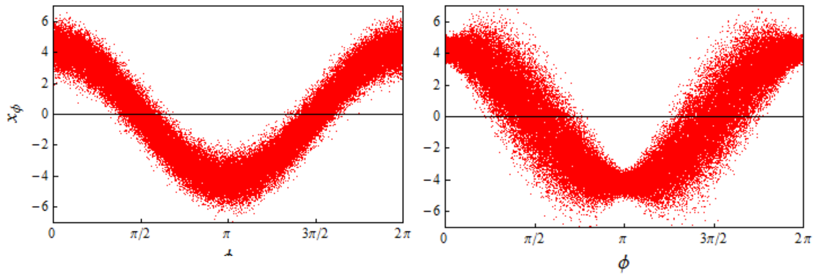
FOLLOW PAPERS BY D. FAUSTI AND Co-WORKERS

BALANCED HOMODYNE DETECTION

Quantum state reconstruction



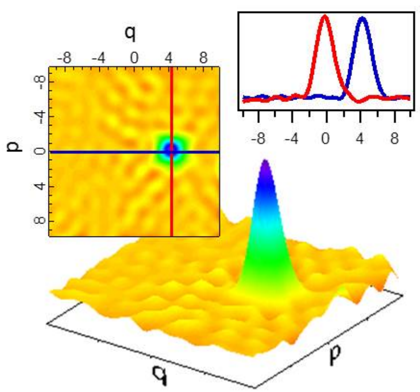
Coherent state N~9 Squeezed stateN~9



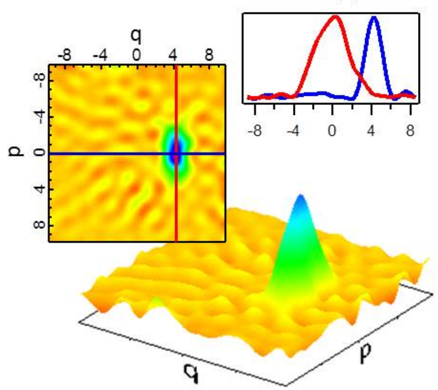
C. Butucea, M. Guta and L. Artiles, *Ann. Stat.* **35**, 465 (2007)

WIGNER FUNCTION RECONSTRUCTION

Coherent state $D(\alpha)|0\rangle$

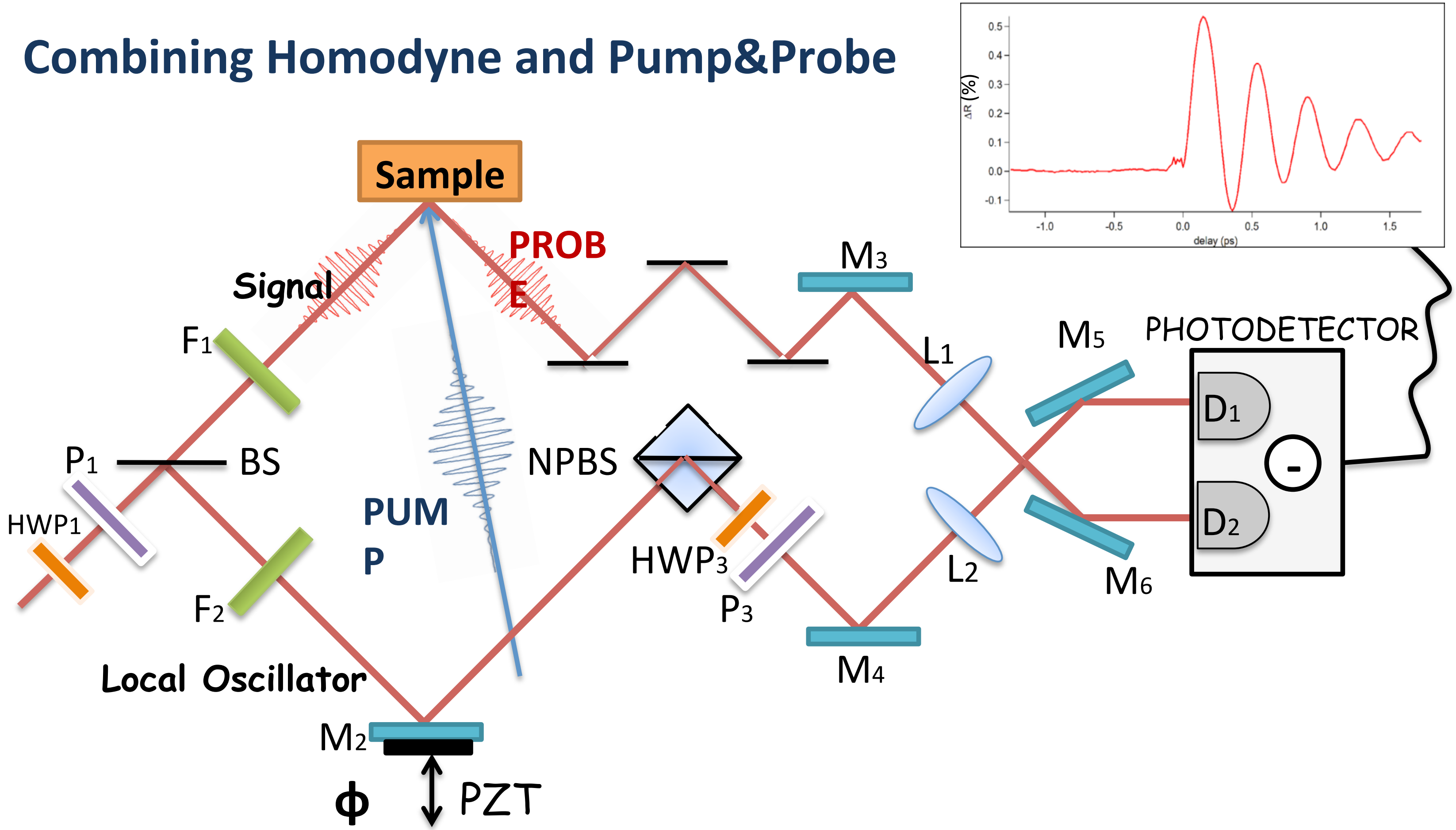


Coherent-squeezed state $D(\alpha)S(\xi)\,|0\rangle$



BALANCED HOMODYNE DETECTION

Combining Homodyne and Pump&Probe



POLARIZATION

ANGULAR MOMENTUM IN TERMS OF PHOTONS

- Spin angular momentum
  - Circular polarisation
  - $\sigma\hbar$ per photon
- Orbital angular momentum
  - Helical phasefronts
  - $\ell\hbar$ per photon

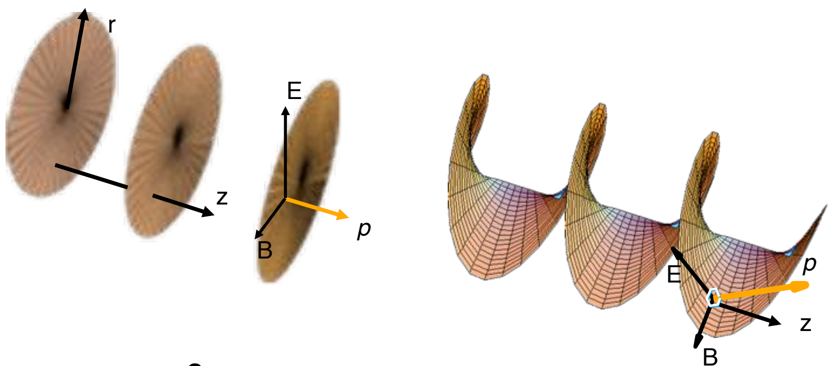
$\sigma = +1$

$\sigma = -1$

Intrinsic angular momentum of the photon formally equivalent to the particle spin

$\ell = 0$ $\ell = 1$ $\ell = 2$ $\ell = 3$ etc

Adapted from Miles Padgett



$p_\theta = 0$ $p_\theta \neq 0$

TUNABILITY

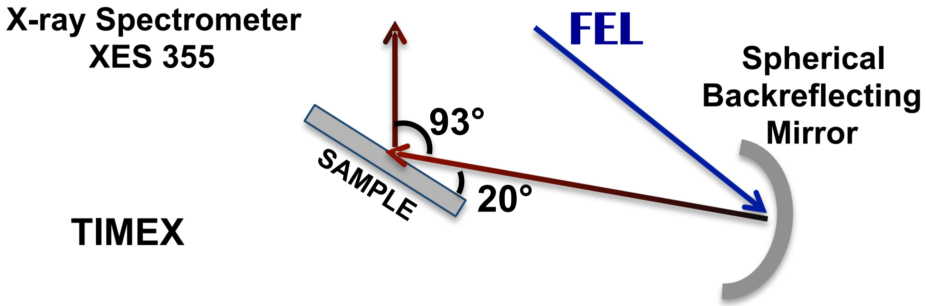
M-edge RIXS @ FERMI

Martina Dell'Angela

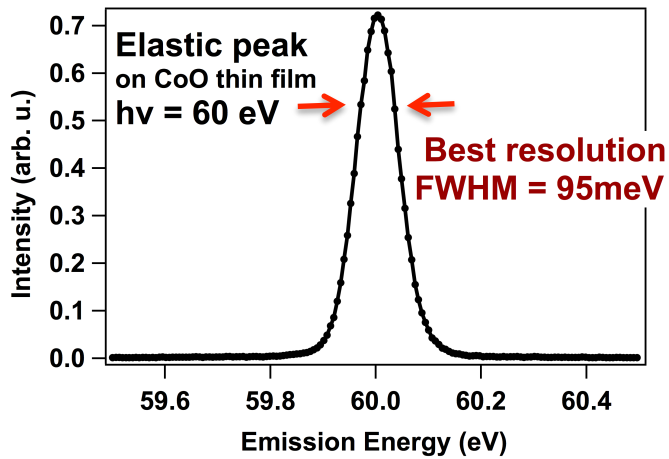
in collaboration with

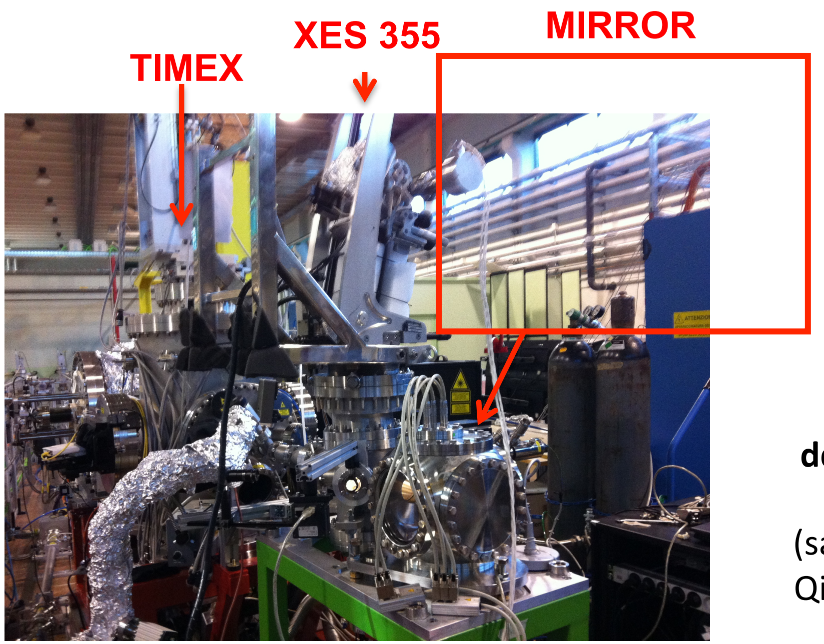
W. Wurth (UNI HH/CFEL) and S. Bajit (CFEL/DESY)

TIMEX, MagneDyn and T-ReX teams



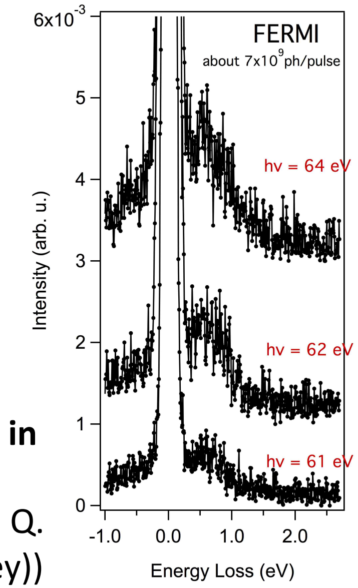
Resolution



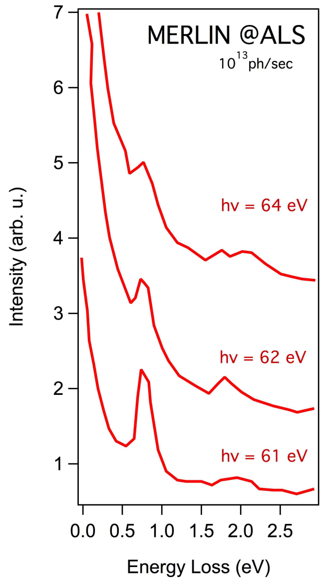


dd- excitations in CoO

(sample from Z. Q. Qiu (UC Berkeley))



FERMI Nov 2014



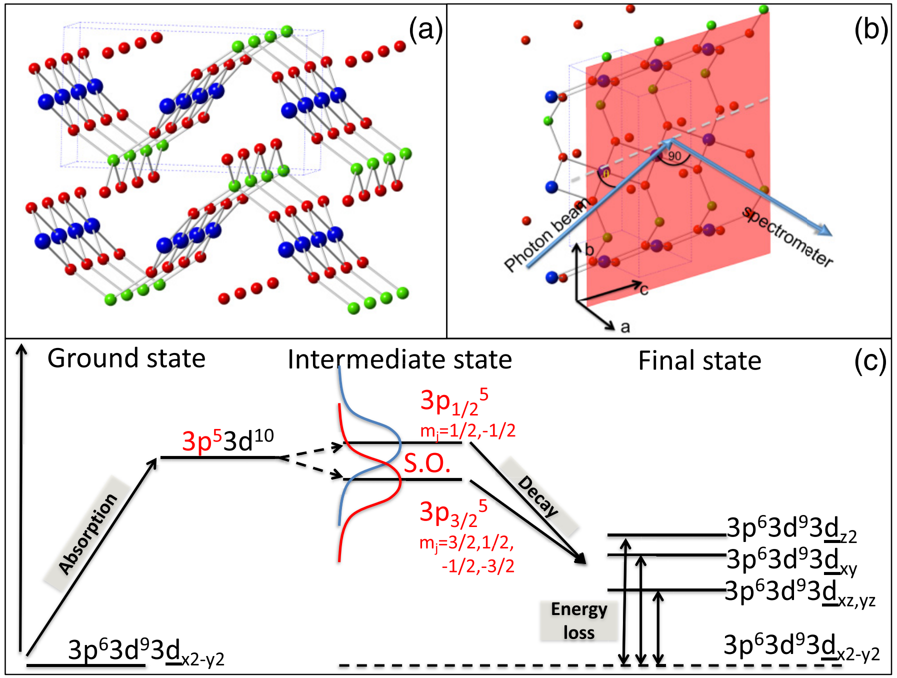
Wray et al.
PRB 88, 035105 (2013)

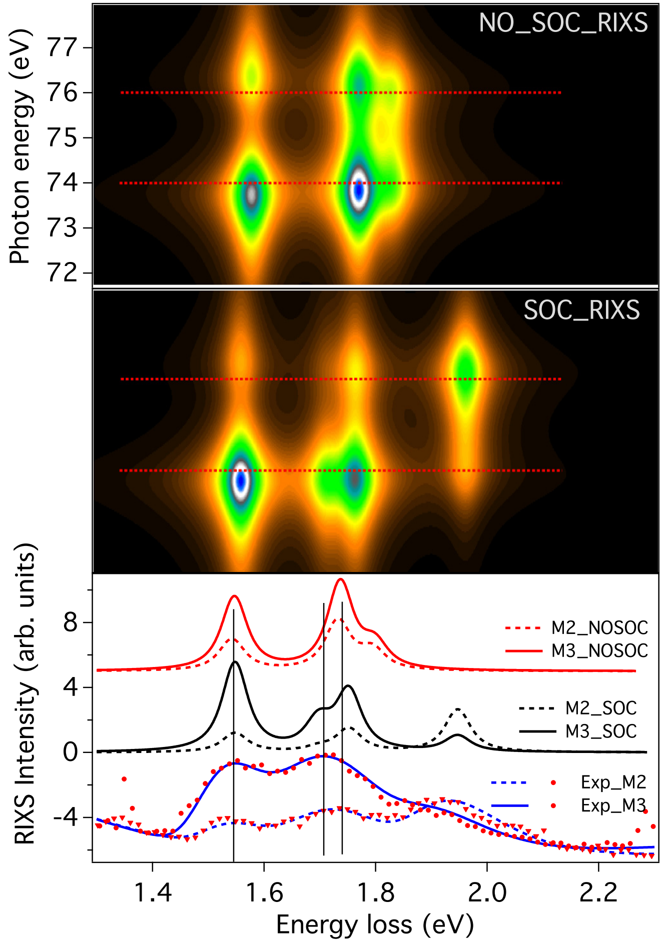
A COMMON PROJECT: TR M-edge RIXS @ FERMI

PHYSICAL REVIEW B **99**, 115120 (2019)

Direct observation of spin-orbit-induced 3*d* hybridization via resonant inelastic extreme ultraviolet scattering on an edge-sharing cuprate

Marco Malvestuto,[1,2,*] Antonio Caretta,[1] Barbara Casarin,[2] Roberta Ciprian,[1,†] Martina Dell'Angela,[3] Simone Laterza,[2] Yi-De Chuang,[4] Wilfried Wurth,[5,6] Alexandre Revcolevschi,[7] L. Andrew Wray,[8] and Fulvio Parmigiani[1,2,9]





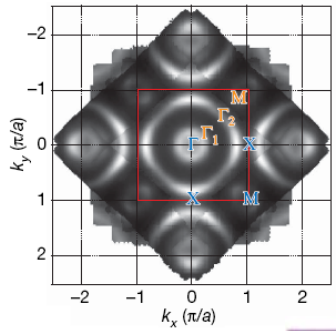
A GLANCE TO THE FUTURE

ARPES AND QM

Structures in momentum space

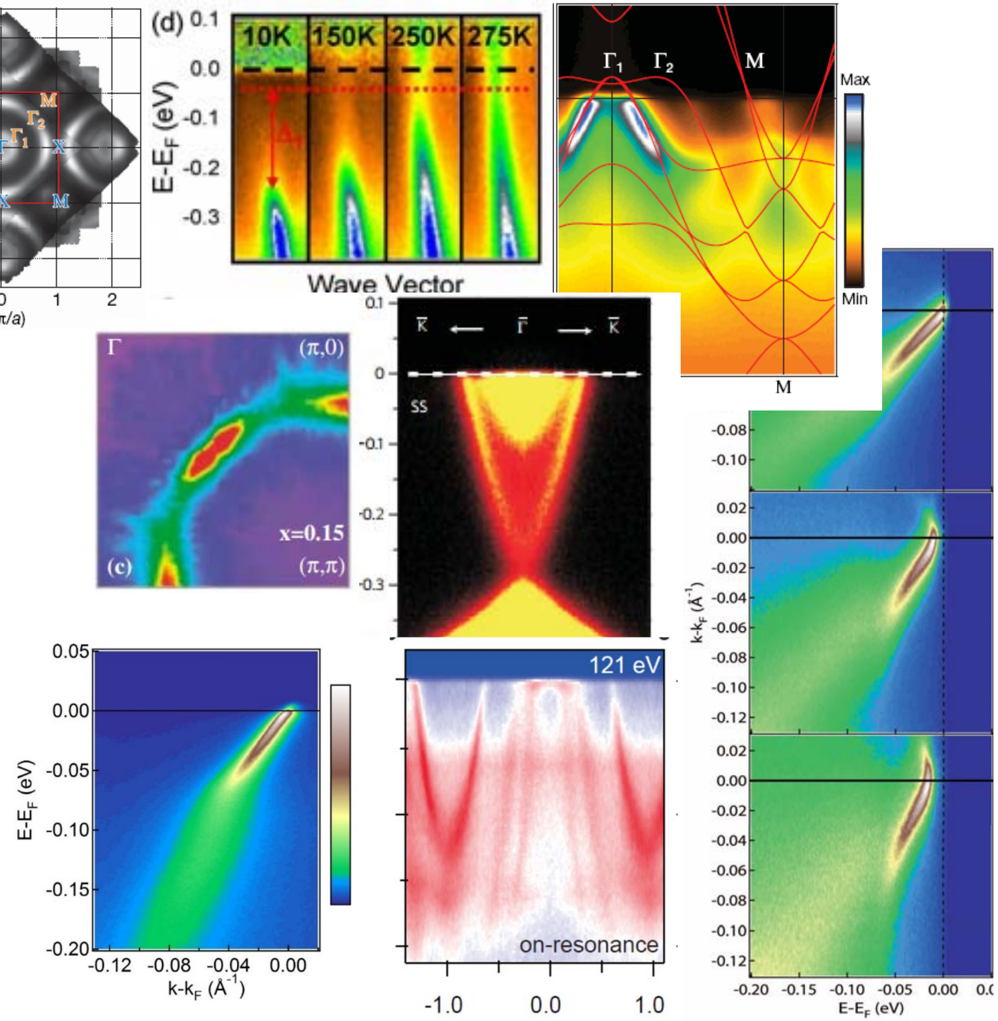
All materials

- Brillouin zones
- Fermi surfaces
- Band dispersion

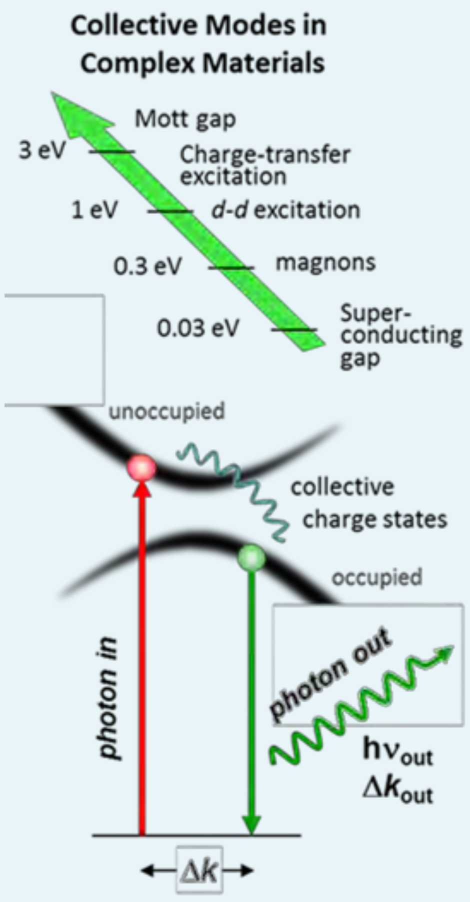


Related to quantum materials

- Charge density wave gaps
- Superconducting gaps
- Spin density wave gaps
- Electron-boson coupling
- Heavy fermion hybridization gaps
- Spin momentum locking
- Dirac dispersions
- Surface states
- …



RIXS AND QM



Momentum-resolved
Resonant Inelastic X-ray Scattering
(***q***-RIXS)

- Bulk sensitive
- Element-specific (L-edge, or K-edge)
  - Transition-metals (Fe, Cu, Mn, Ti, Ir etc.)
  - Oxygen, Arsenic
- Energy resolution (~20 meV)
- Momentum resolution
- Time resolution (20 meV⇔100 fs)
- Monolayer/interface sensitivity
- Applied fields (magnetic, electric)

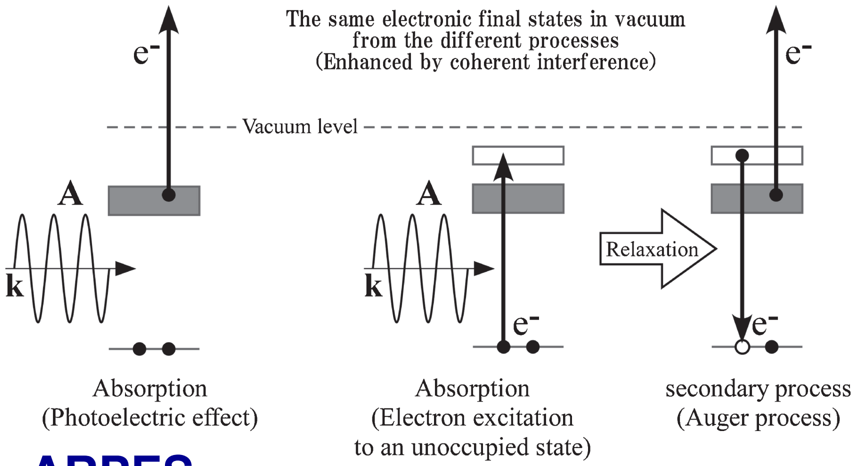
Current limitations:

- Photon flux in narrow bandwidth (coherent X-ray source ⇒ laser)

COMBINING ARPES/RES-ARPES WITH RIXS

ARPES RES-ARPES RIXS

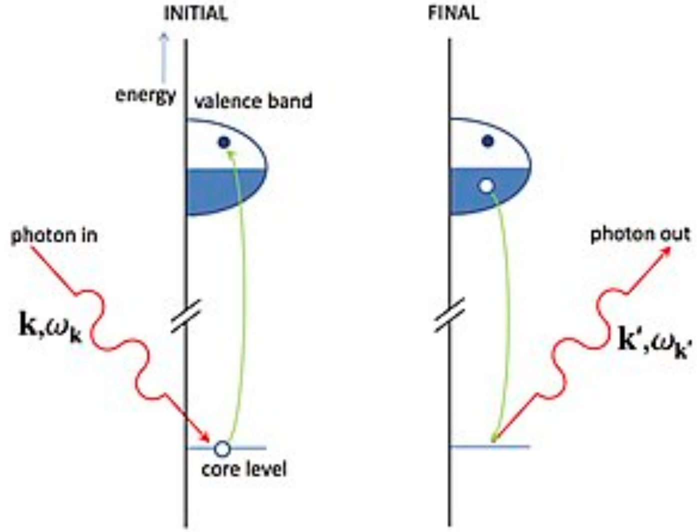
COMPLEMENTARY INFORMATION
AT ONCE
IN THE MOMENTUM SPACE



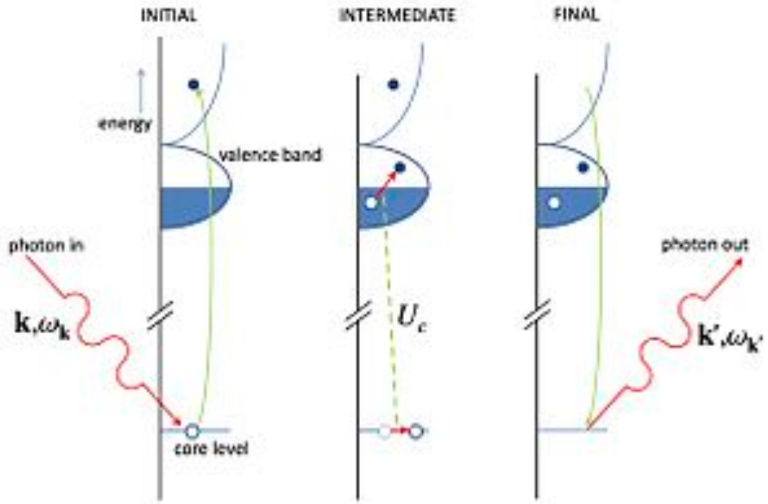
ARPES

RES-ARPES

FANO RESONANCE
SPIN TEXTURE IN AF SYSTEM



DIRECT RIXS



INDIRECT RIXS

A PARADIGMATIC EXAMPLE:
MAGNETIC INTERACTIONS IN QM

MEASURING THE SPIN TEXTURE IN AF-QM

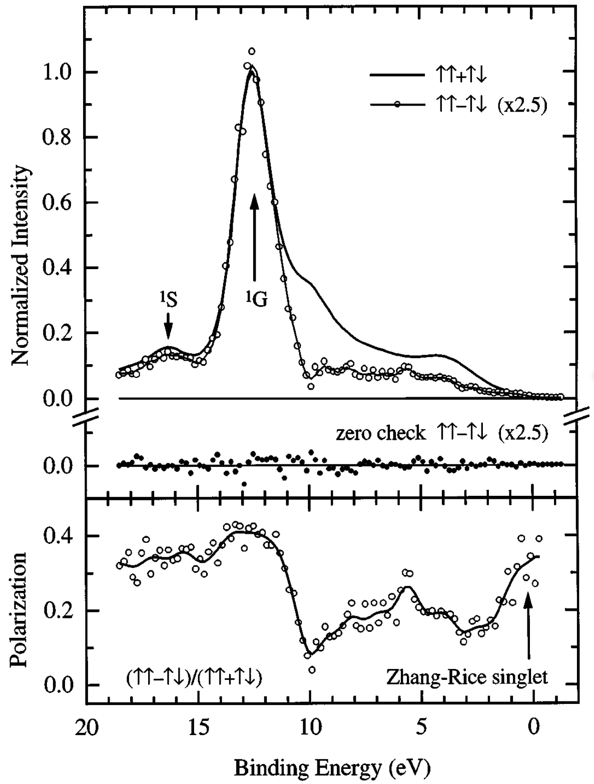
VOLUME 78, NUMBER 6 PHYSICAL REVIEW LETTERS 10 FEBRUARY 1997

Spin-Resolved Photoemission on _Anti_-Ferromagnets: Direct Observation of Zhang-Rice Singlets in CuO

L. H. Tjeng,[1] B. Sinkovic,[2] N. B. Brookes,[3] J. B. Goedkoop,[3] R. Hesper,[1] E. Pellegrin,[1] F. M. F. de Groot,[1] S. Altieri,[1] S. L. Hulbert,[4] E. Shekel,[2,*] and G. A. Sawatzky[1]

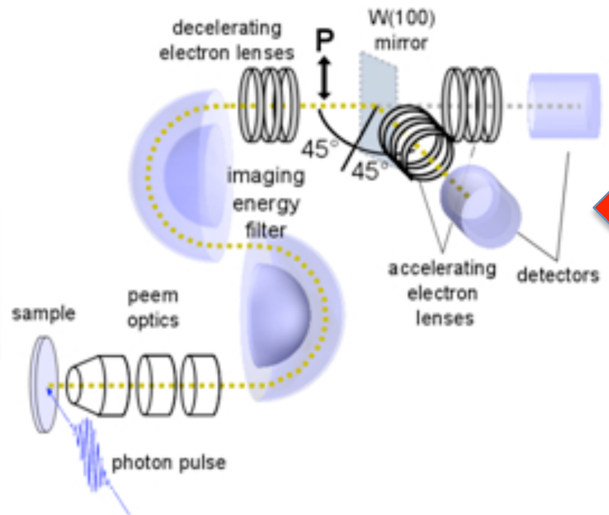
In this photoemission work on CuO we make use of the $2p_{3/2}$ (L_3) resonance condition: when the photon energy is near the Cu $2p$ ($L_{2,3}$) absorption edges, the photoemission consists not only of the direct channel ($3d^9 + h\nu \rightarrow 3d^8 + e$) but also, and, in fact, overwhelmingly, of the deexcitation channel in which a photoabsorption process is followed by a nonradiative Auger decay ($2p^6 3d^9 + h\nu \rightarrow 2p^5 3d^{10} \rightarrow 2p^6 3d^8 + e$). In principle, to observe spin signal one needs only the use of a spin detector and circularly polarized light. It is important to realize, however, that circularly polarized light can only be very effective if a strong spin-orbit splitting is present in the atomic subshell under study, because then angular momenta will govern the selection rules [24]. Conse-

MEASURING THE SPIN TEXTURE IN AF-QM



SPIN TEXTURE IN AF MAT

Intrinsic angular momentum of the photon formally equivalent to the particle spin



electron spin

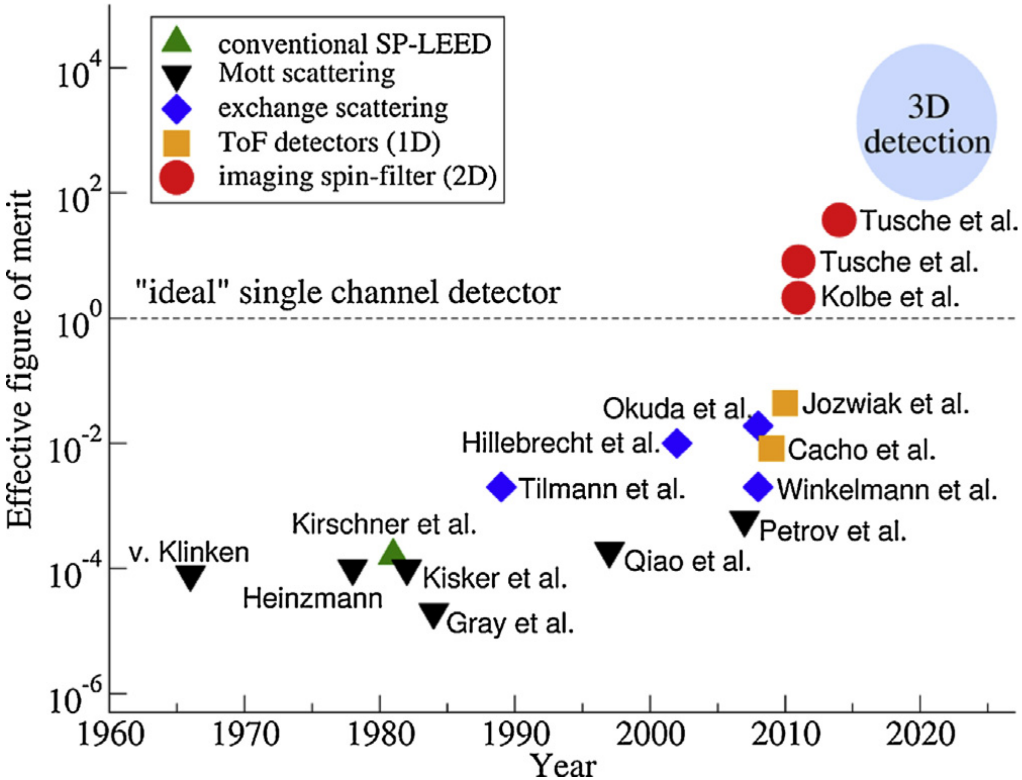
We demonstrate that it is possible to obtain spin-resolved valence band spectra with a very high degree of spin polarization from antiferromagnetic transition metal materials if the excitation light is circularly polarized and has an energy close to the cation $2p_{3/2}$ (L_3) white line. We are able to unravel the different spin states in the single-particle excitation spectrum of CuO and show that the top of the valence band is of pure singlet character, which provides strong support for the existence and stability of Zhang-Rice singlets in high-T_c superconductors. [S0031-9007(97)02343-0]

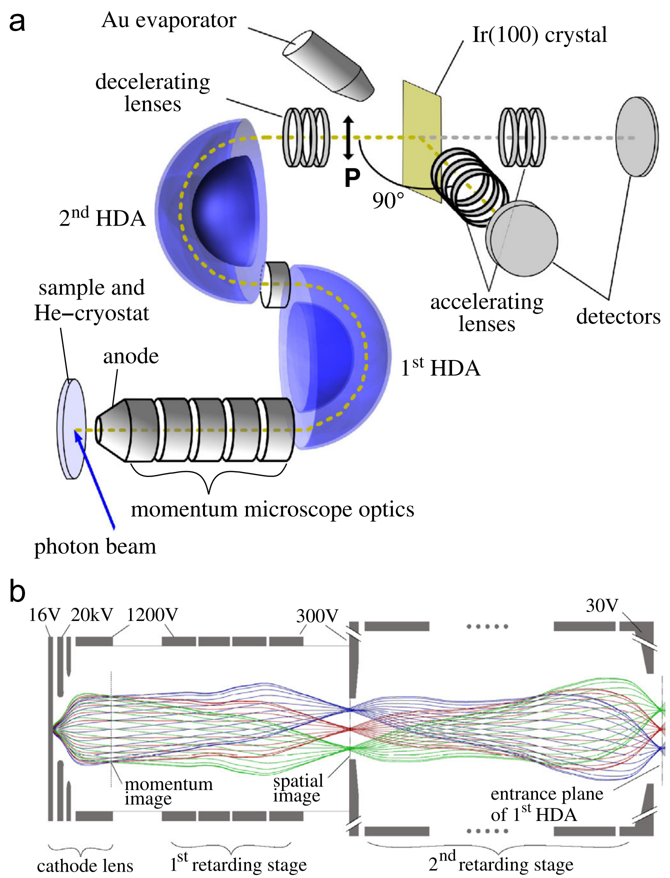
MEASURING THE SPIN TEXTURE IN AF-QM

Spin resolved bandstructure imaging with a high resolution momentum microscope

Christian Tusche [a,*], Alexander Krasyuk [a], Jürgen Kirschner [a,b]

[a] *Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle, Germany*
[b] *Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, 06099 Halle, Germany*

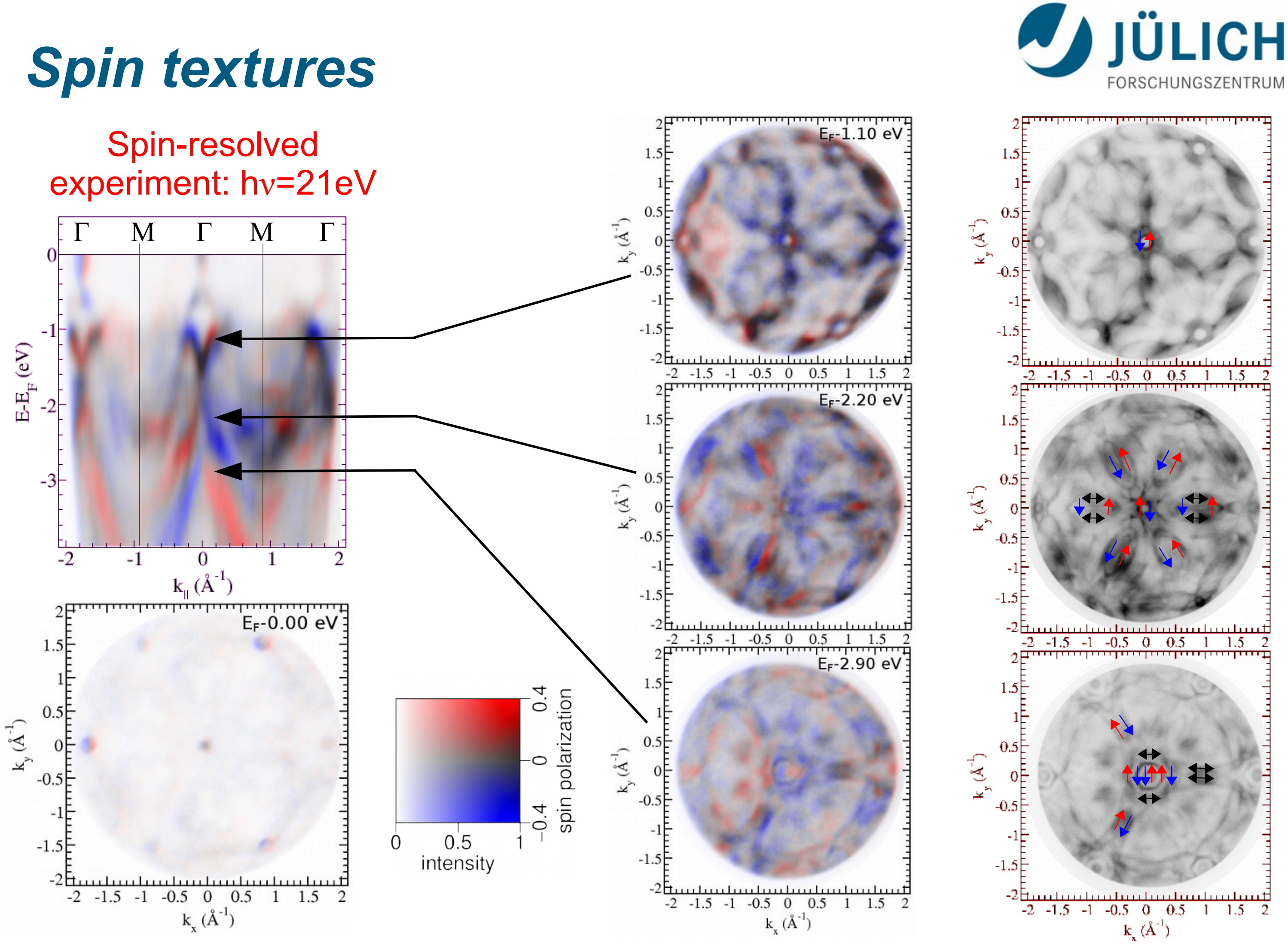




S. Suga, C. Tusche / Journal of Electron Spectroscopy and Related Phenomena 200 (2015) 119–142

MEASURING THE SPIN TEXTURE IN T.I.

Spin textures

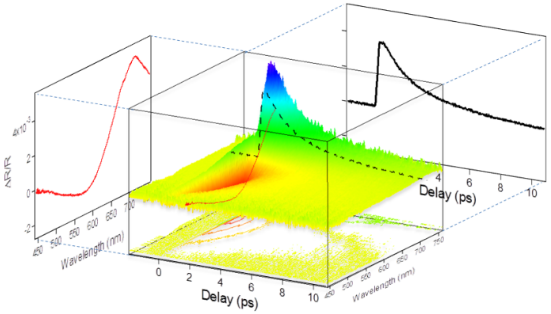


Forschungszentrum Jülich, Y.-J. Chen and C. Tusche

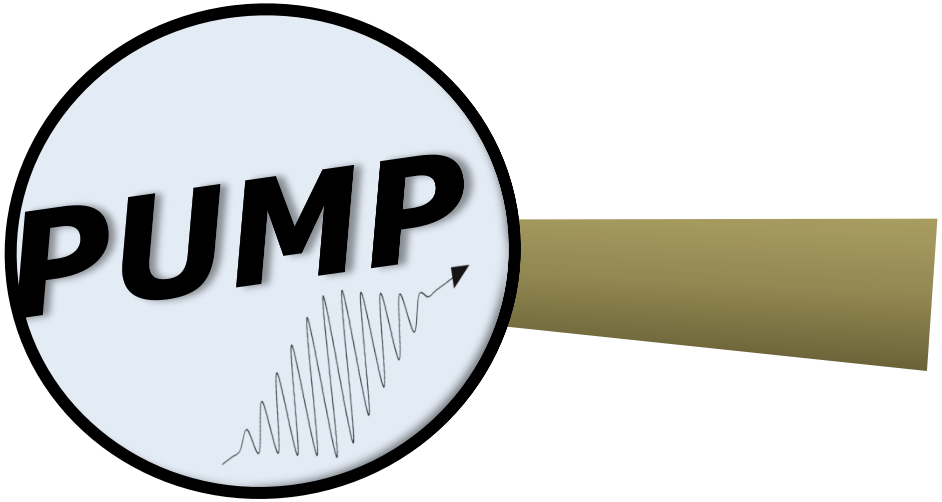
SELECTIVE EXCITATIONS

AVAILABLE SPECTROSCOPIC TOOLS

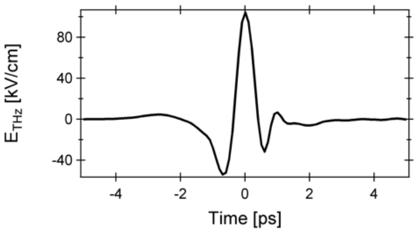
Optical Pump & Broadband Probe



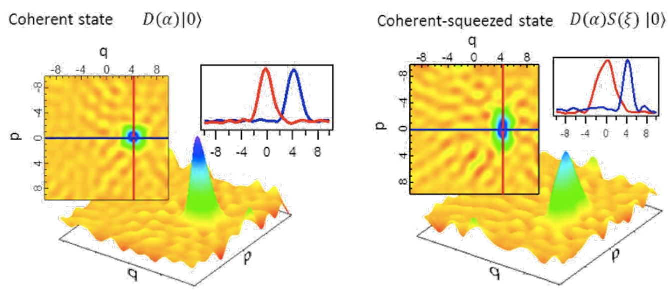
Phase Stable Near-IR Pump & WL



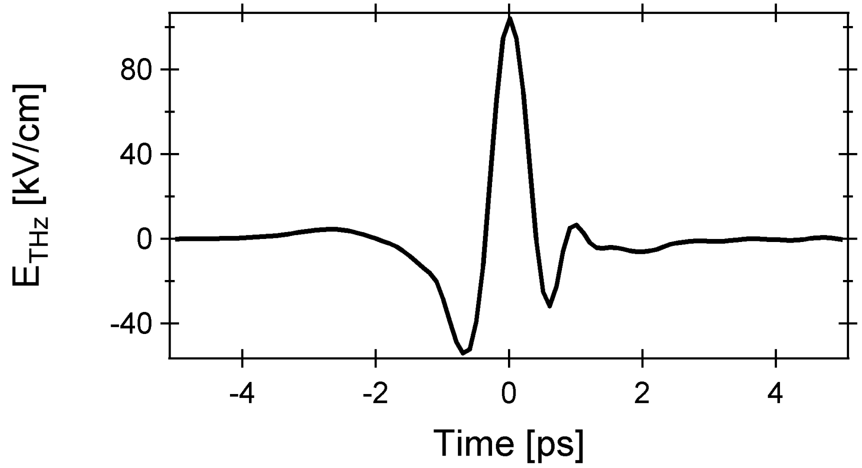
THz-Excitations

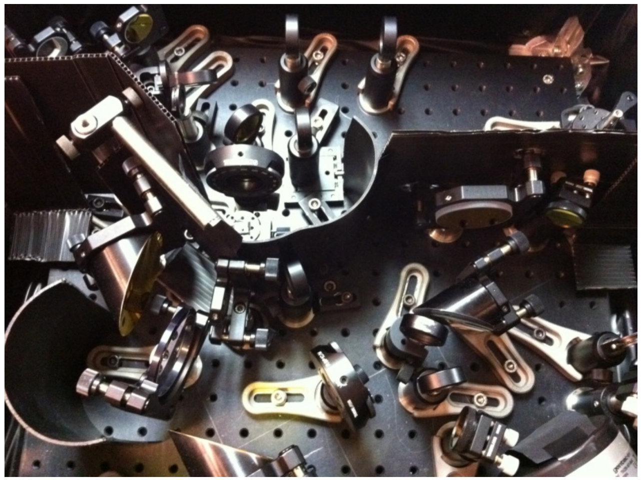


Homodyne reconstruction of the quantum state

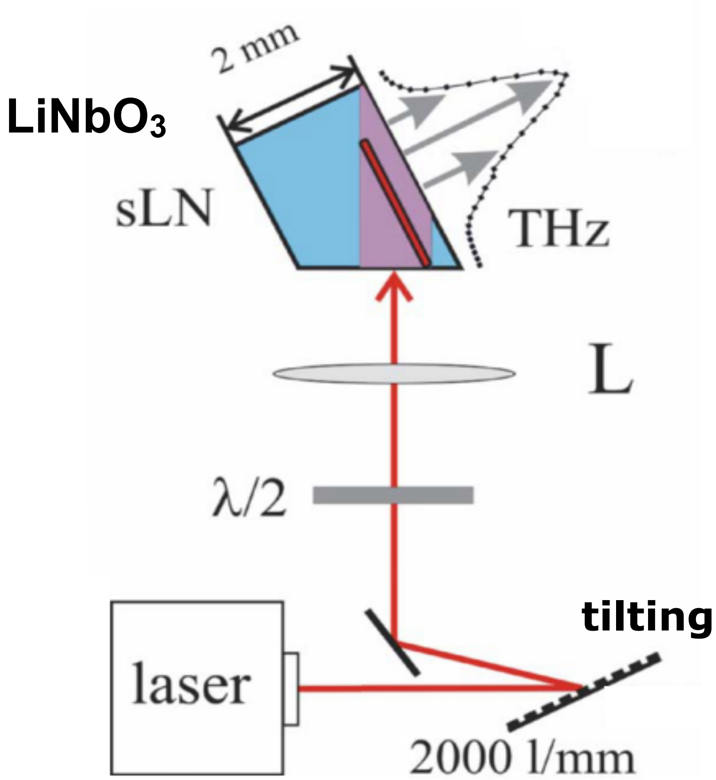


THZ: GENERATION OF INTENSE PULSES





Novelli & al. Scientific Reports 3, Article number: 1227, 2013



Hebling et al. J. Opt. Soc. Am. B Vol. 25 No. 7 (2008)

ARPES AND QM

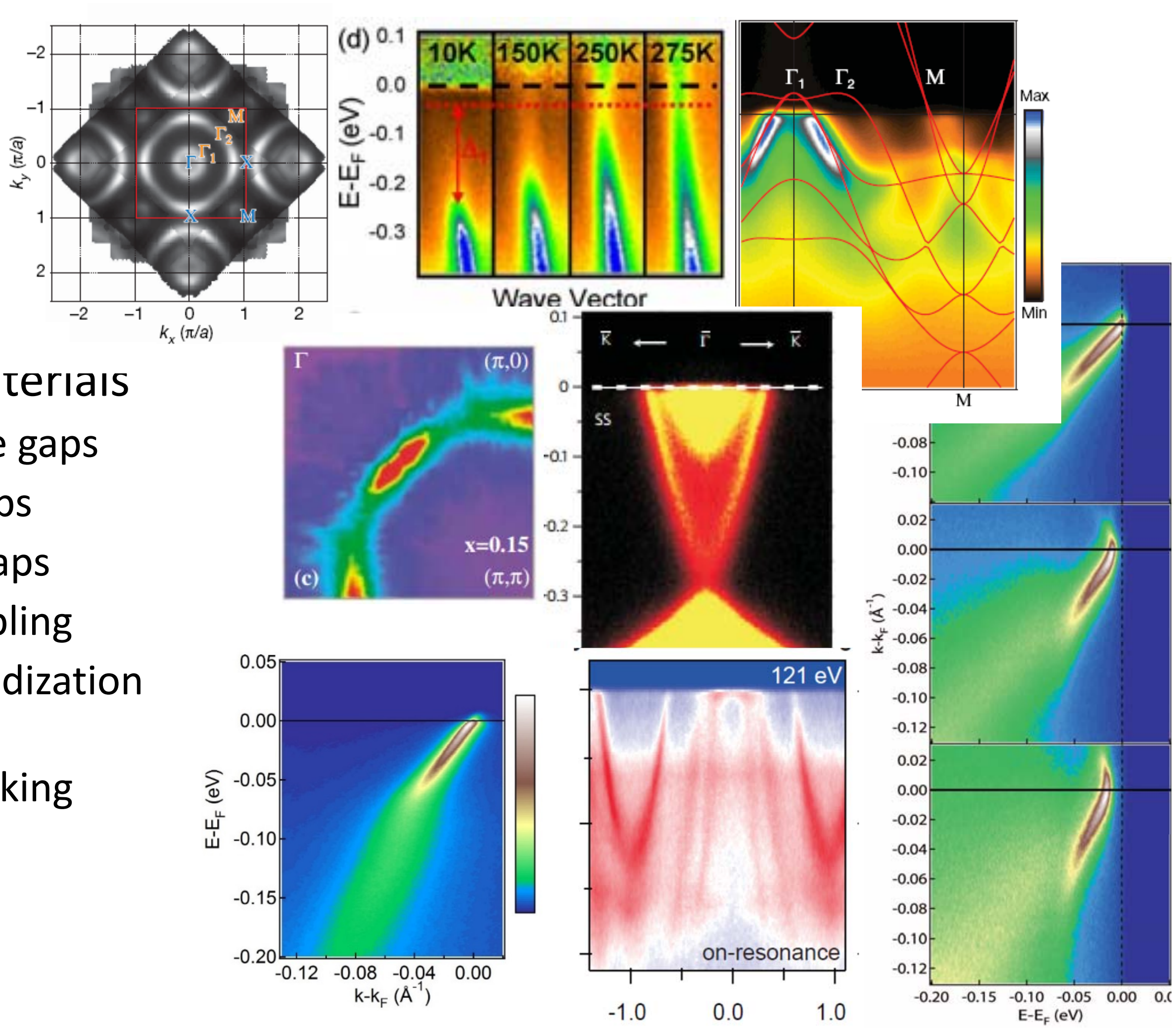
Structures in momentum space

All materials

- Brillouin zones
- Fermi surfaces
- Band dispersion

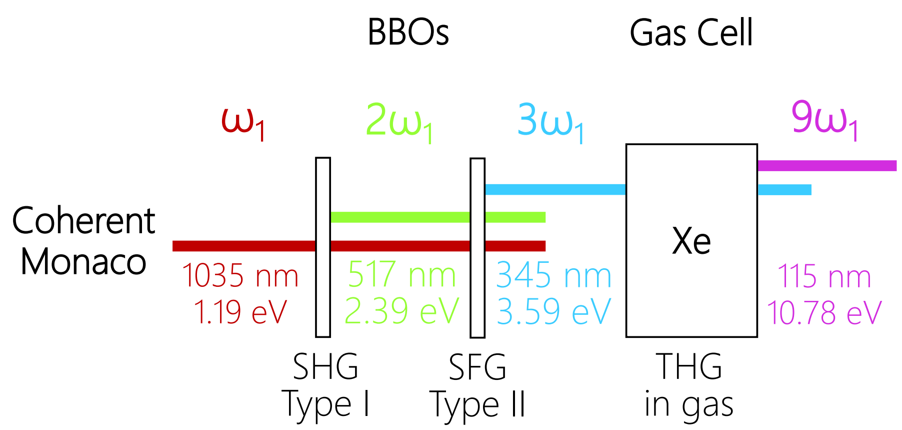
Related to quantum materials

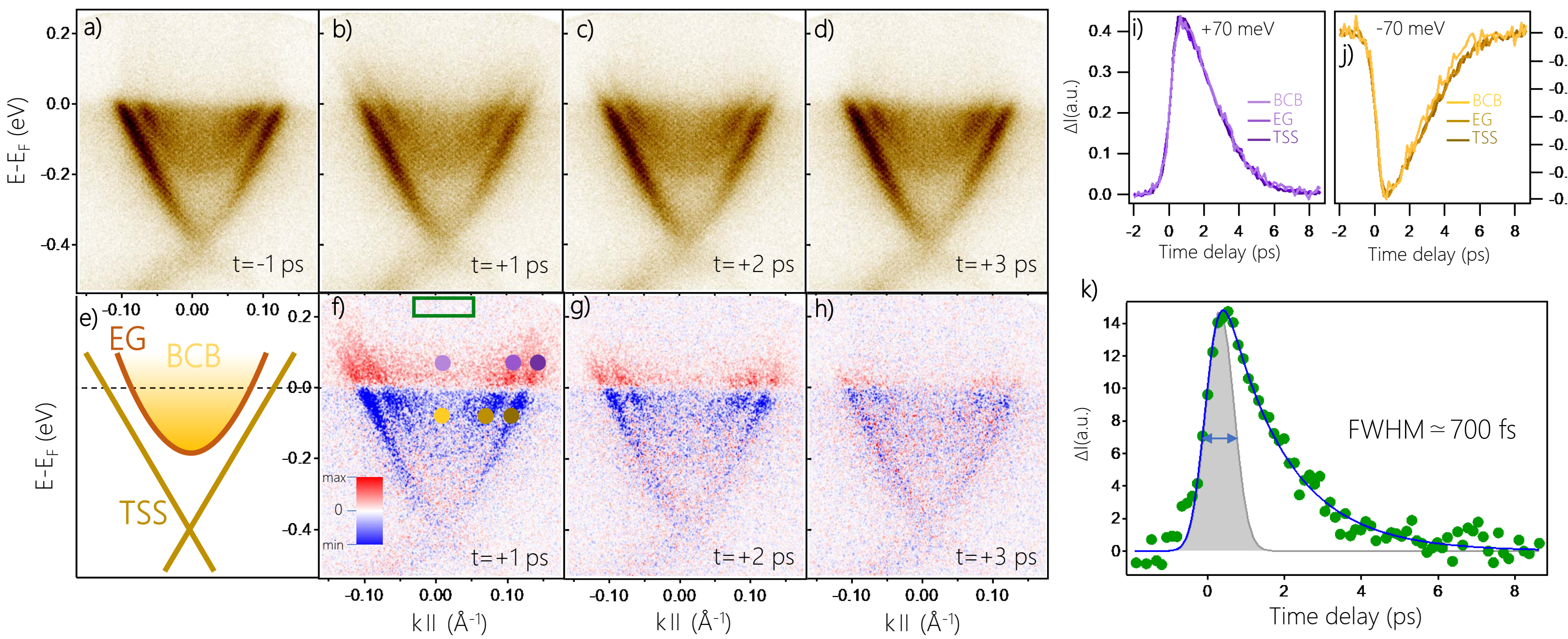
- Charge density wave gaps
- Superconducting gaps
- Spin density wave gaps
- Electron-boson coupling
- Heavy fermion hybridization gaps
- Spin momentum locking
- Dirac dispersions
- Surface states
- …



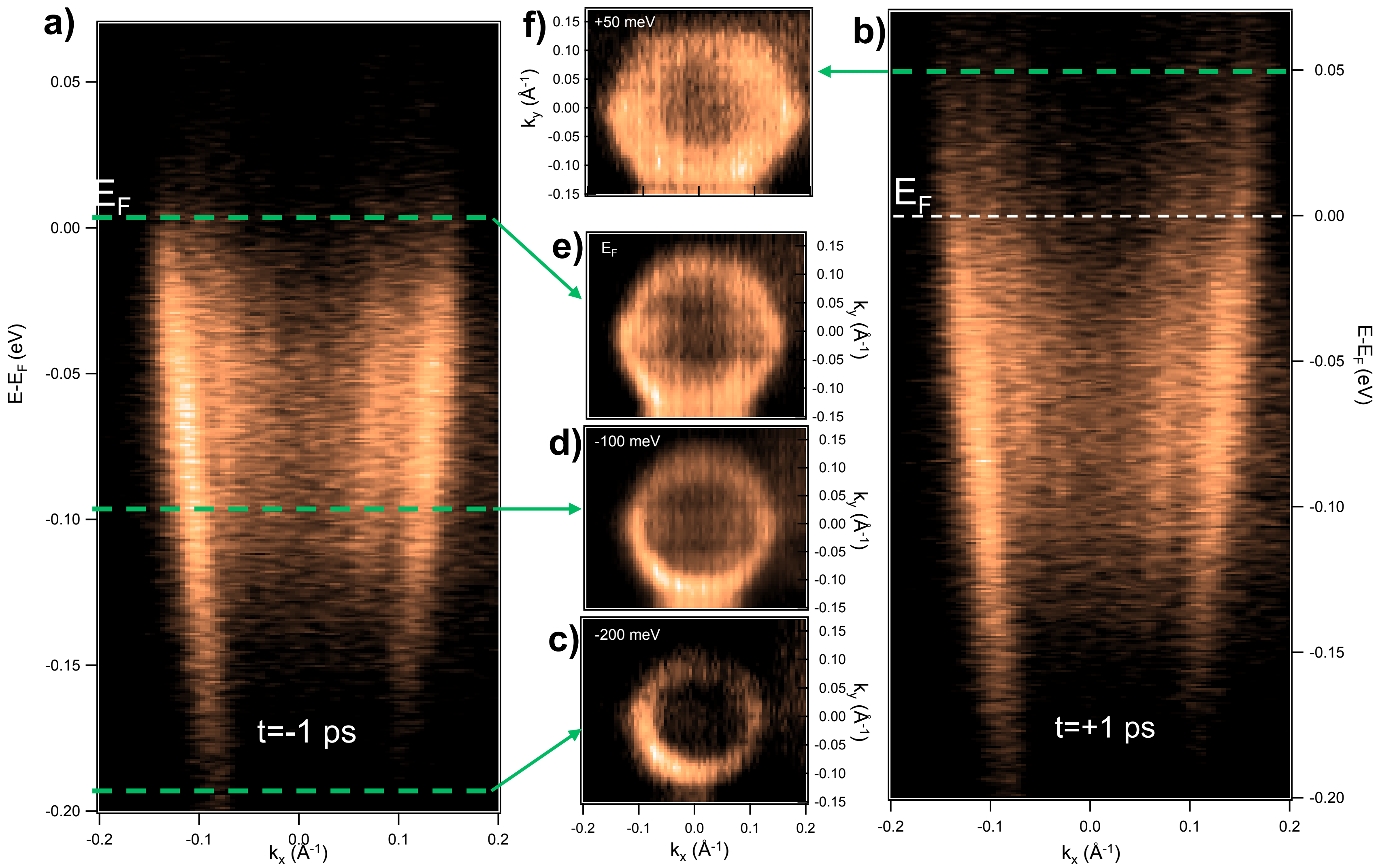
PLANTY OF ROOM FOR IMPROVING HHG SOURCES

CUTTING EDGE TR-ARPES AND QM

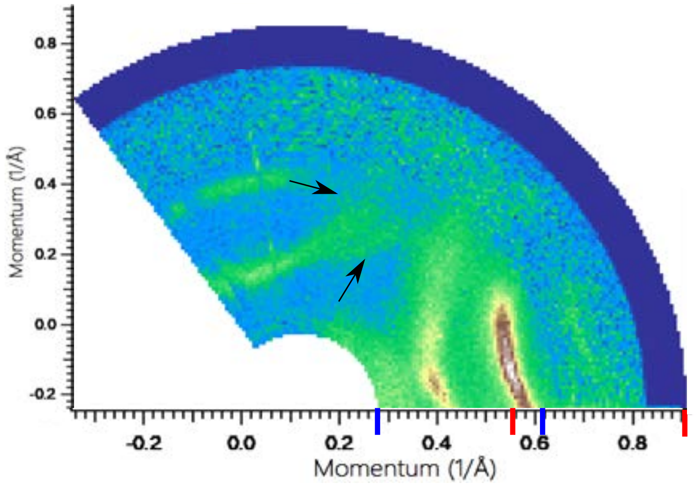


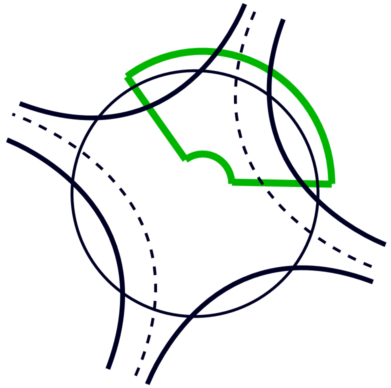


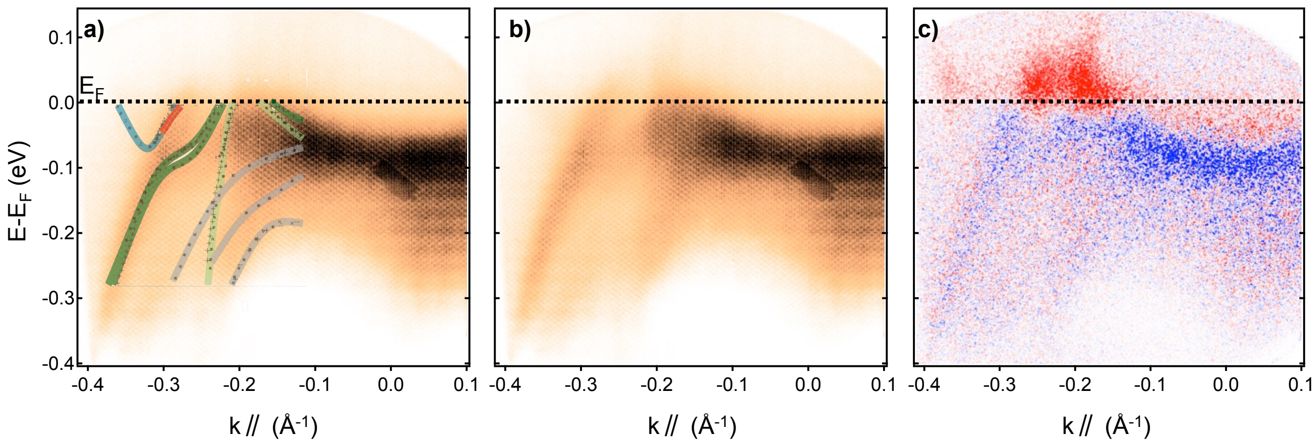
CUTTING EDGE TR-ARPES AND QM

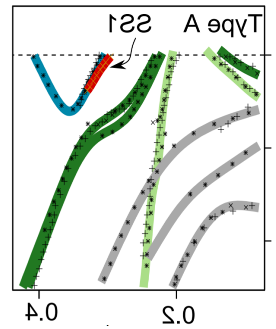


CUTTING EDGE TR-ARPES AND QM



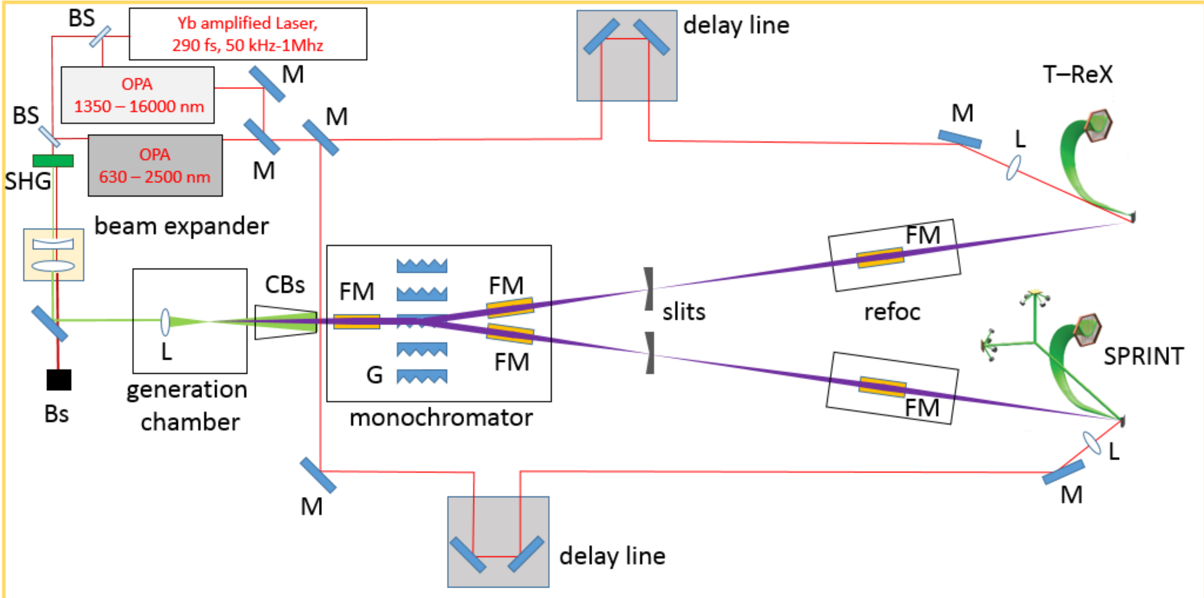




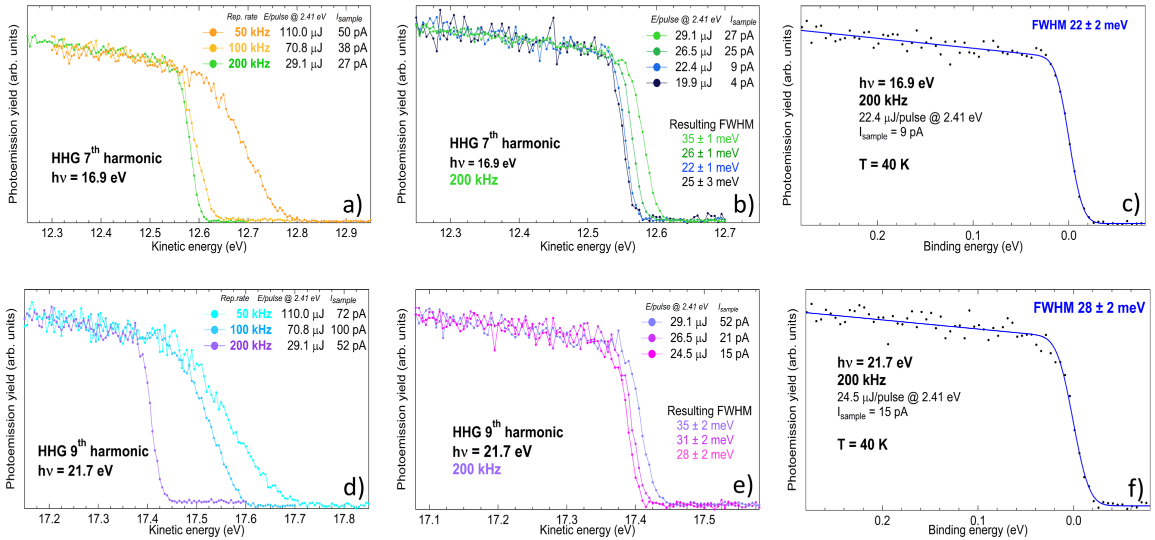


CUTTING EDGE TR-ARPES AND QM

Space Charge Free Ultrafast Photoelectron Spectroscopy on Solids by a Narrowband Tunable Extreme Ultraviolet Light Source



Parameter	Value
Photon energy (eV)	16.9
Rep. Rate (kHz)	200
ΔE_{exp} (meV)	22
Δt_{exp} (fs)	300
ΔE_{probe} (meV)	19
Δt_{probe} (fs)	105
Δt_{FT} (fs)	96



CONCLUSIONS

Future Scenario

- The way to produce fully coherent EUV soft X-ray radiation is paved.
- Tunability
- Variable polarization
- Full coherence
- High repetition rate ⟹ Controlling peak vs. average Brilliance

The future scenario

Coherent X-ray Optics

Quantum X-ray optics

Stroboscopic phase tomography

An extraordinary effort is needed to develop a suitable science program