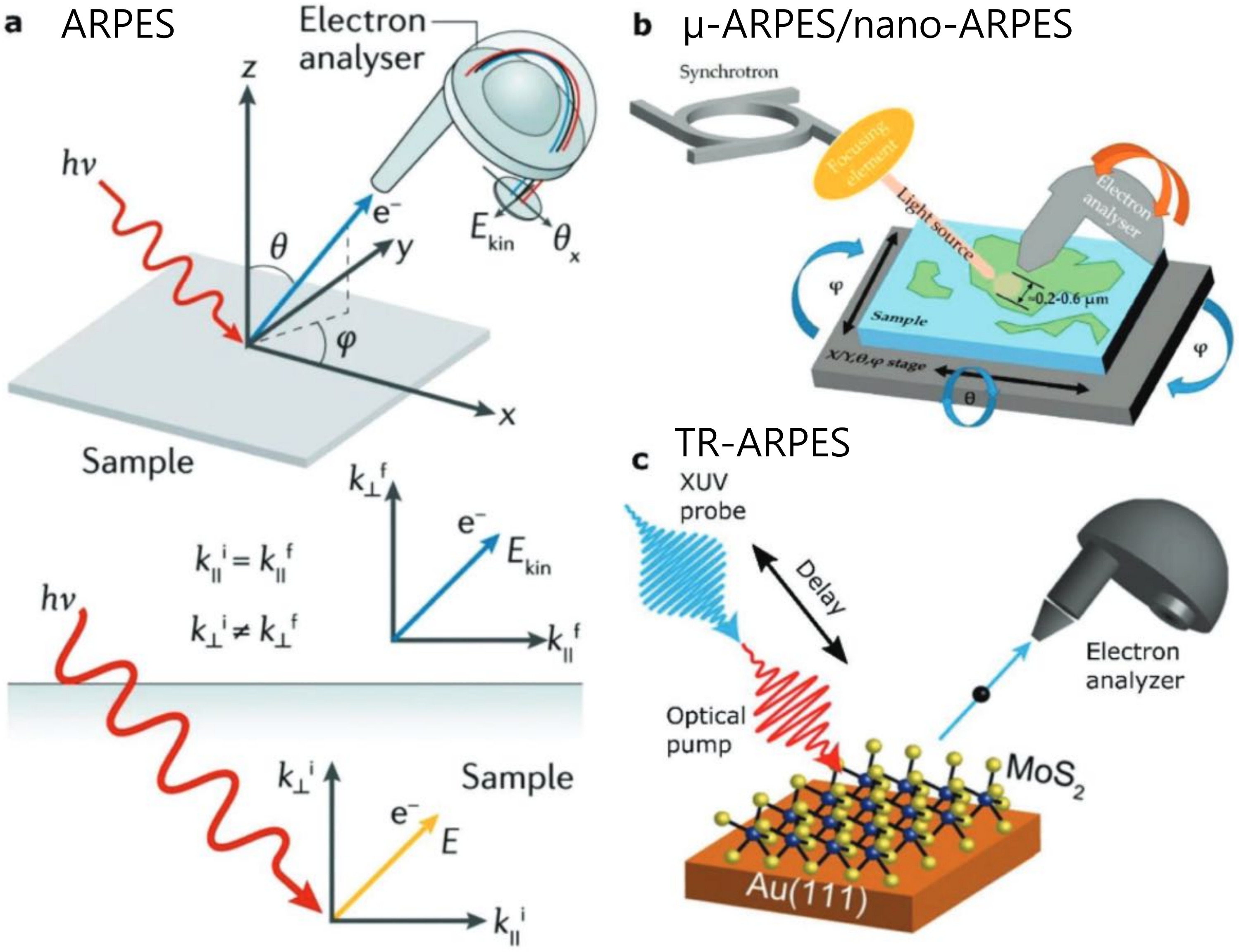
Through the Lens of a Momentum Microscope: Viewing Light-Induced Quantum Phenomena in 2D Materials

Ouri Karni, Iliya Esin, and Keshav M. Dani, Adv. Mater. **34**, 2204120 (2022)



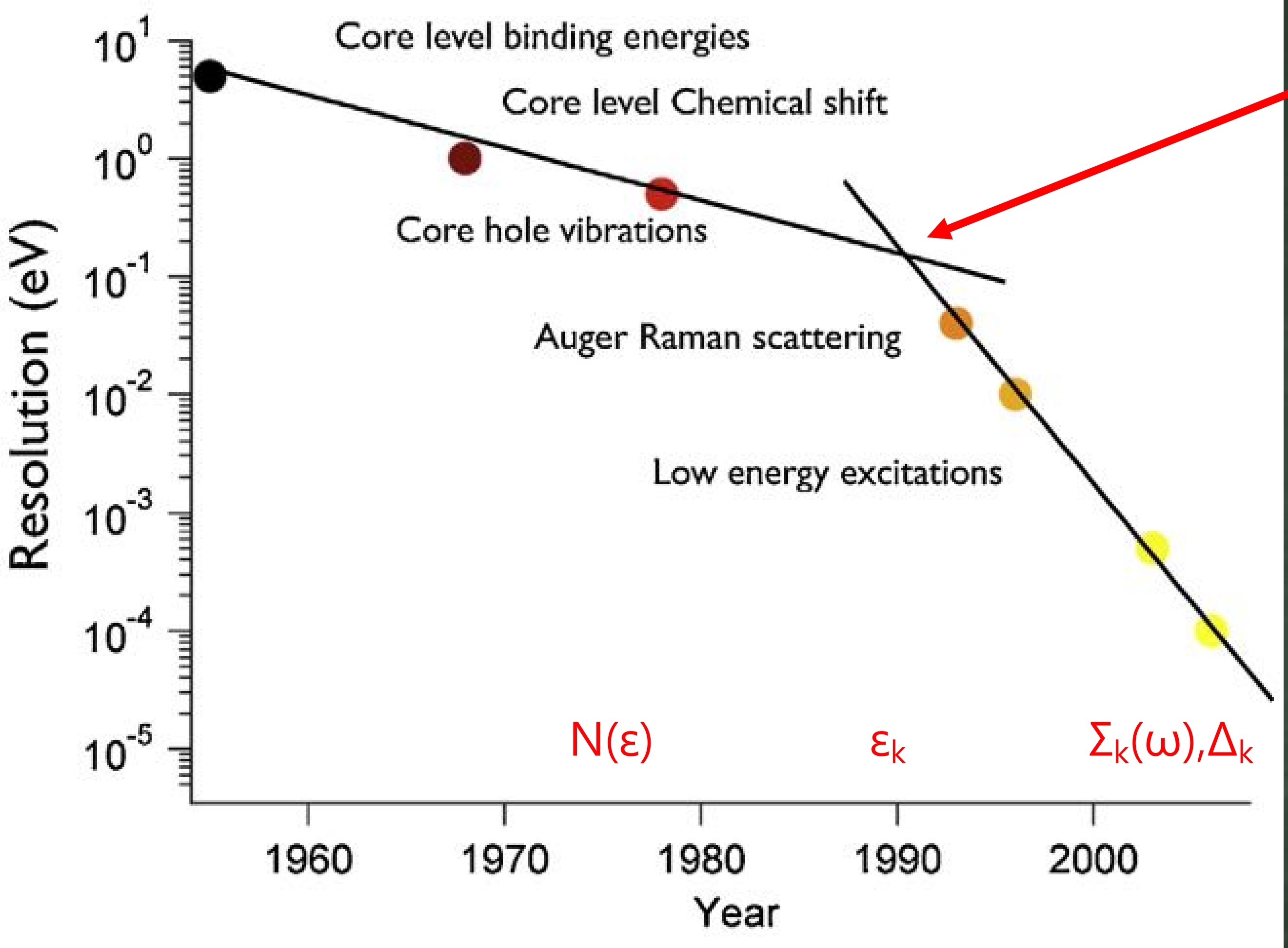
a ARPES

b μ-ARPES/nano-ARPES

c TR-ARPES

Toward Higher E/ΔE in Electron Analyzer

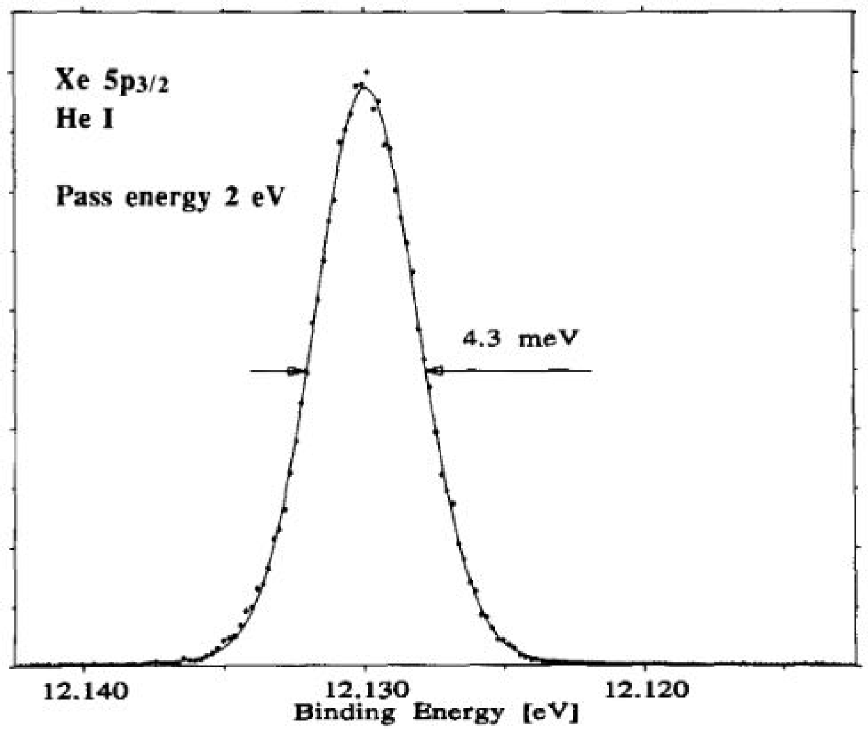
Concentric hemispherical analyzer (CHA) has been preferred.



Ovsyannikov et al., JESRP **191**, 92 (2013)

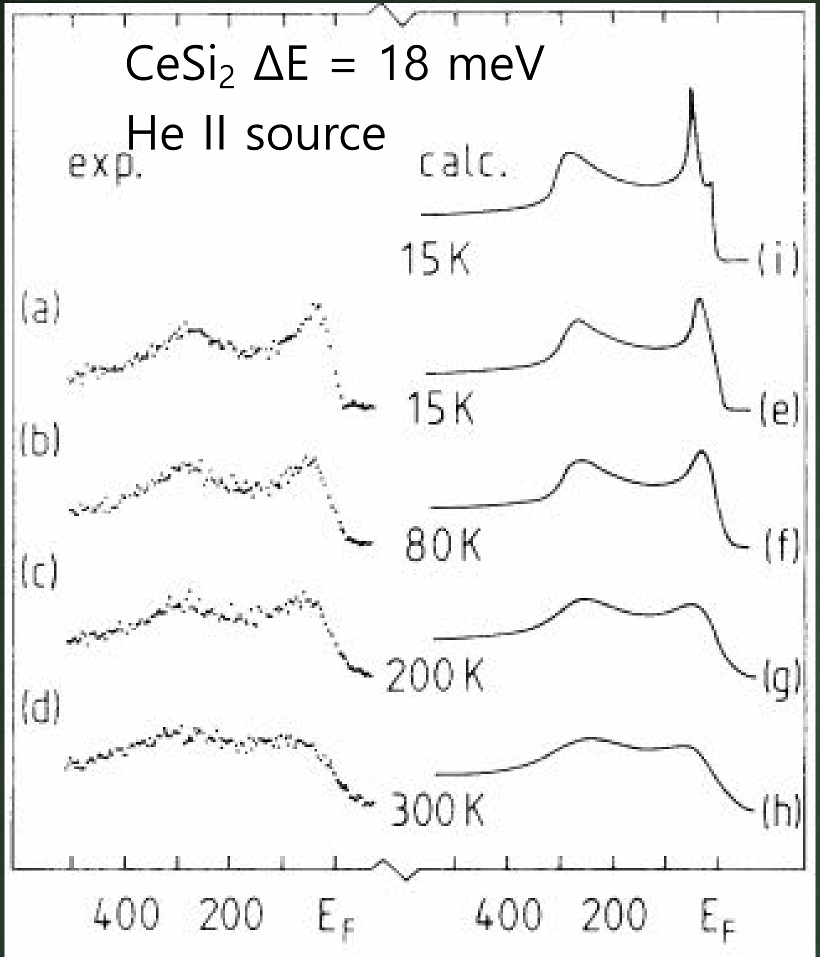
perfect control of electrostatic lens system (SIMION+PC?)

SES-200: Mårtensson et al., JESRP **70**, 117 (1994)



electrostatic lens aberration correction + high spatial resolution in electron detector

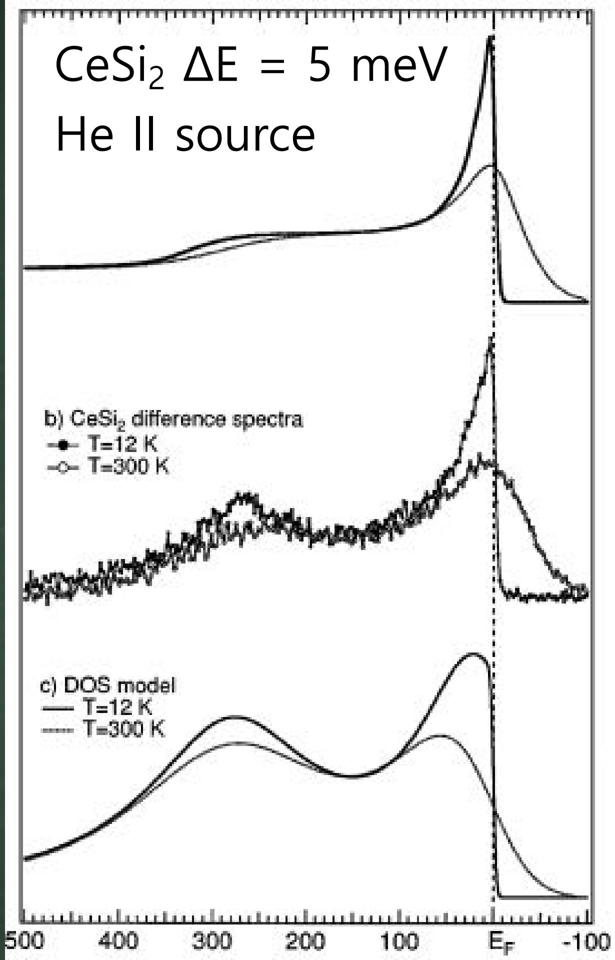
Toward Higher E/ΔE in Electron Analyzer



CeSi₂ ΔE = 18 meV

He II source

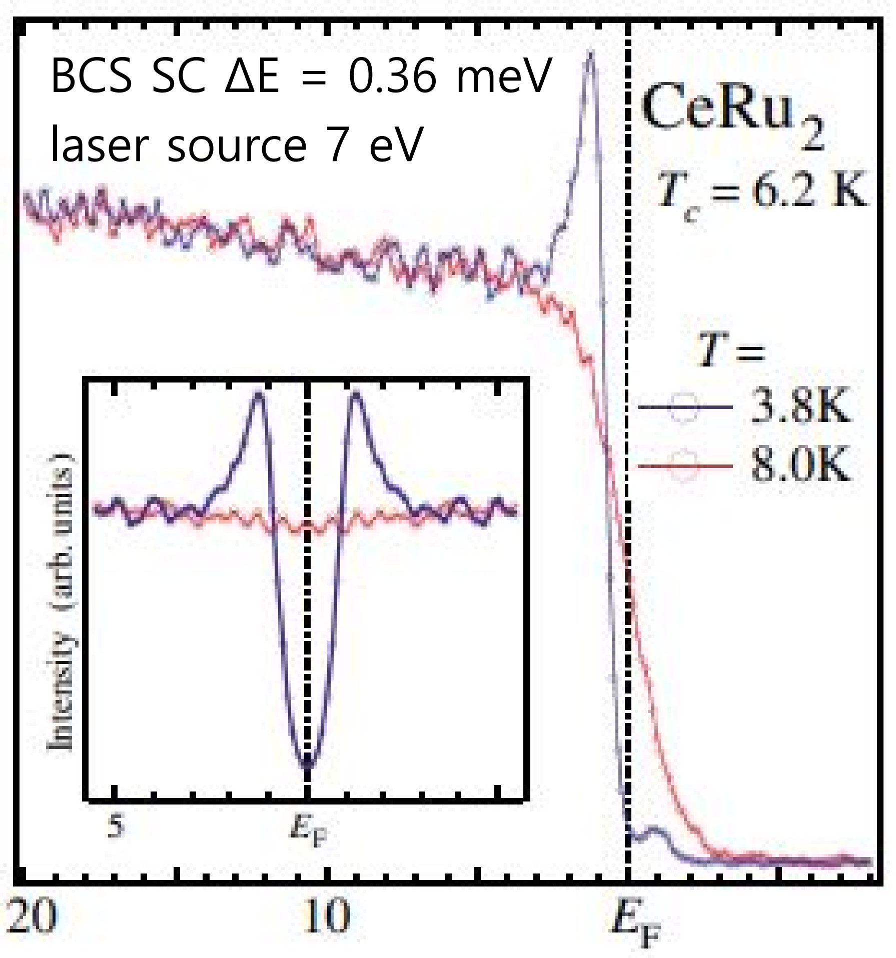
Baer group, PRL 58, 2810 (1987)



CeSi₂ ΔE = 5 meV

He II source

Baer group, PRL 78, 4127 (1997)



BCS SC ΔE = 0.36 meV

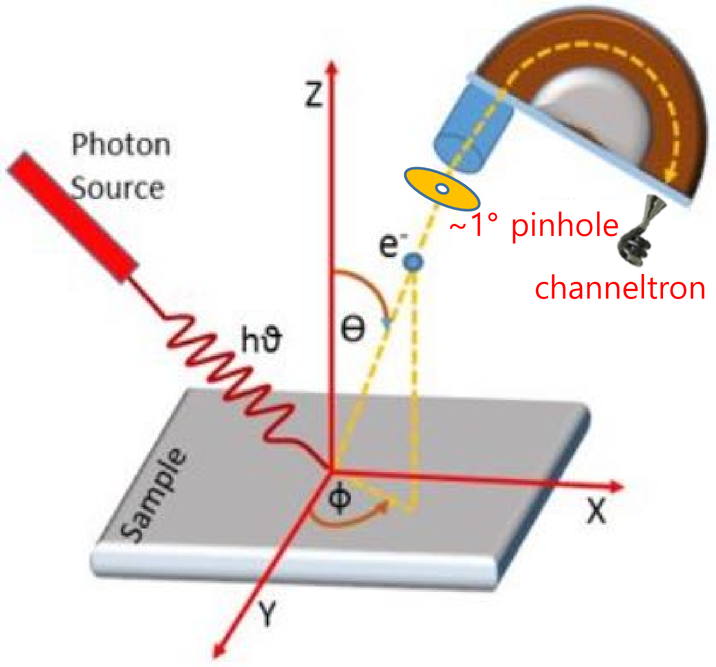
laser source 7 eV

Shin group, PRL 94, 057001 (2005)

Stable electronic ground (< 0.1 meV) is essential.

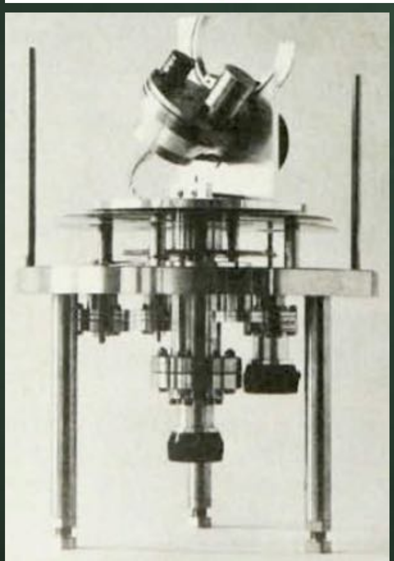
High energy resolution requires low temperature < 10 K (300 K ~ 25 meV → 0.1 eV width in Fermi function)

ARPES in Old Days



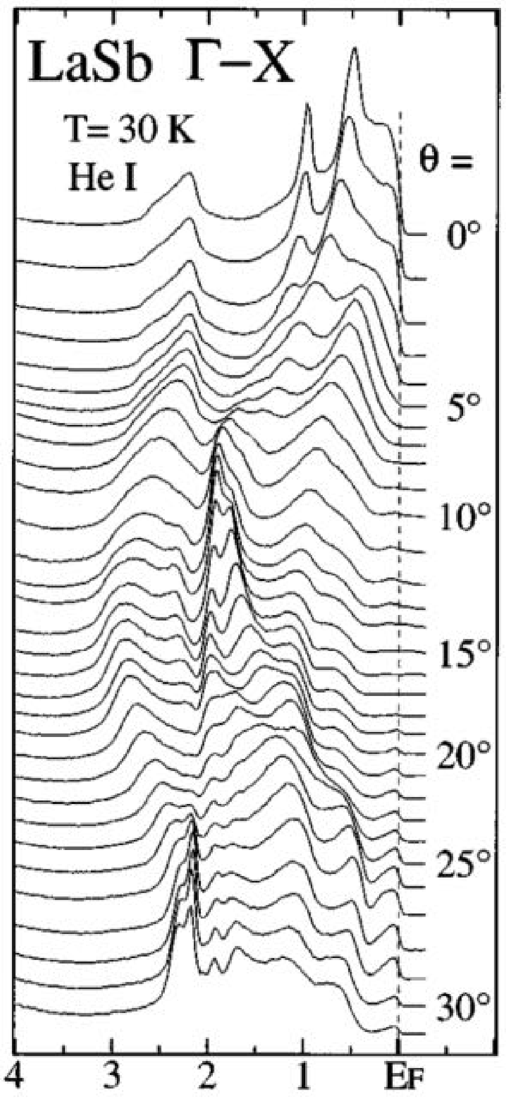
~1° pinhole

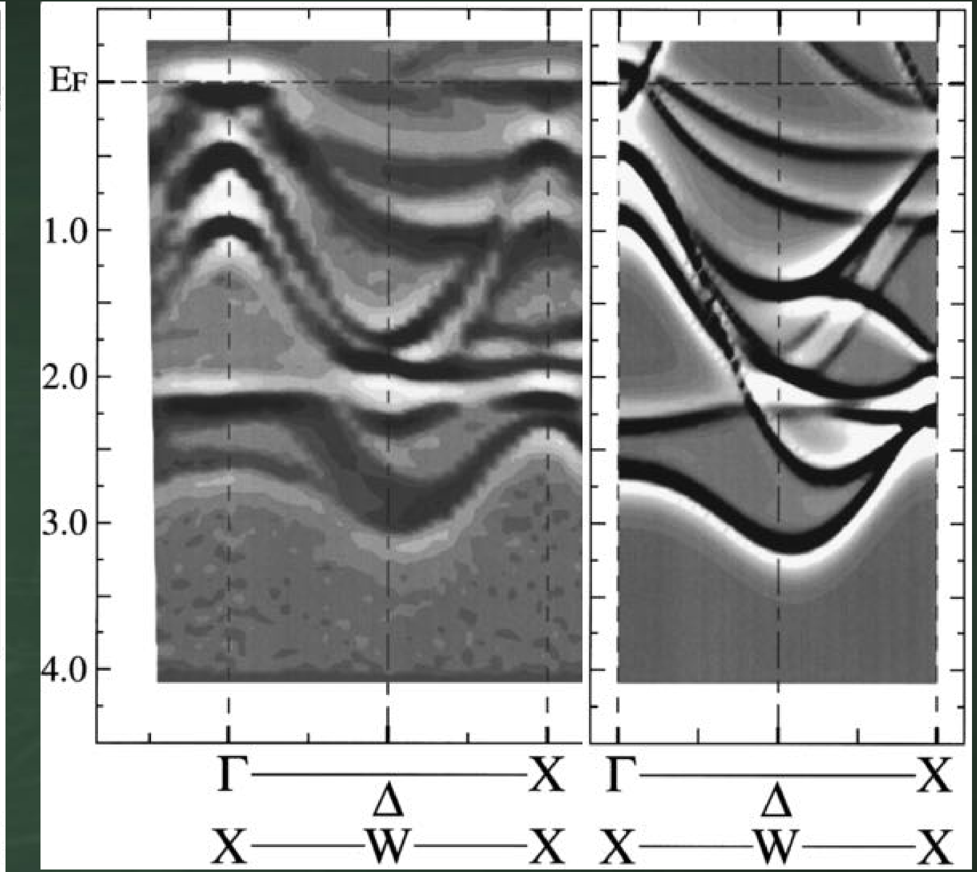
channeltron



rotate sample
or analyzer

VSW Ltd. Phys. Today '83

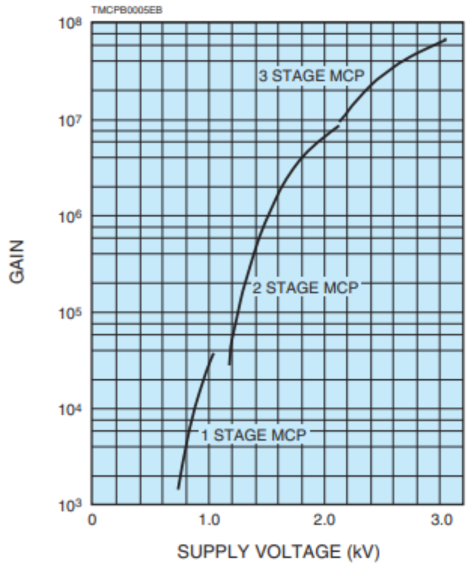


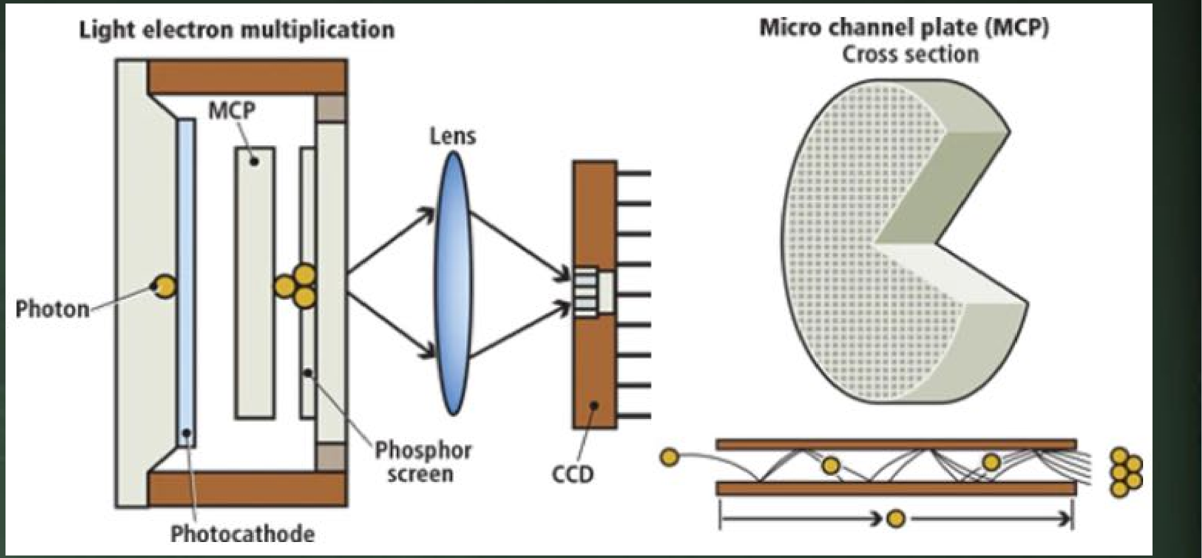


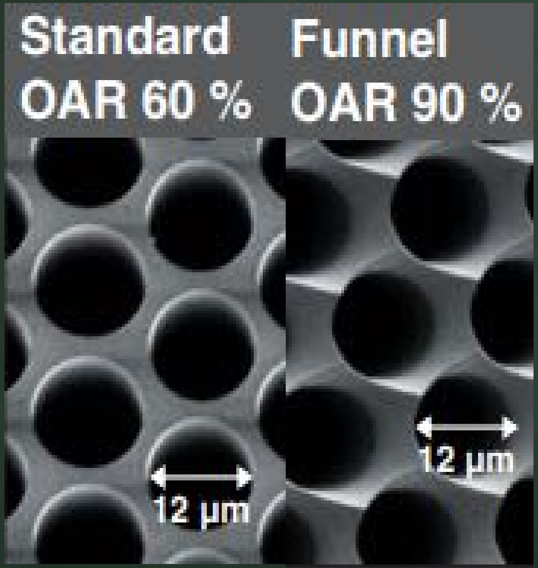
Takahashi group, PRB **58**, 7675 (1998)

2 day measurements!

Two-Dimensional Electron Detector: MCP + Phosphor Screen + CCD







MCP size: up to ϕ77 mm by Hamamatsu https://www.hamamatsu.com

up to 20 x 20 cm^2 by Incom, Inc. https://incomusa.com

spatial resolution ~20 μm

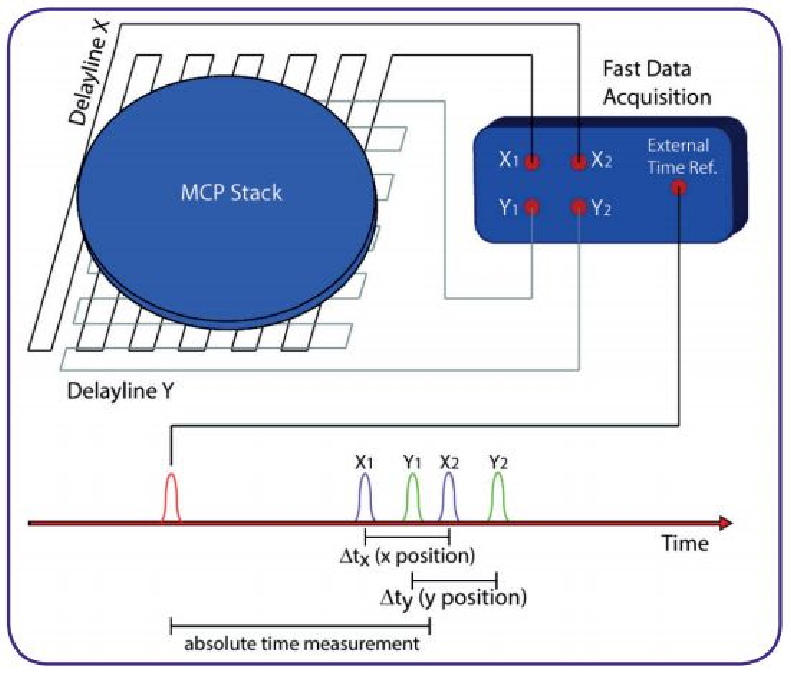
phosphor screen P43(Gd_2O_2S:Tb):

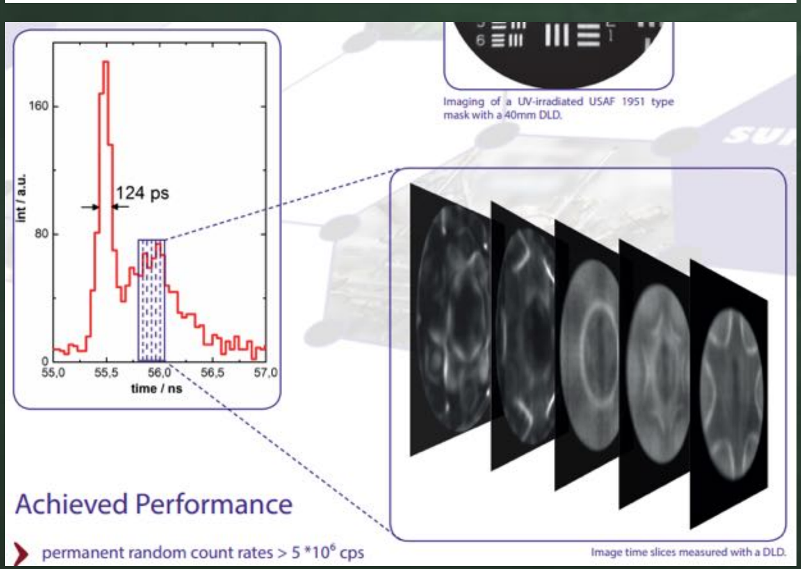
best efficiency, λ = 545 nm (green),

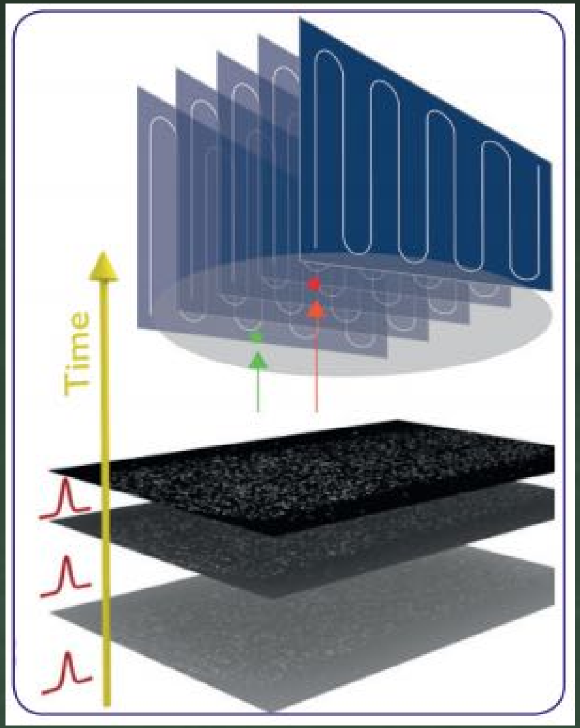
spatial resolution: ~100 μm (~300 ch for ϕ40 MCP → $\Delta\theta \sim 0.1°$ for $\theta \sim \pm 15°$)

10% decay time 1 ms → not suitable for time-resolved measurements

Two-Dimensional Electron Detector: MCP + DLL







multi-hit design

Active diameter: 10 – 150 mm

Spatial resolution: down to 30 μm

Multi-Hit designs: > 10 hits

Time resolution: < 200 ps

Repetition rate: 9 MHz

cf. Ti:Sapphire fs laser up to 80 MHz

single-bunch storage ring ~ 1 MHz

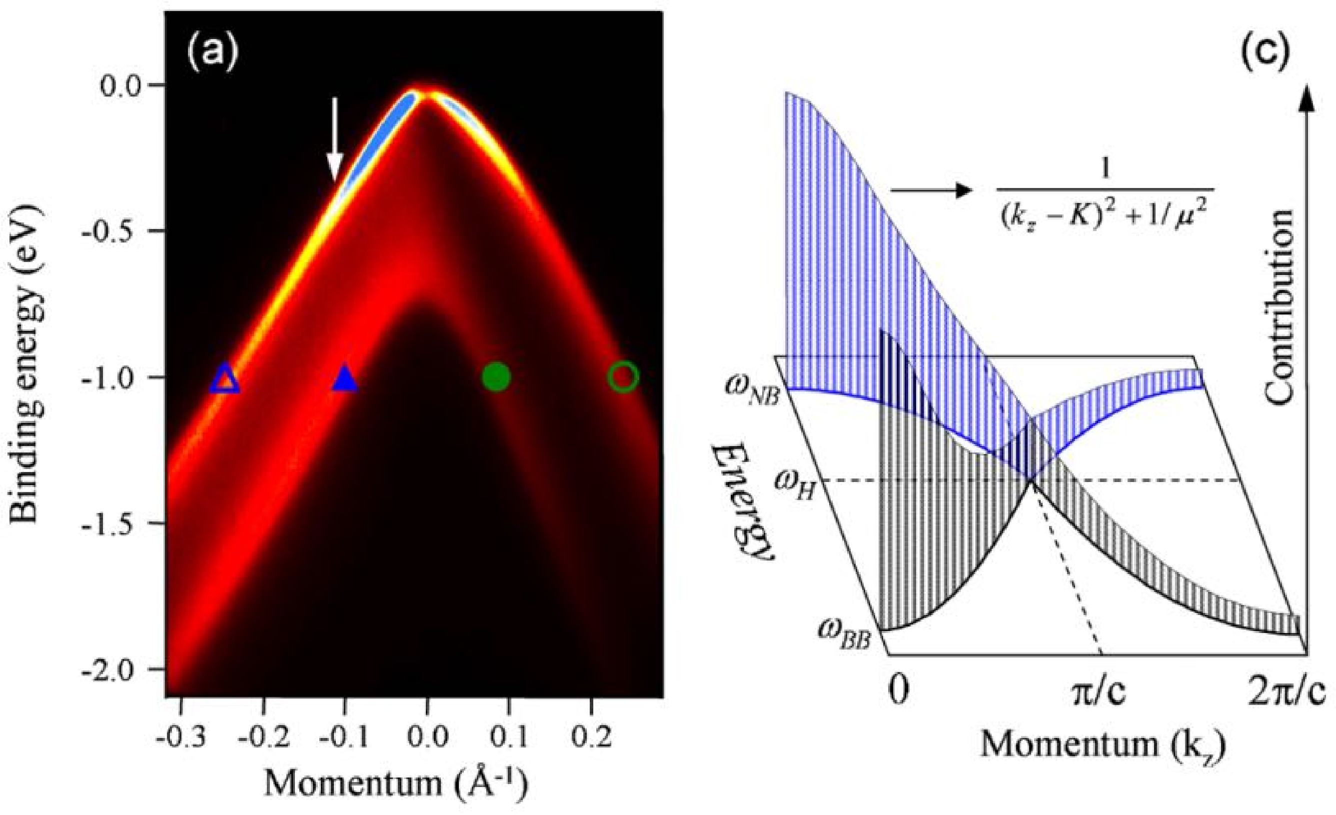
https://www.surface-concept.com/downloads/info/delaylinedetectors.pdf

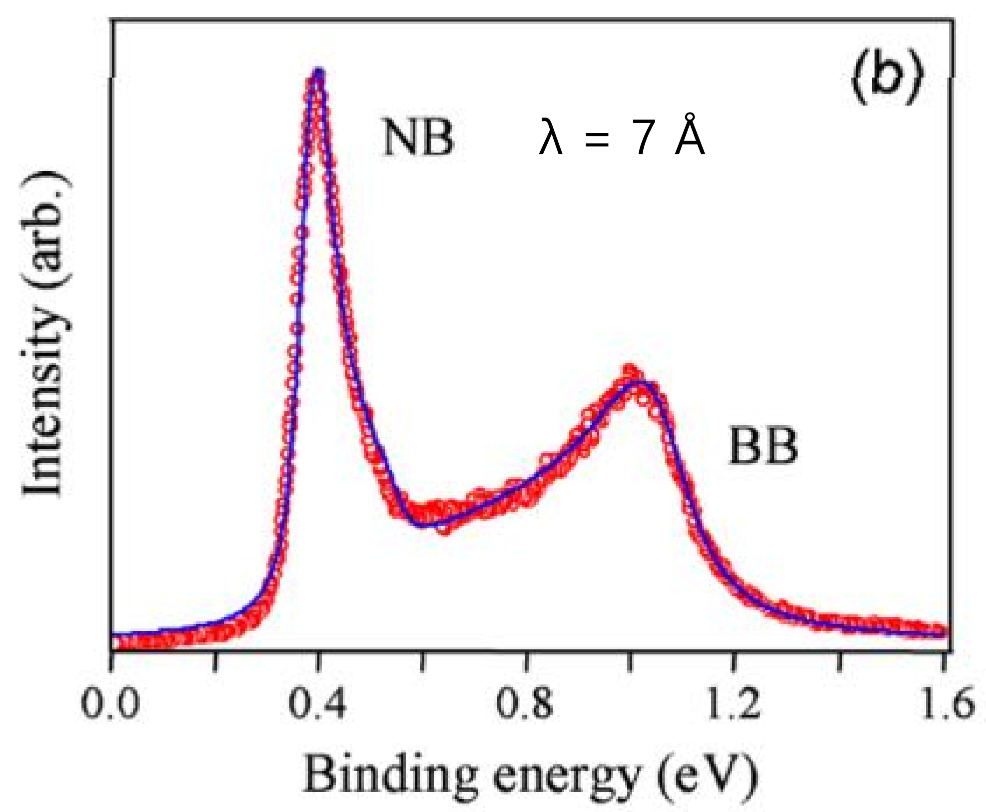
Effects of Finite $\Delta k_\perp$ in ARPES Even in Quasi-2D System

Leem et al., PRL **100**, 016802 (2008)

In most ARPES, $\Delta k_\perp > 0.1$ Å^{-1} due to photoelectron escape depth

graphite

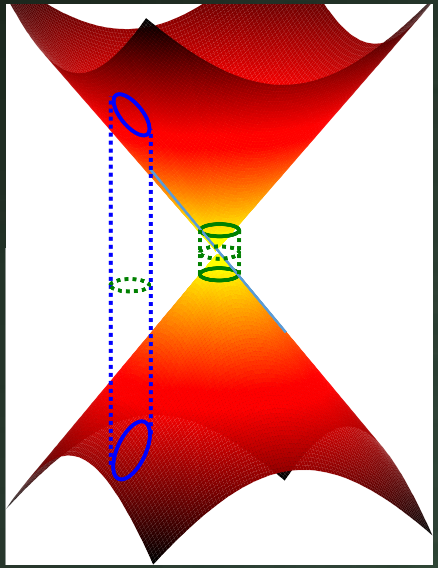


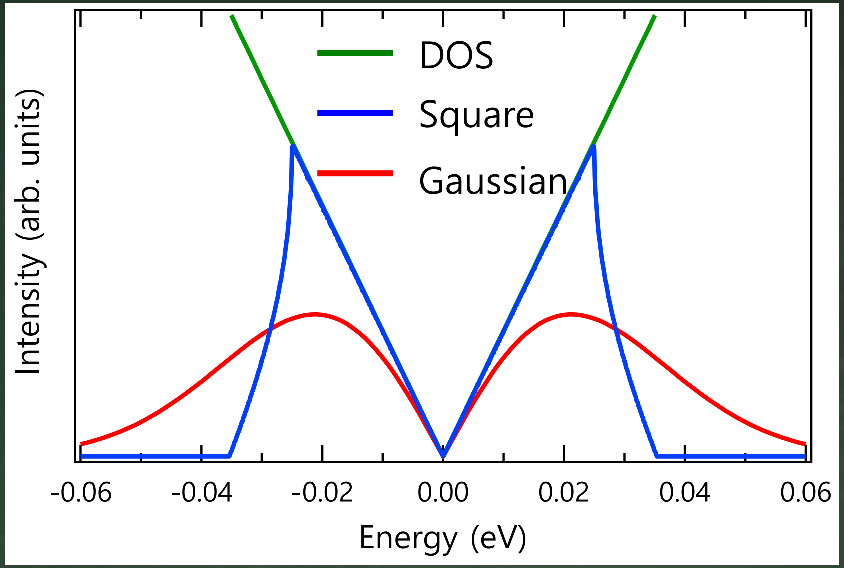


Small beam size is essential for Van der Waals 2D materials.

Importance of $\Delta k_{\parallel}$ in ARPES especially for Dirac fermions

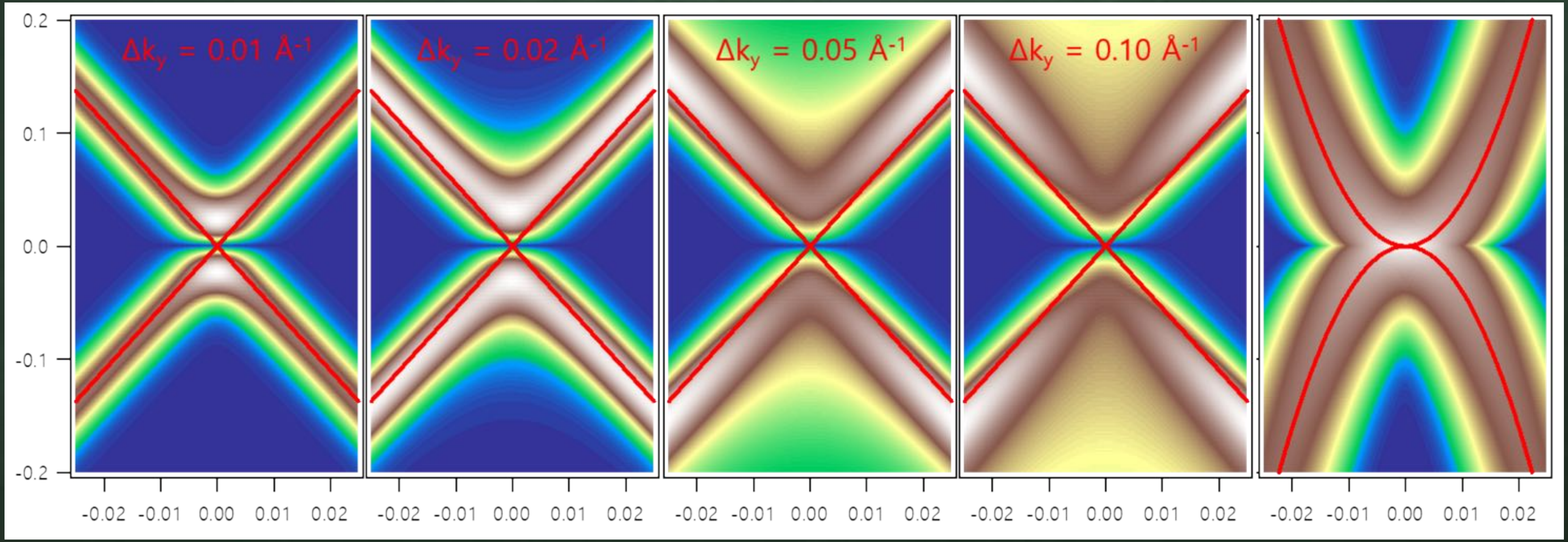
HD Kim unpublished

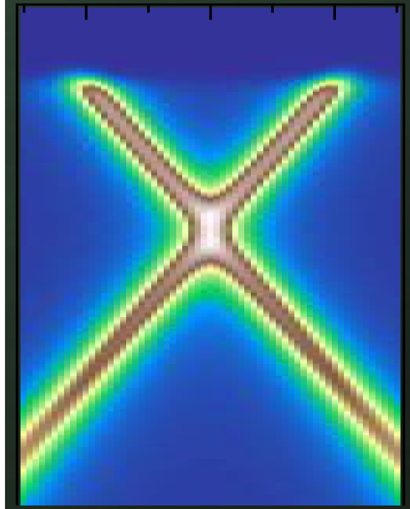




High $\Delta k_{\parallel}$ is essential to probe Dirac/Weyl fermions.

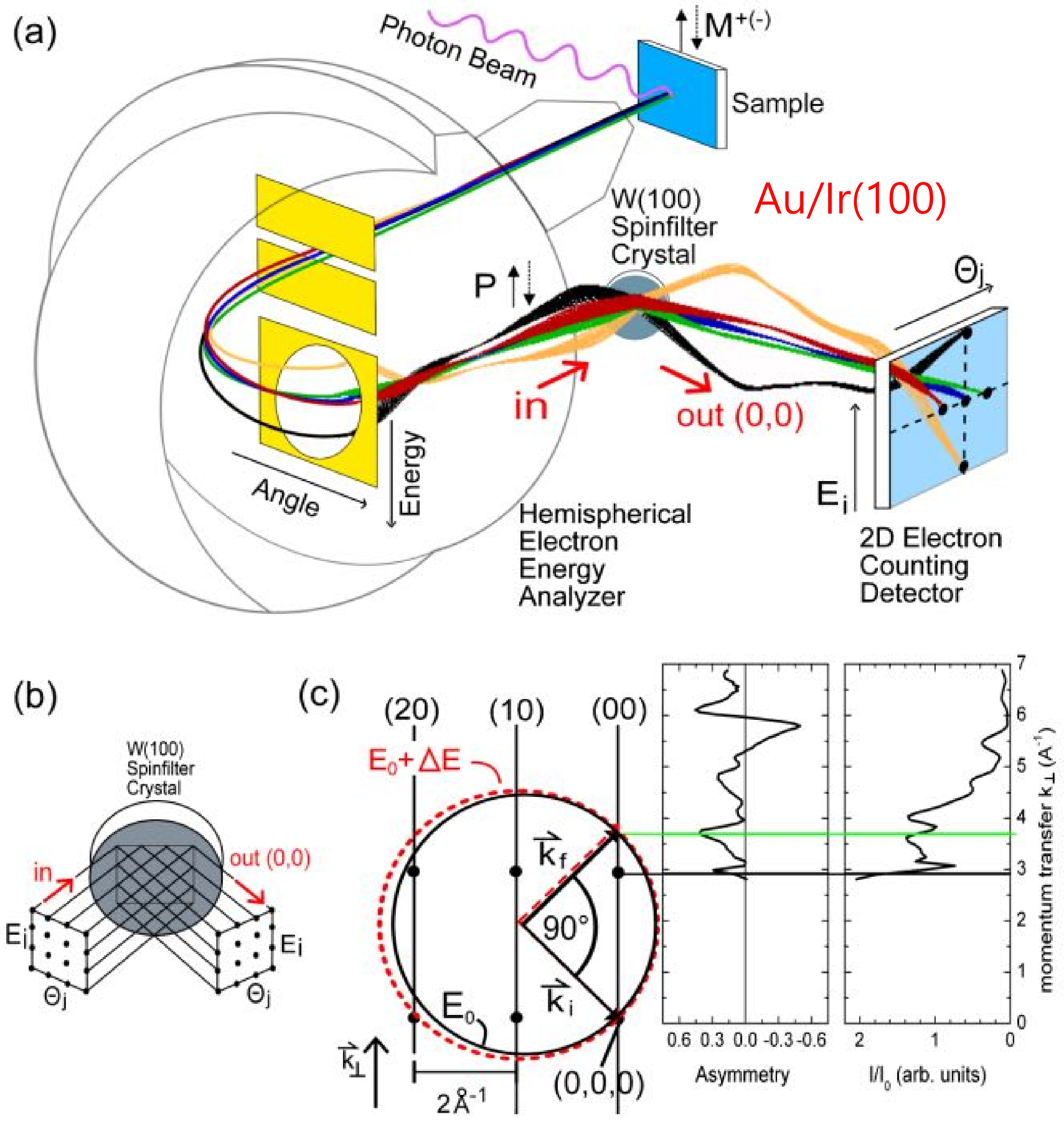
Single Dirac energy can be measured only in STM.





Multichannel Spin-Polarization Detector @ Mainz

Kolbe et al., PRL **107**, 207601 (2011)



Using W(100) or Au/Ir(100) spin-filter crystal,

specular geometry

spin-polarized low-energy electron diffraction

$k_{\parallel}$ conservation → 2D lateral image preservation

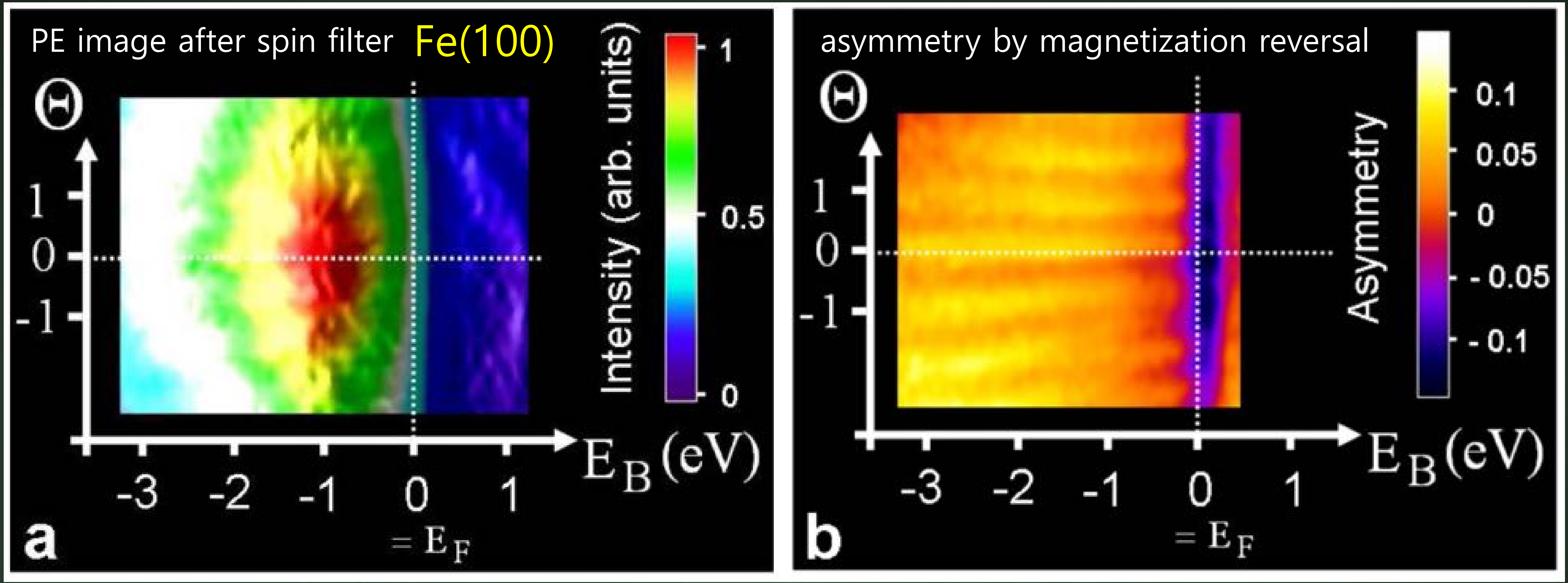
4-bundle electron-optical simulation

optimum working point:

- scattering energy 26 eV
- reflectivity R = 1.2%
- asymmetry S = 0.43

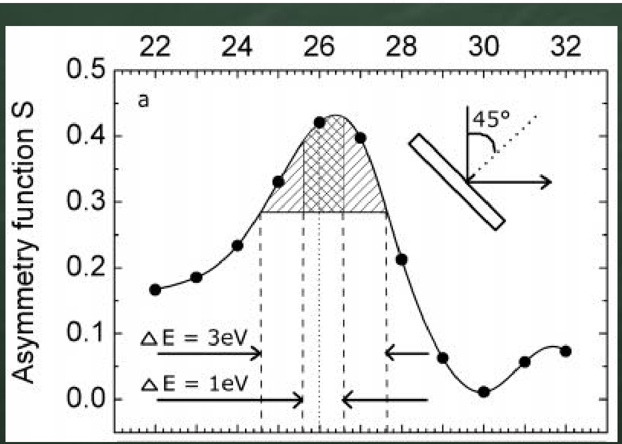
Multichannel Spin-Polarization Detector @ Mainz

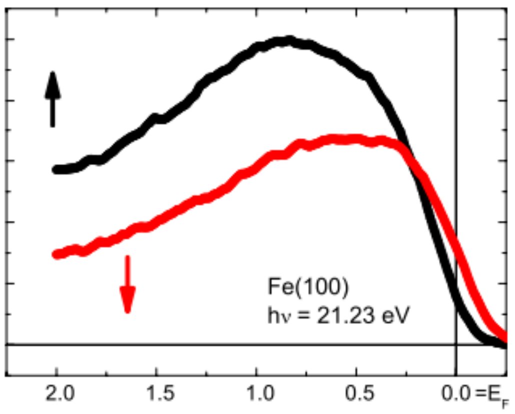
Kolbe et al., PRL **107**, 207601 (2011)



spin polarization

$$P_{ij} = \frac{A_{ij}}{S_{ij}} = \frac{I_{ij}^{+} - I_{ij}^{-}}{I_{ij}^{+} + I_{ij}^{-}}\frac{1}{S_{ij}}$$

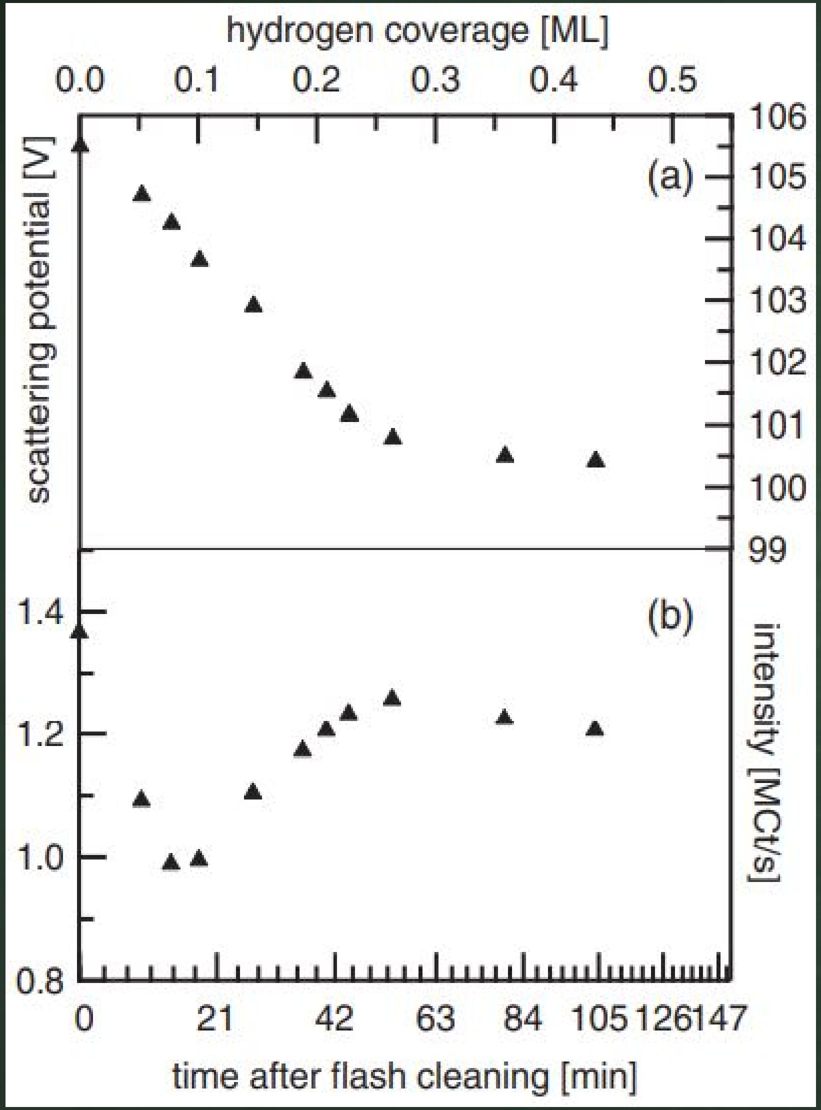


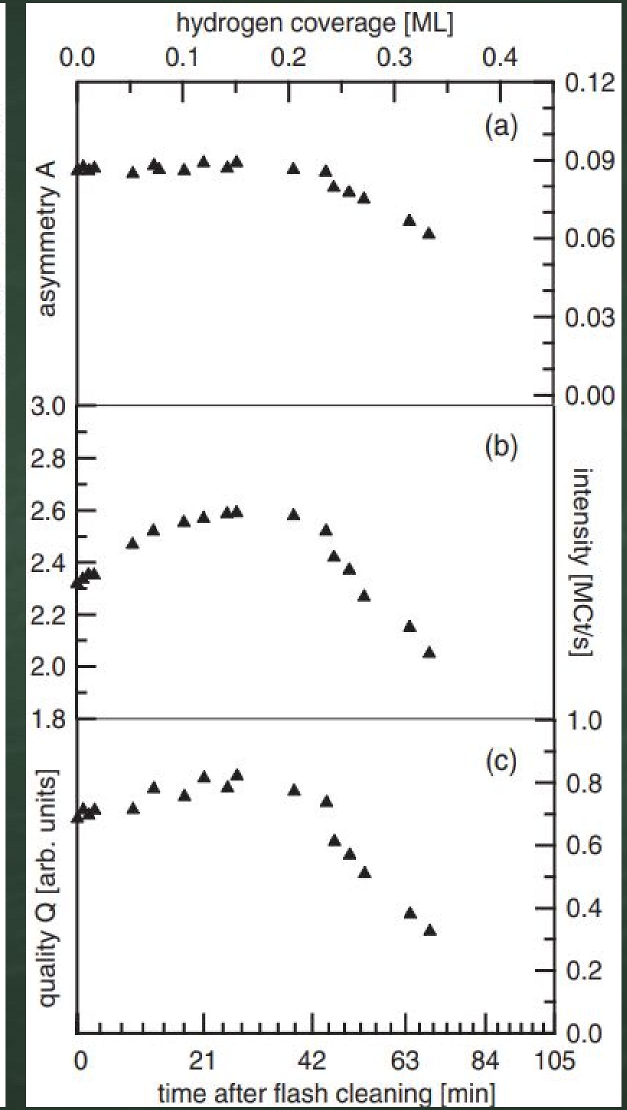


Au-Passivated Ir(100) Spin Filter by MPI Halle

Kirschner group, PRB **88**, 125419 (2013)

surface degradation of W(100) spin filter





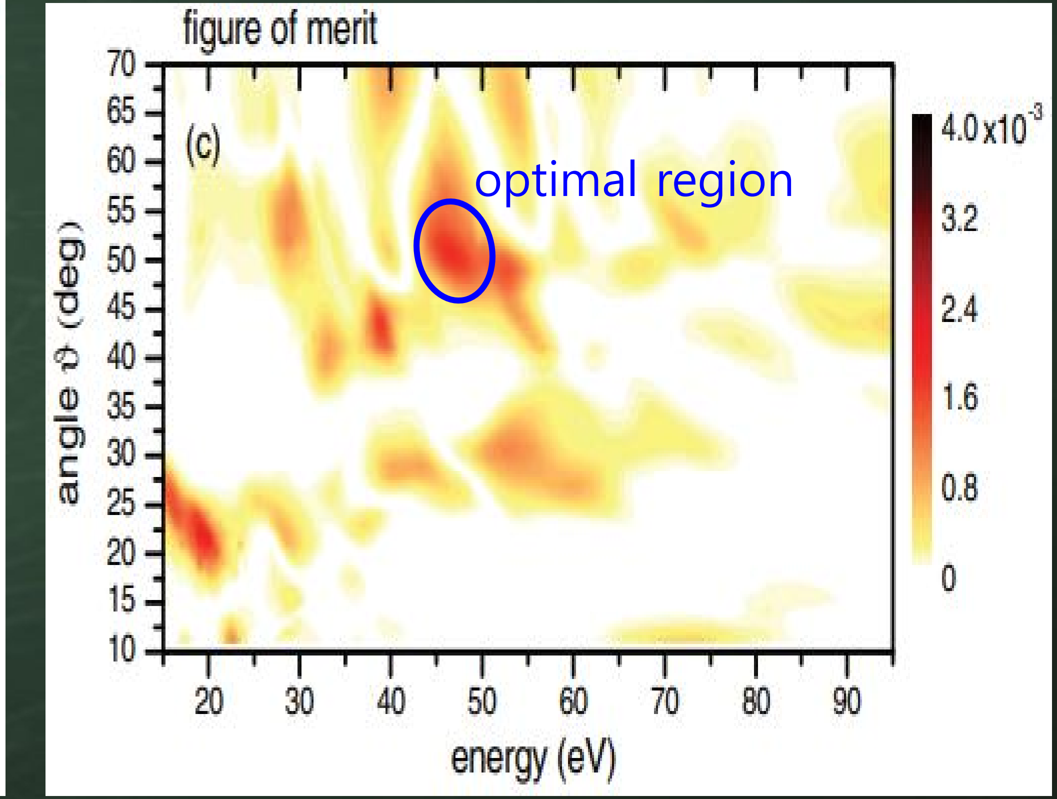
Lofink et al., RSI **83**, 023708 (2012)

Au/Ir(100)

several month lifetime by Au passivation

high spin-asymmetry up to 77%

useful width ~2 eV



Photoemission Momentum Microscopy (PEkM)

Measure ARPES in 2D k-space

(1) High-Resolution Time-of-Flight Analyzer + MCP + DLL

2D k-space image + spectrum (pulsed photon source (pulse width < 10 ps))

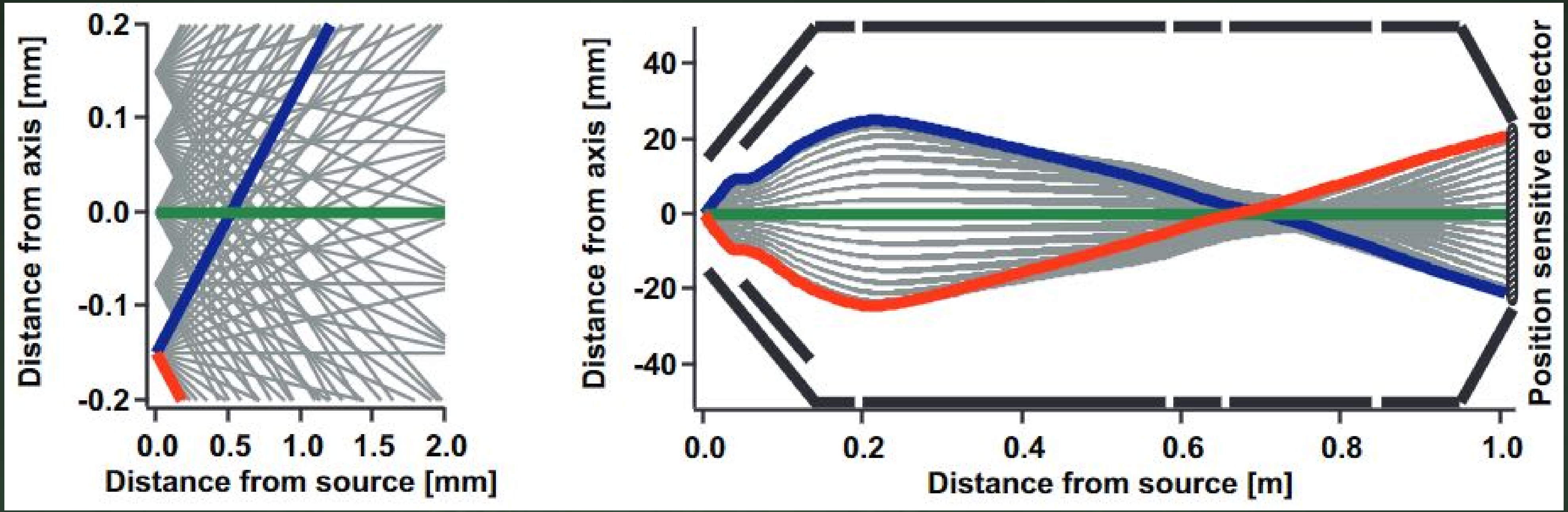
(2) PEEM Electrostatic Lens + 2 CHA + MCP + Phosphor Screen + CCD Camera

2D k-space image with fixed energy (CW or pulsed photon source)

(3) PEEM Electrostatic Lens + High-Resolution Time-of-Flight Analyzer + MCP + DLL

High-Resolution Time-of-Flight Analyzer (ArToF10k)

Ovsyannikov et al., JESRP **191**, 92 (2013)



$$v = \sqrt{2E/m} = \sqrt{\frac{2E \times 1.6 \times 10^{-19}\ \mathrm{J/eV}}{9.11 \times 10^{-31}\ \mathrm{kg}}} = 0.6 \times 10^{6}\sqrt{E(\mathrm{eV})}\ \mathrm{m/s}$$

$$E = \frac{1}{2}m(d/t)^2 \longrightarrow |E/\Delta E| = |t/2\Delta t|$$

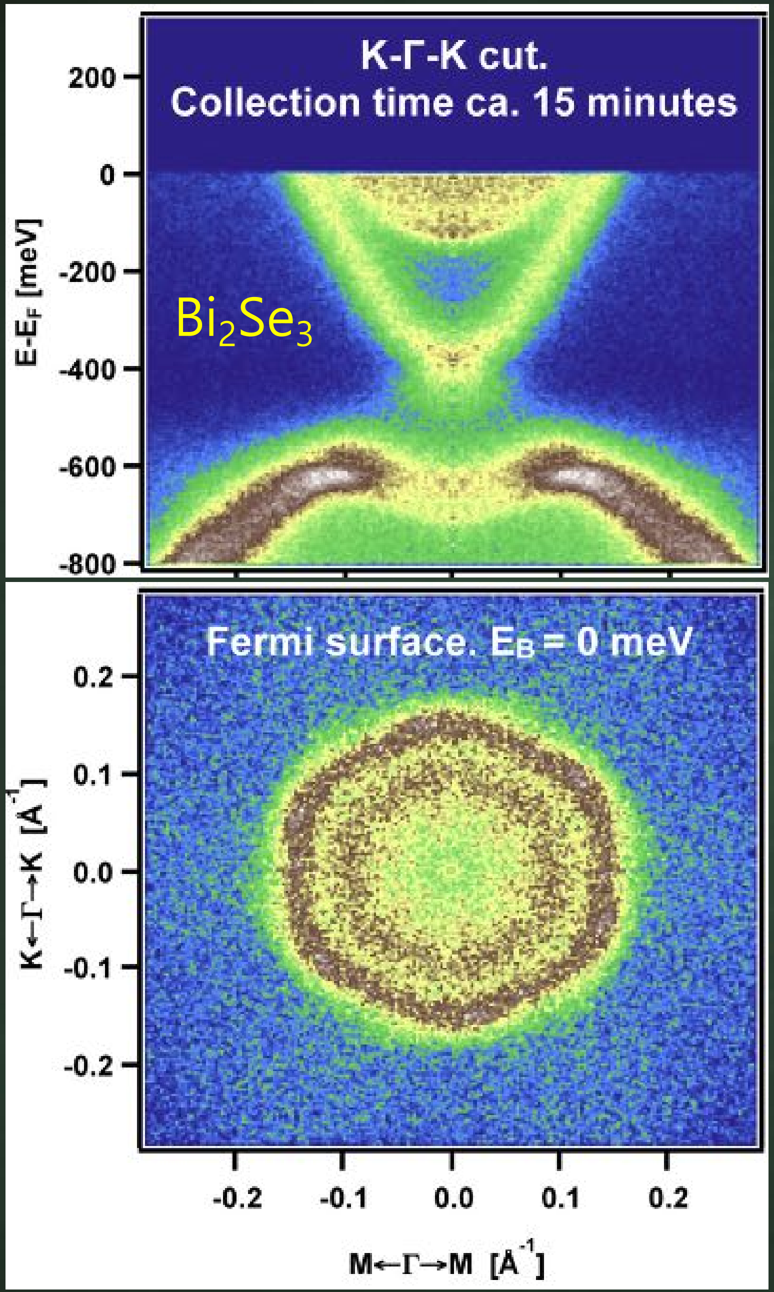
drift energy E_d = 3 eV, v = 10^6 m/s, 1 m drift → 1 μs

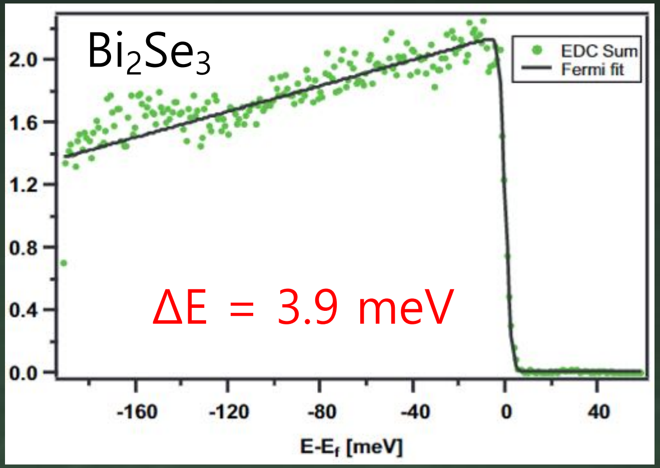
time resolution of electronics and detector ~ 0.1 ns → E/ΔE = 5,000

$\Delta E = \sqrt{(\alpha E^{3/2}\Delta t)^2 + (\beta E\Delta d^{\gamma})^2} \longrightarrow \Delta E = \alpha E^{3/2}\Delta t$ when E > 50 eV and beam size < 100 μm

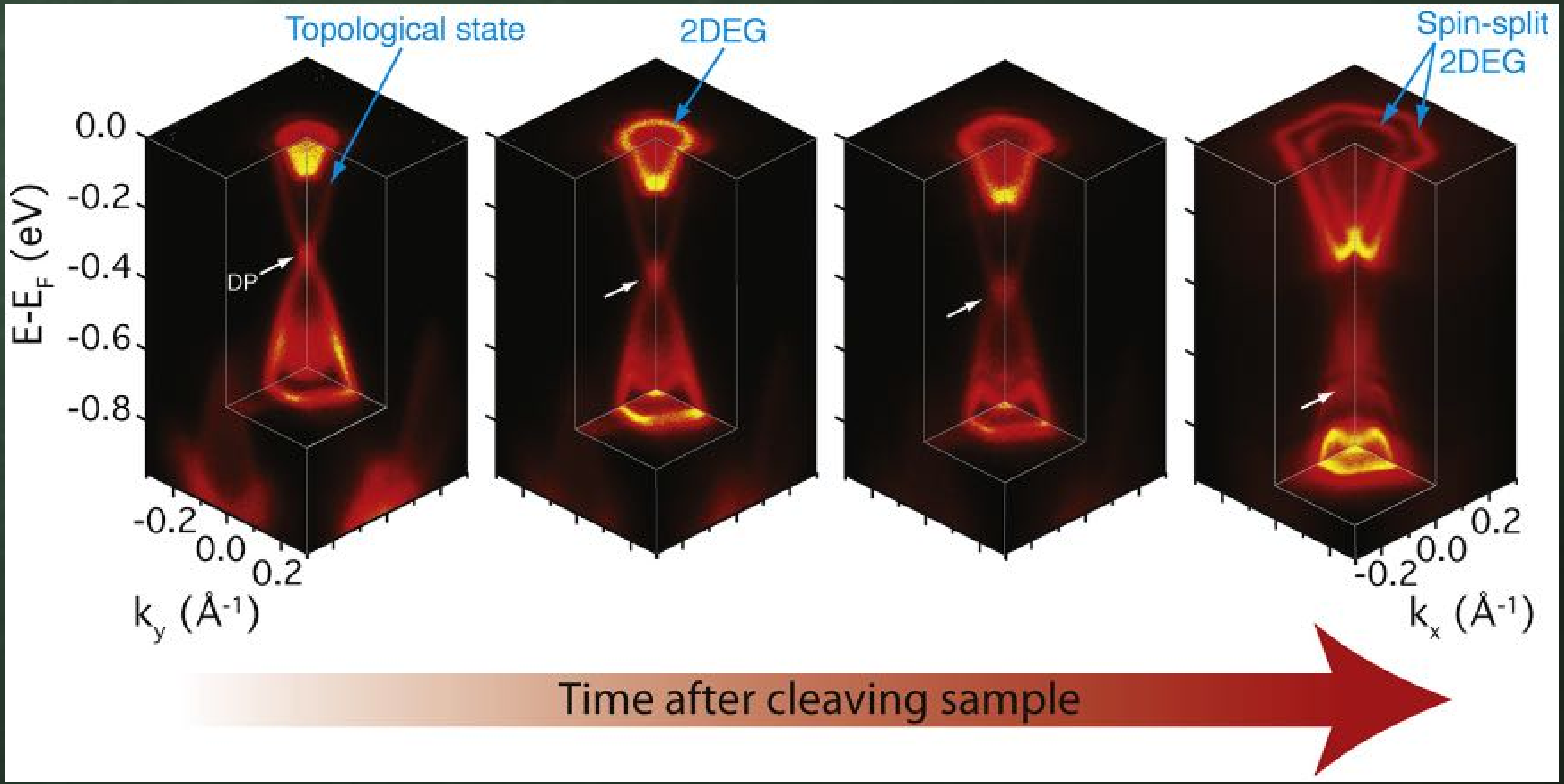
High-Resolution Time-of-Flight Analyzer (ArToF10k)

Ovsyannikov et al., JESRP **191**, 92 (2013)



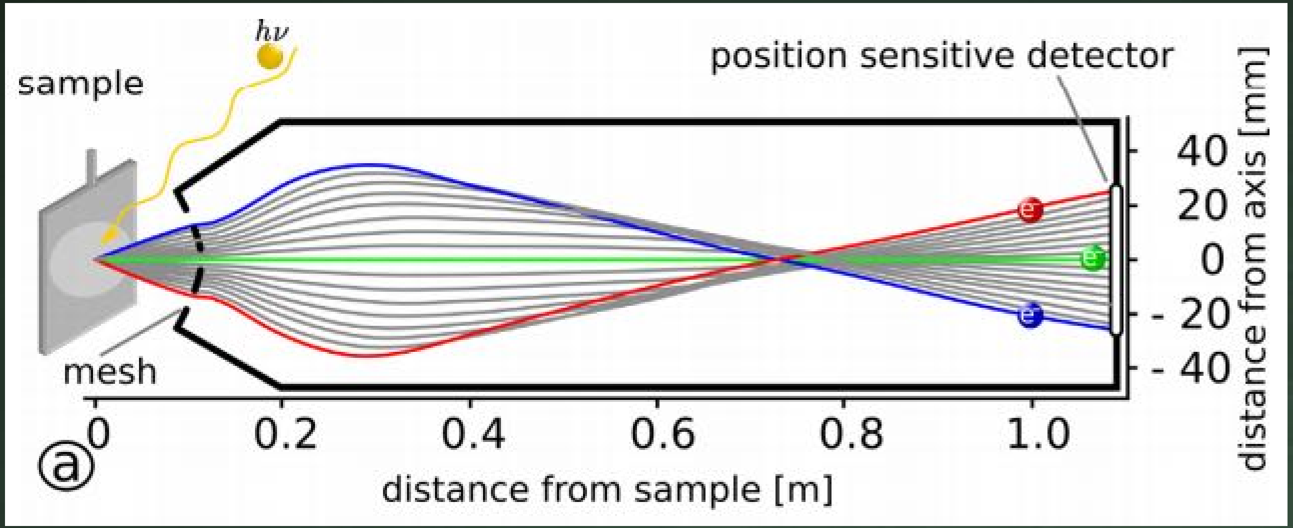


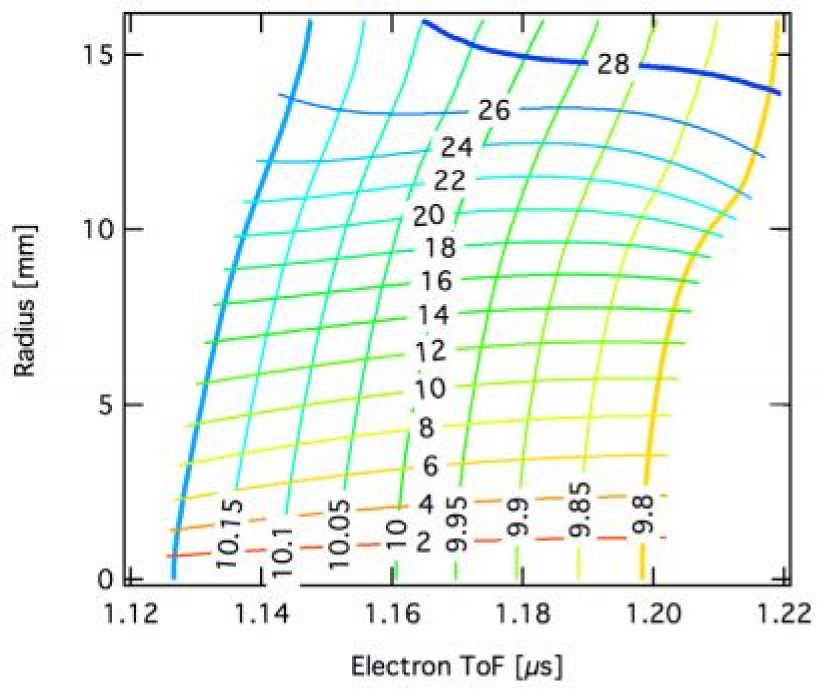
Test with single-bunch mode @ BESSY II

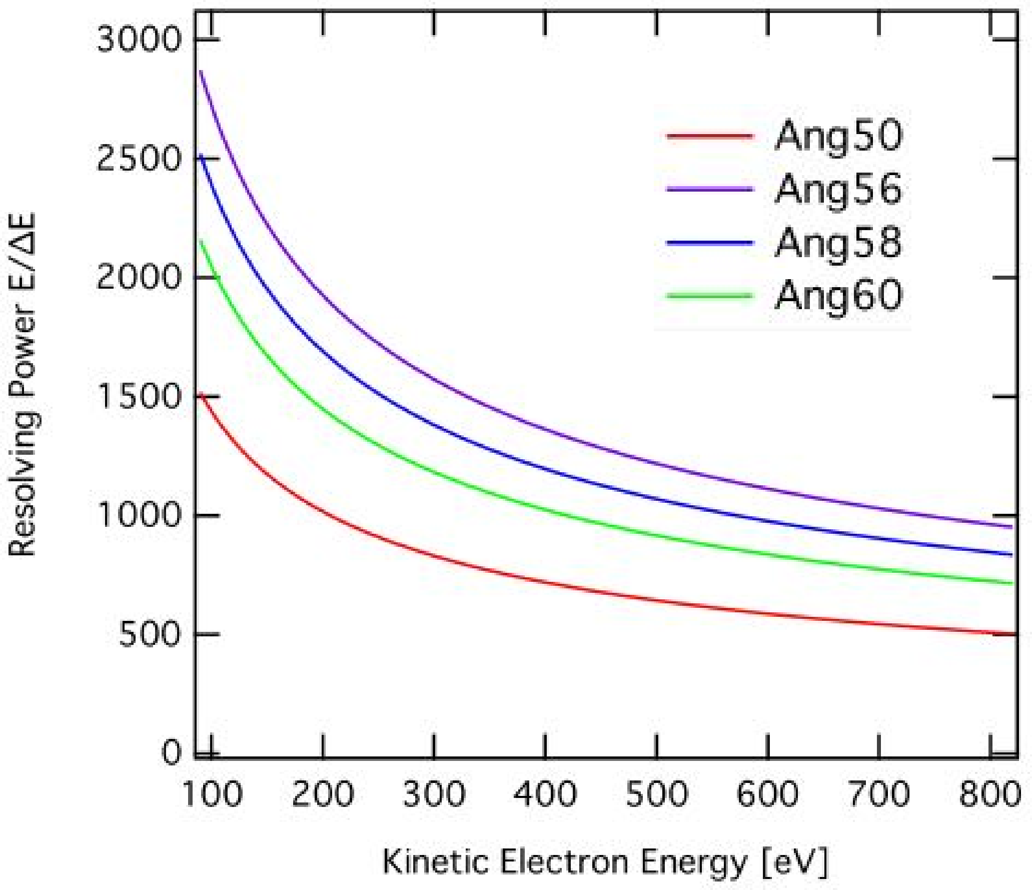


60° ToF @ Soft X-Ray Femto Slicing Beamline of BESSY II

Kühn et al., JESRP **224**, 45 (2018)

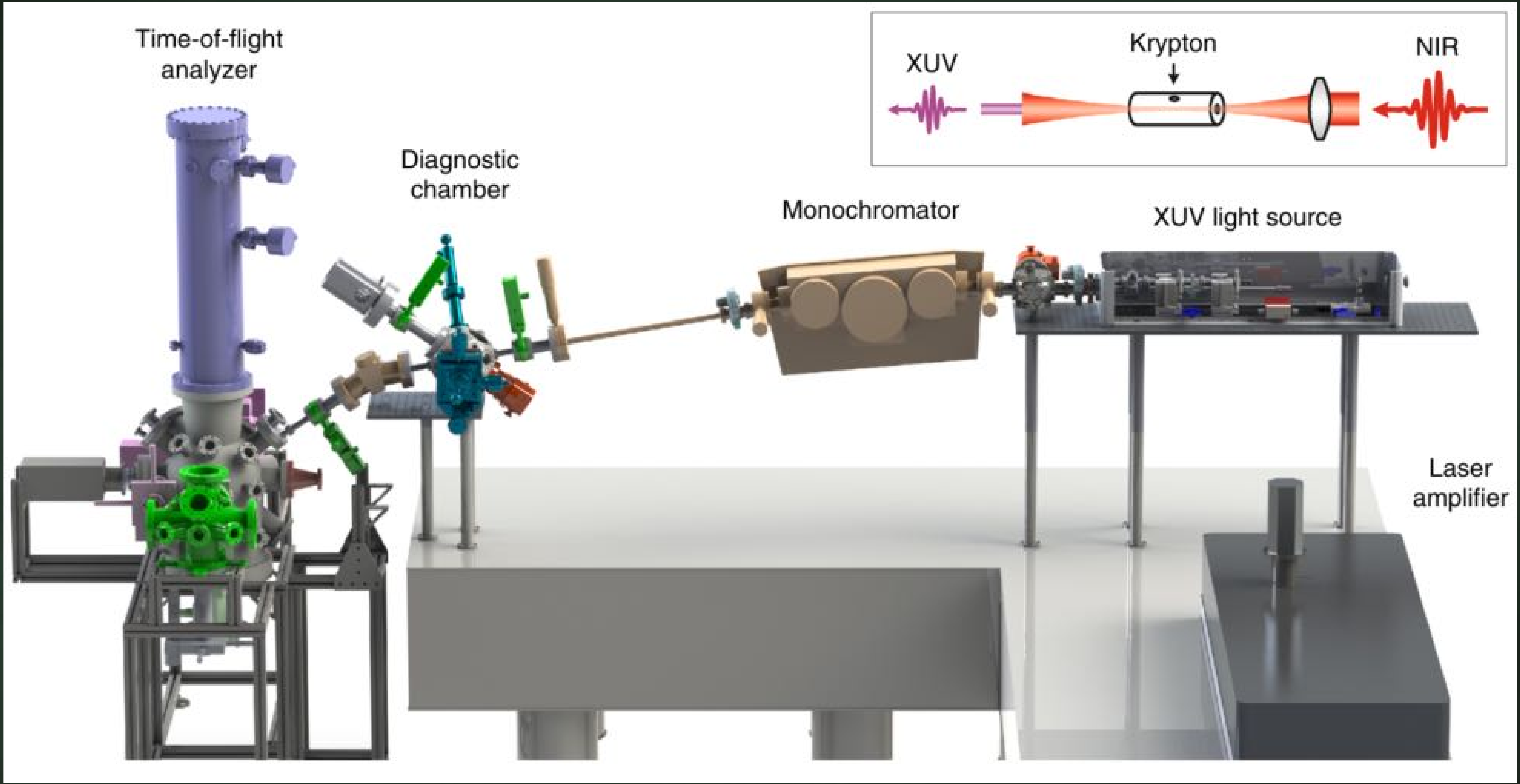






TR-ARPES with ArToF10k Using XUV Pulses @ MIT

Gedik group, Nat. Comm. **10**, 1038 (2019)



TR-ARPES with ArToF10k Using XUV Pulses @ MIT

Gedik group, Nat. Comm. **10**, 1038 (2019)

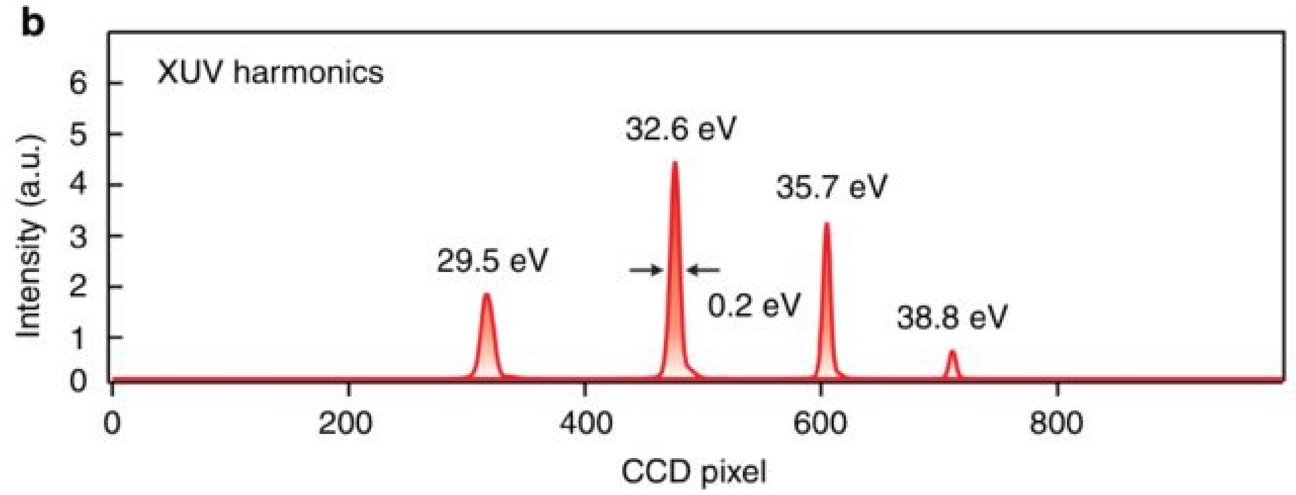
photon energy: 24–33 eV

photon flux: 10^8 – 10^9 photons/sec @ 30 eV

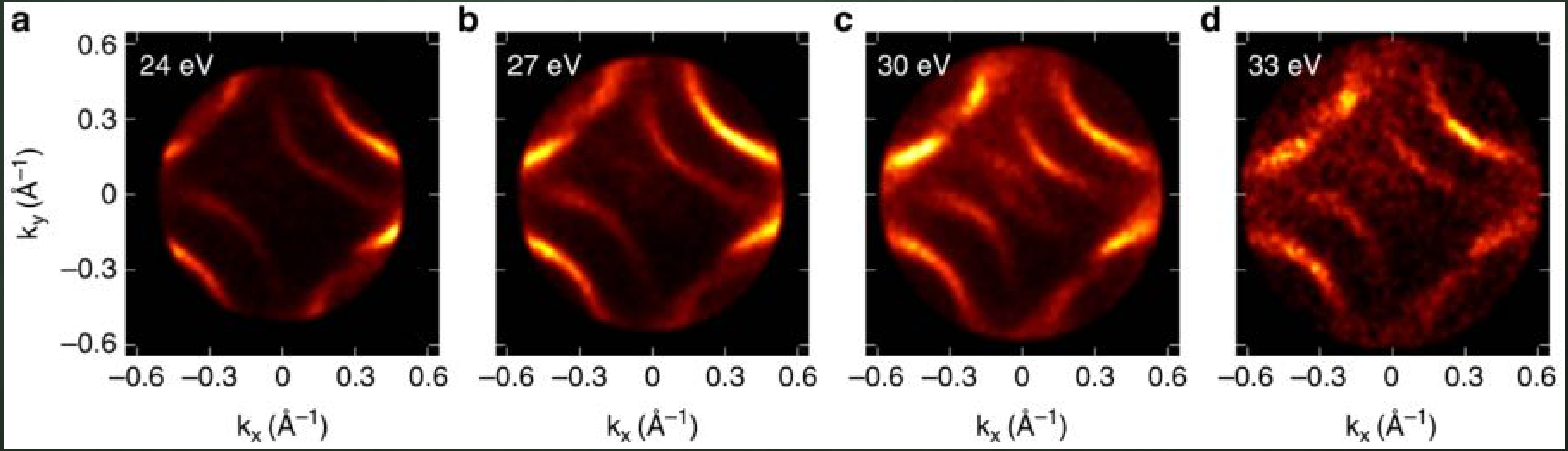
repetition rate: 30 kHz

time resolution: 200 fs

energy resolution: 30 meV @ 33 eV



$Bi_2Sr_2CaCu_2O_{8+\delta}$

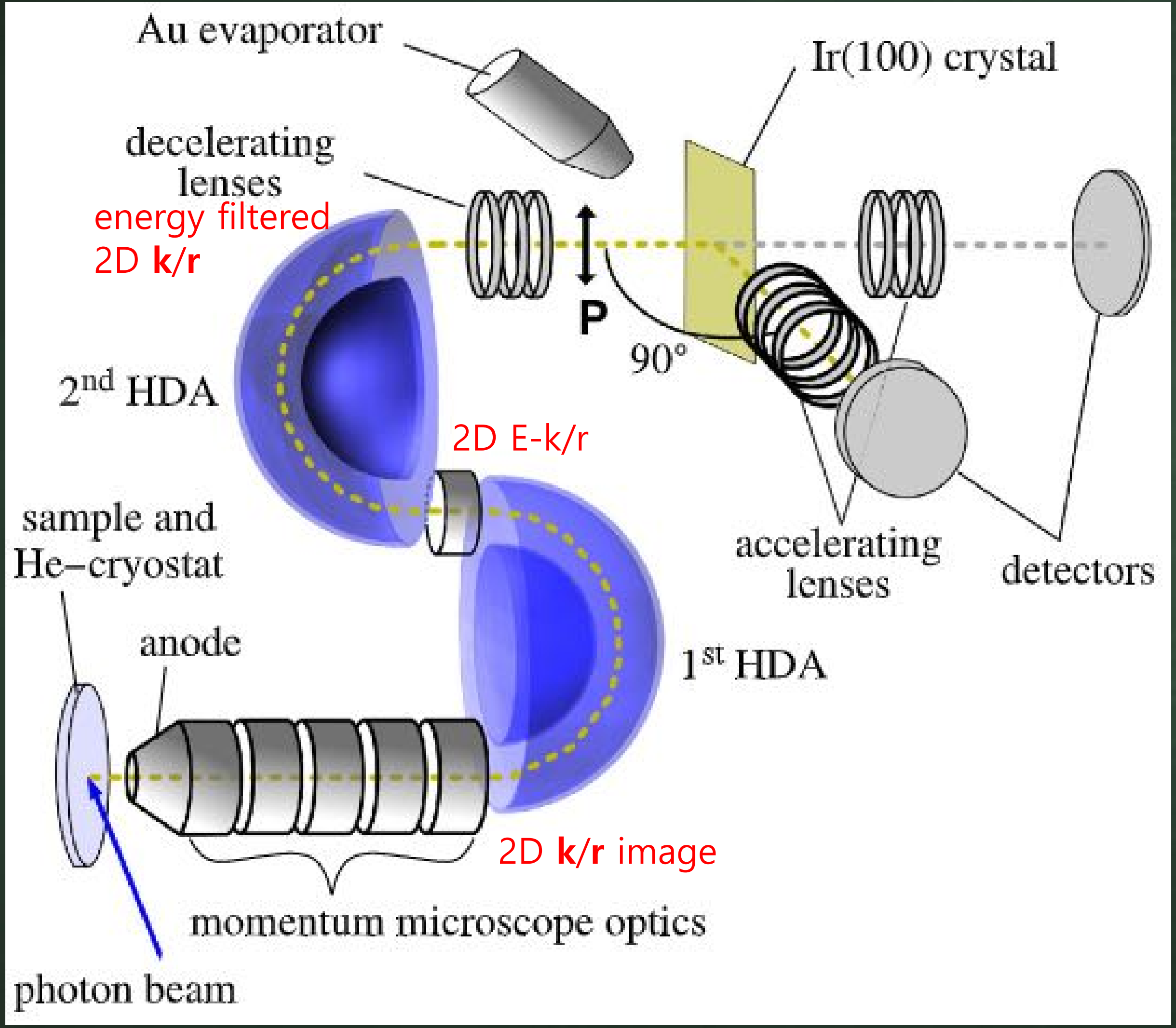


SP-HR-PEkM @ MPI Halle

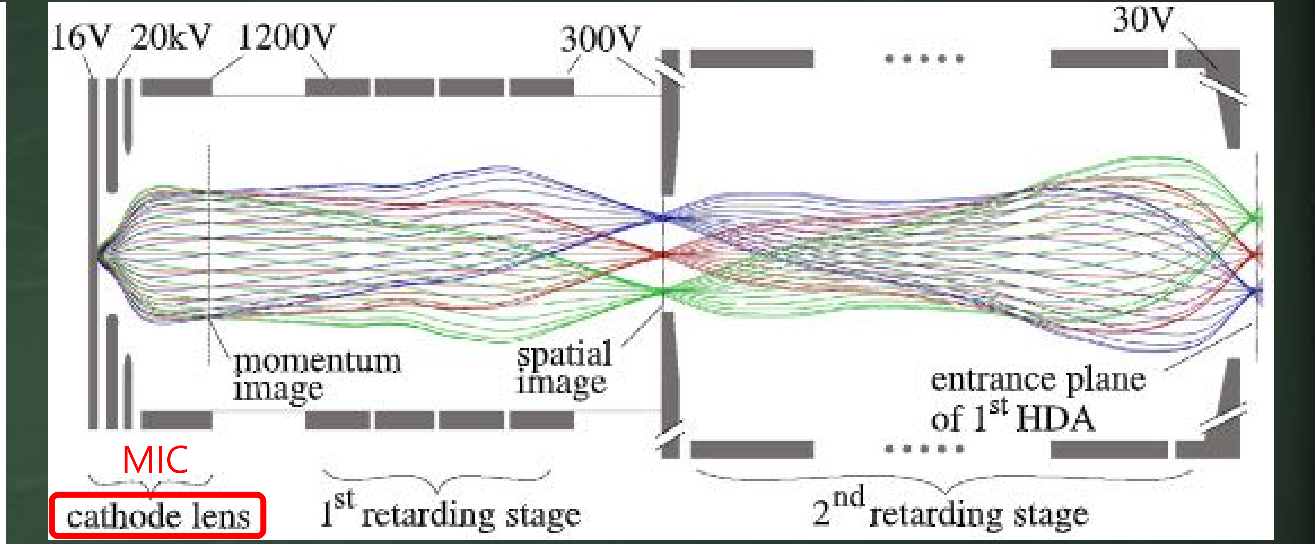
Tusche, Krasyuk, Kirschner, Ultramicroscopy **159**, 520 (2015)

1st design: Omicron+Focus & MPI Halle, Krömker et al., RSI **79**, 053702 (2008)

SP-HR-PEkM system



k-microscope optics



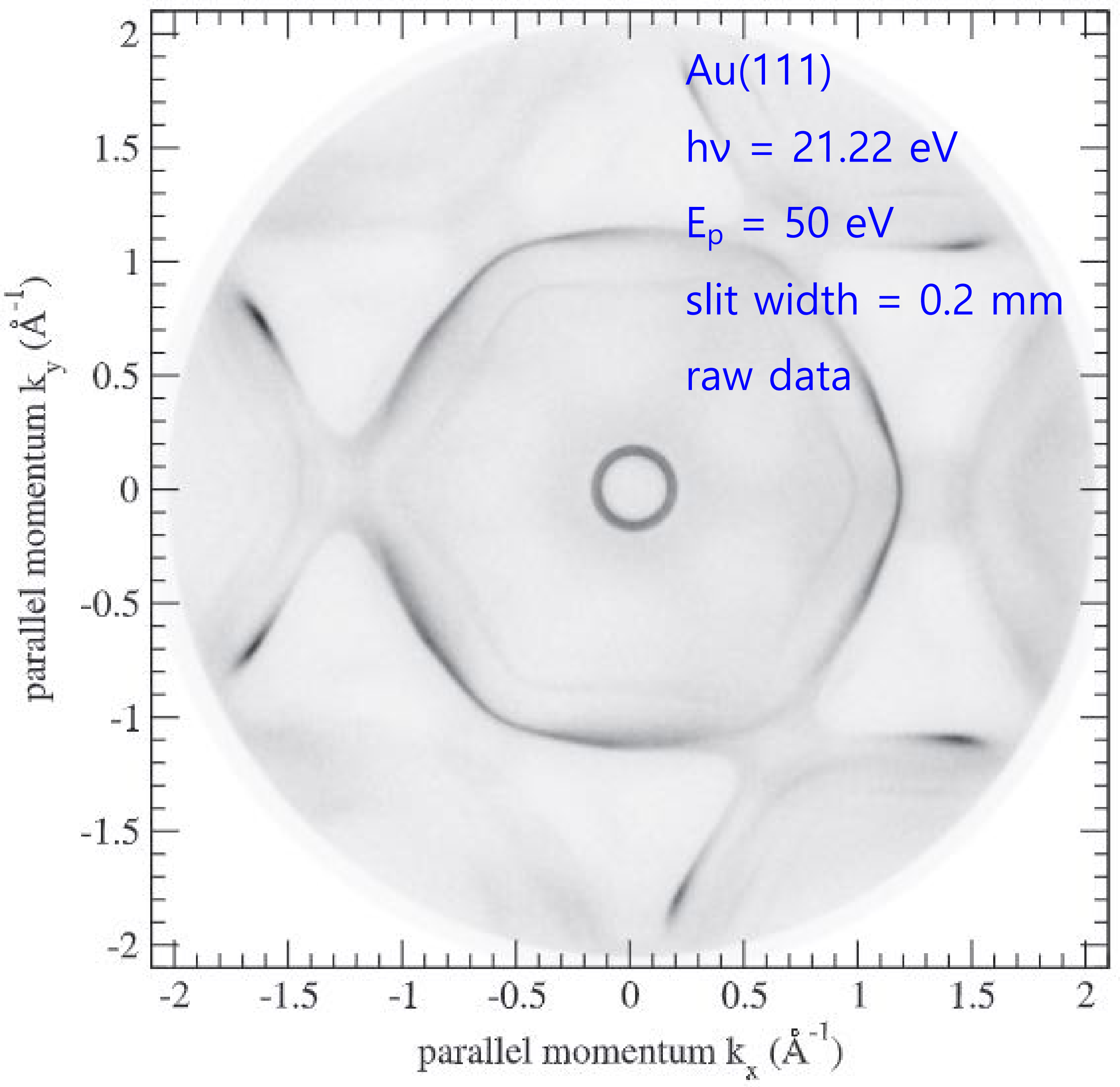
To take whole photoelectrons, apply high voltage
cf. ±15°: 3.4%, ±30°: 13.4%

simulation with beam size 100 μm,
E_{kin} = 16 eV, E_p = 30 eV

energy filter by two HDAs
→ same entrance and exit images
by 1/r potential symmetry

SP-HR-PEkM @ MPI Halle

Tusche, Krasyuk, Kirschner, Ultramicroscopy **159**, 520 (2015)



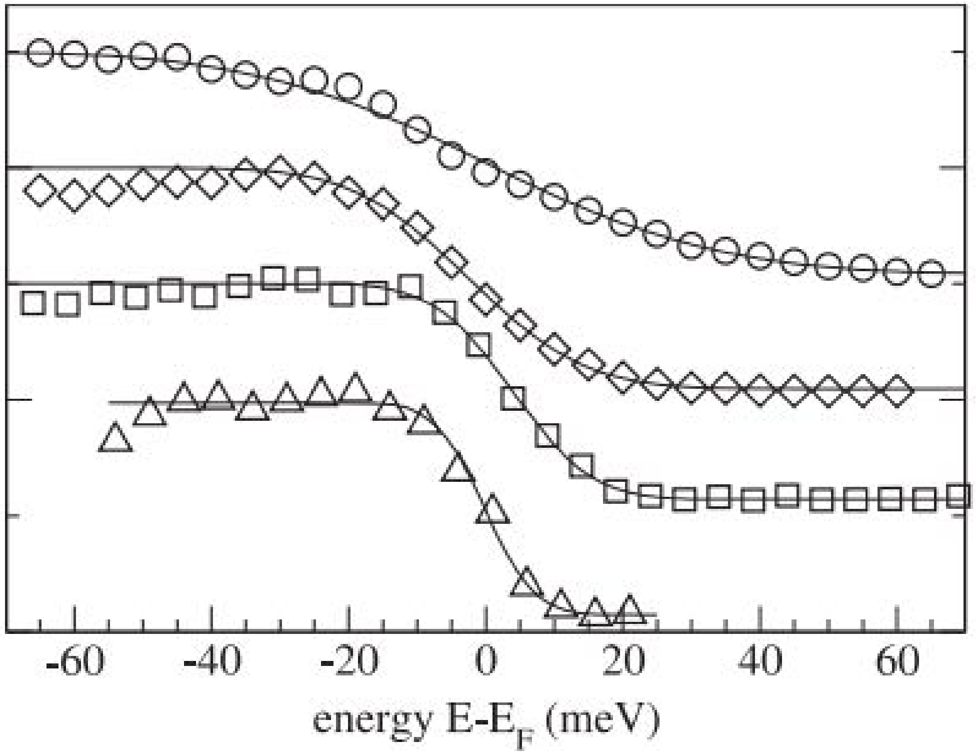
Au(111)

hν = 21.22 eV

E_p = 50 eV

slit width = 0.2 mm

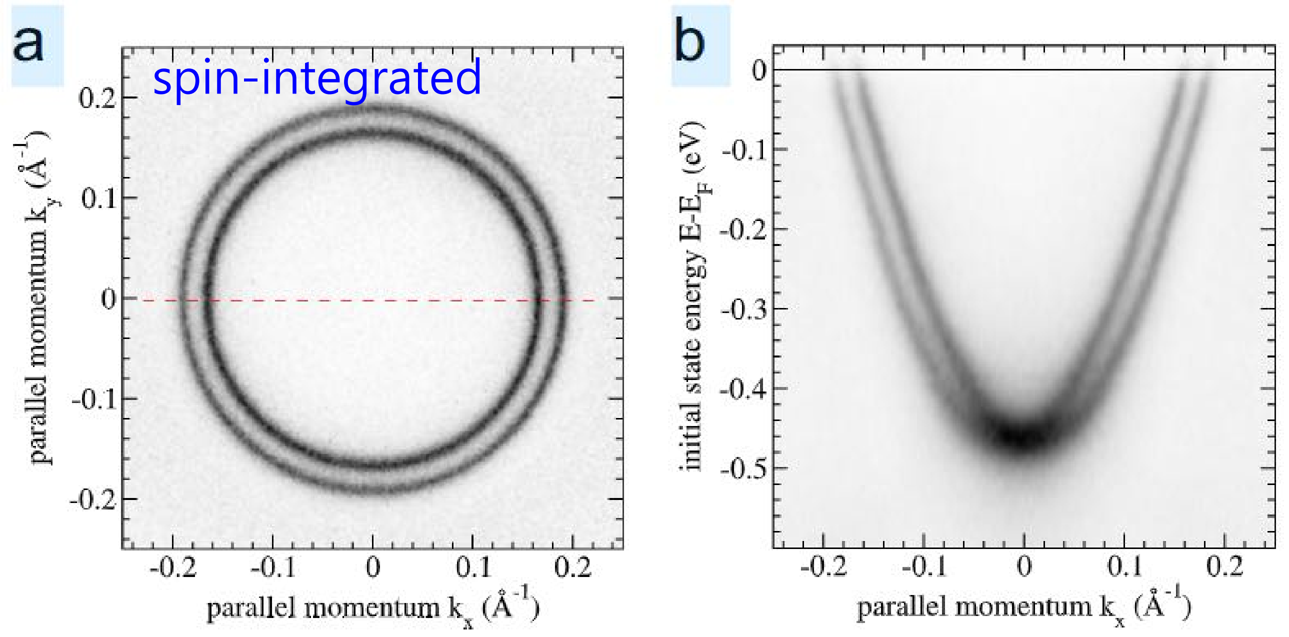
raw data

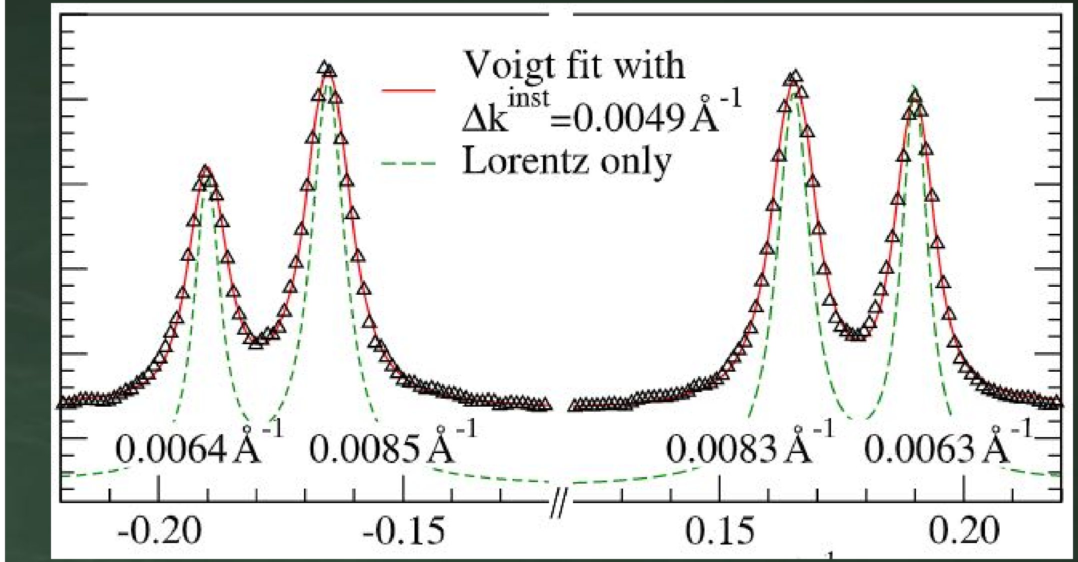


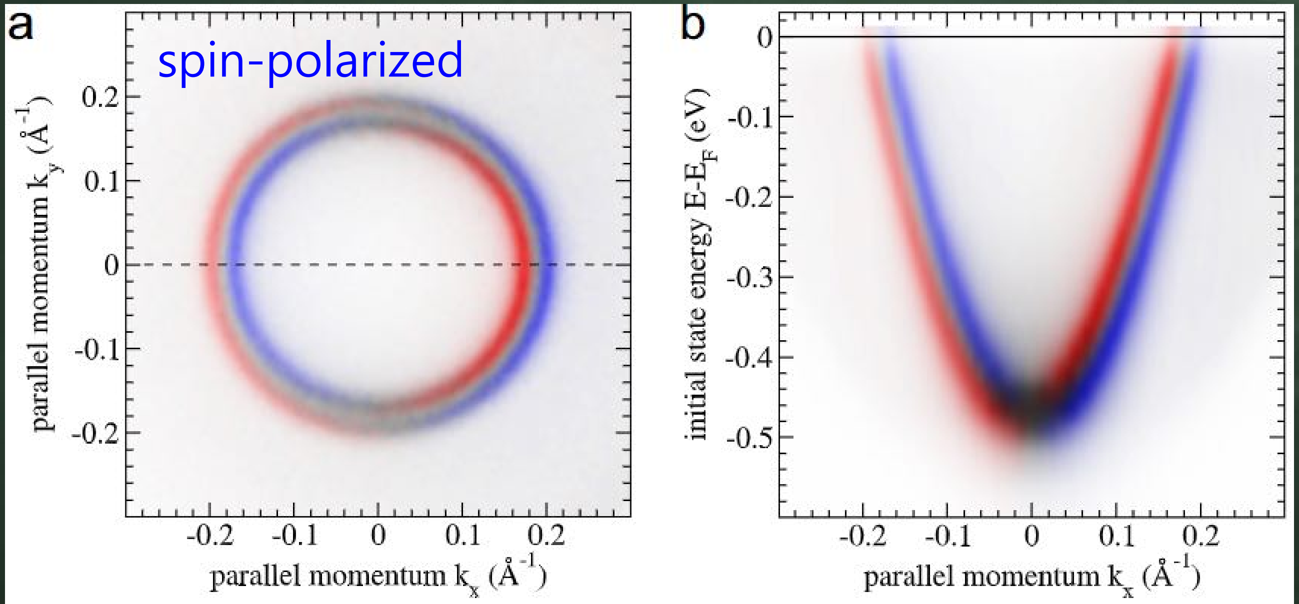
E_{Pass}	ΔE_{ideal} (meV)	ΔE_{exp} (meV)
100	66.67	56.9 ± 4.0
50	33.33	30.2 ± 1.9
30	20.00	20.1 ± 3.1
15	10.00	11.9 ± 1.8

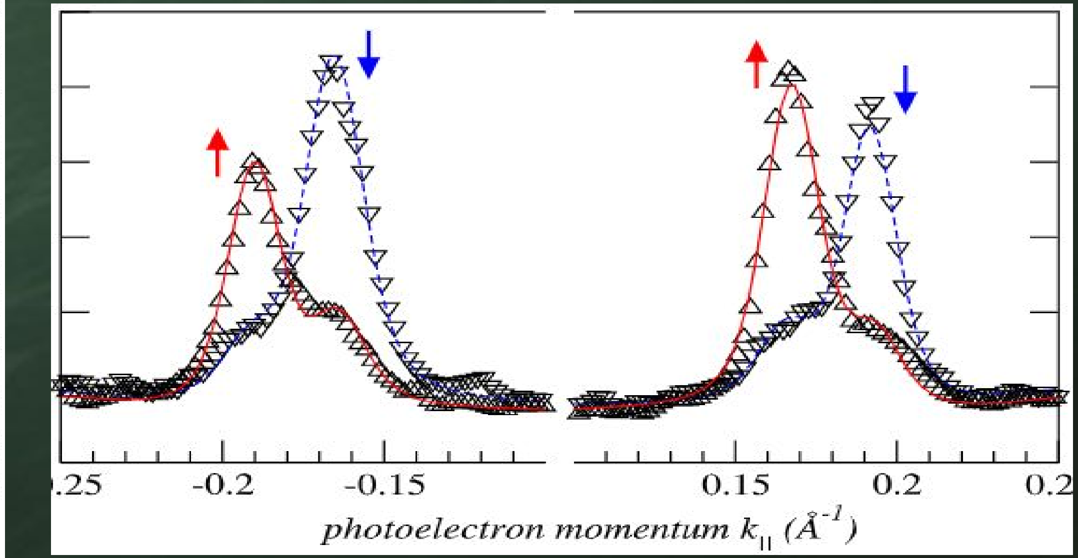
SP-HR-PEkM @ MPI Halle

Tusche, Krasyuk, Kirschner, Ultramicroscopy **159**, 520 (2015)



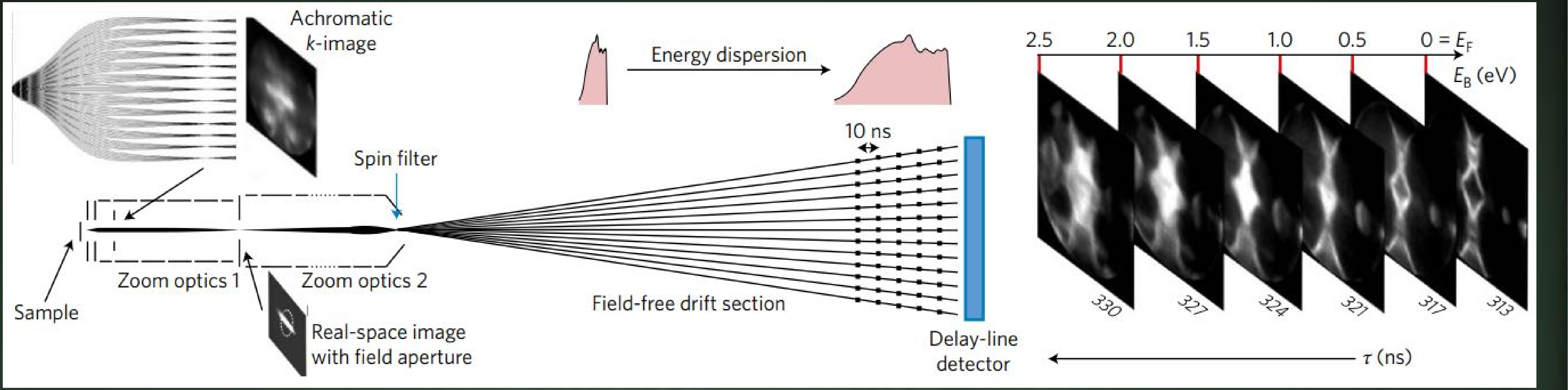






ToF-PEkM for 3D Band Mapping @ PETRA III, DESY

Medjanik et al., Nat. Mater. **16**, 615 (2017)



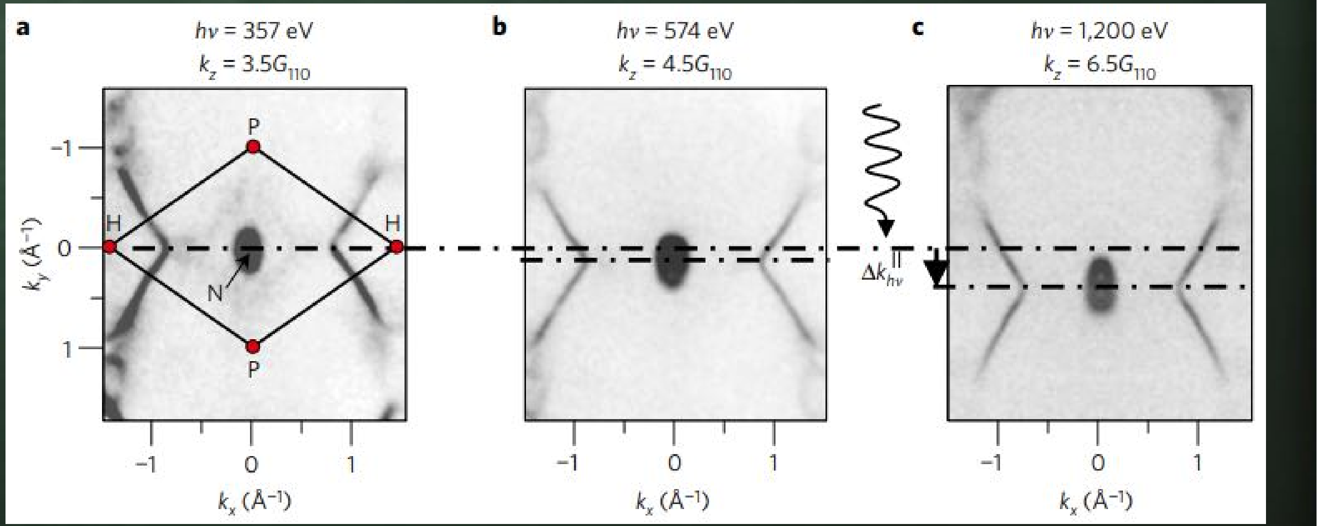
k-resolution: 0.01 Å^{-1}

energy resolution: ~55 meV @ 350 – 1200 eV

spatial resolution: 50 nm

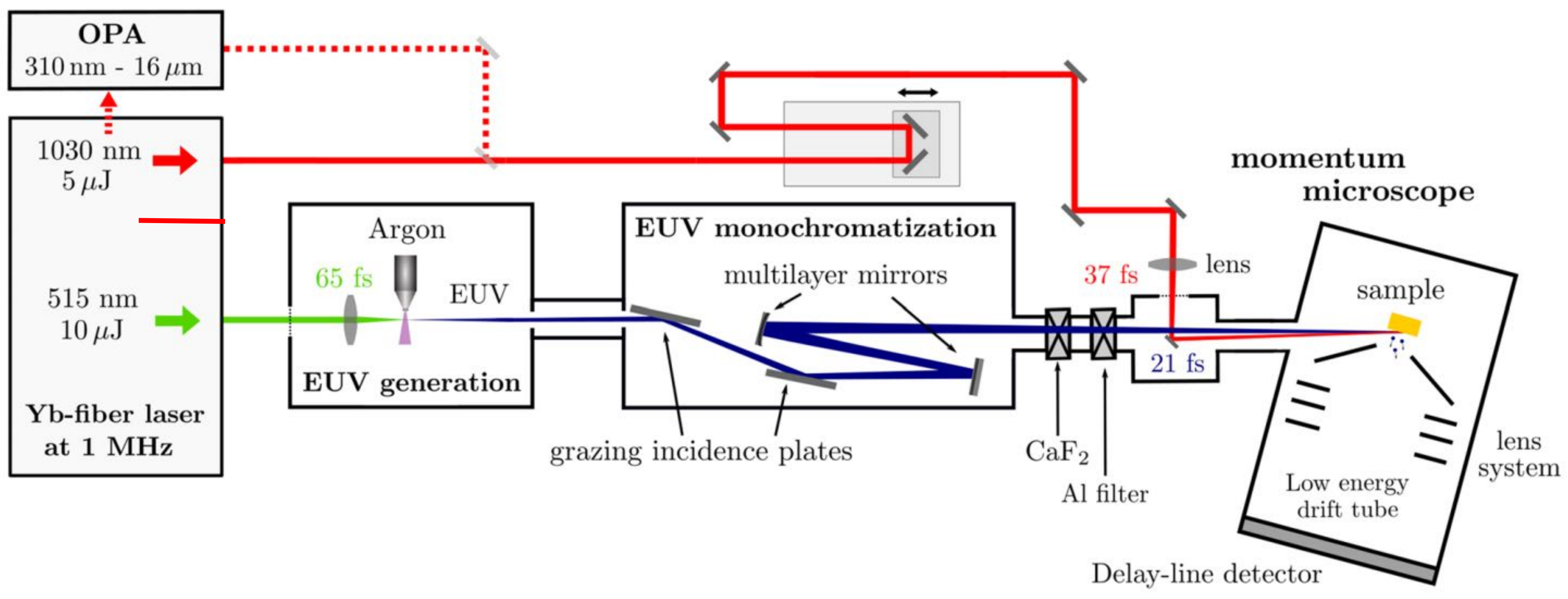
3D band structure of W(110) taken within 3 h!

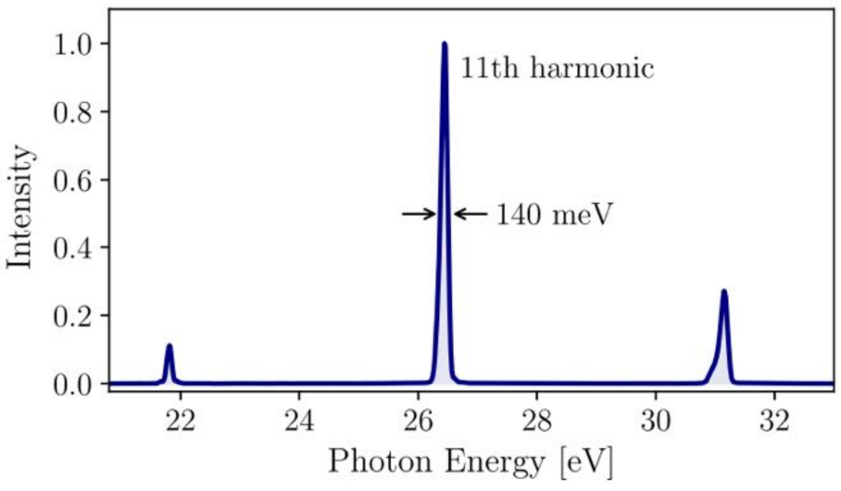
Now available at SPECS GmbH with a spin filter



TR-ToF-PEkM with 1 MHz-RR Table-Top EUV

Mathias group @ Göttingen, RSI **91**, 063905 (2020)





photon flux: 2.7 x 10^{12}/s

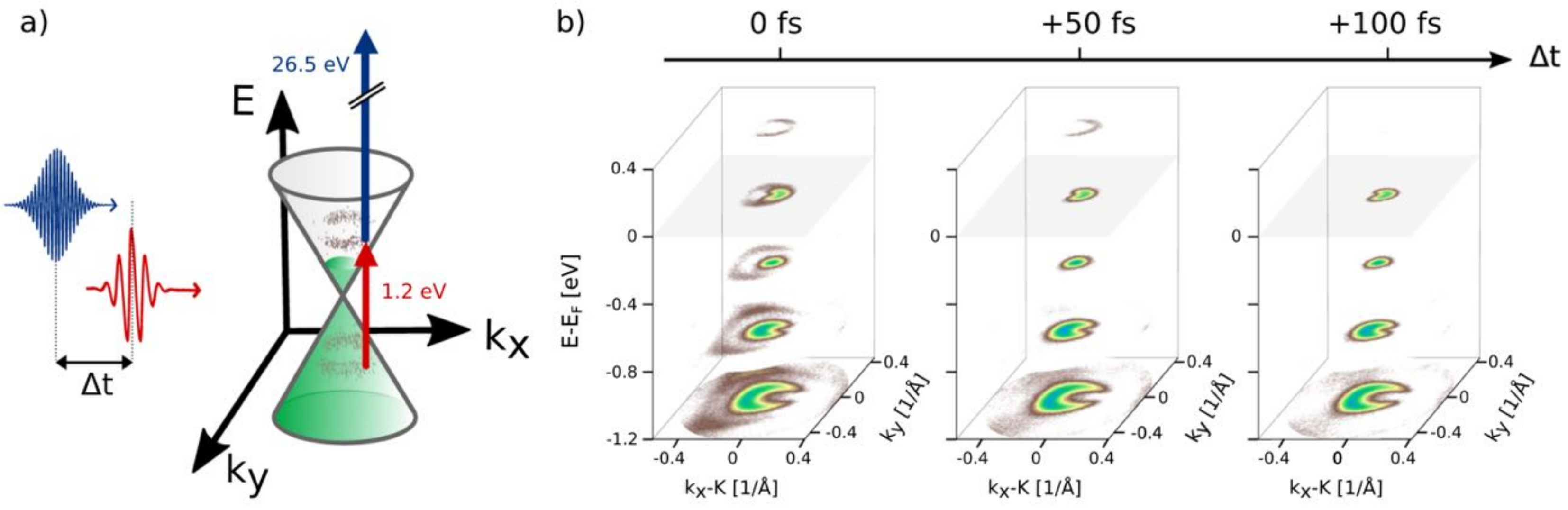
→ 8.5 x 10^{3}/pls (0.3% reduction due to Al filter)

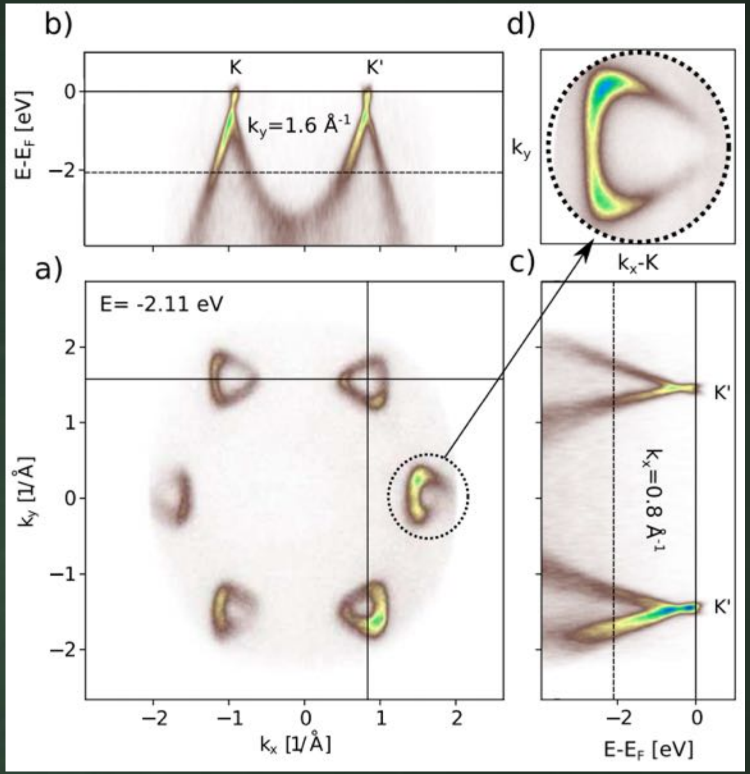
→ > 1 photoelectron/pls

bandwidth: 140 meV

TR-ToF-PEkM with 1 MHz-RR Table-Top EUV

Mathias group @ Göttingen, RSI **91**, 063905 (2020)





Limitations of PEkM

Only one photoelectron detection per laser pulse due to DLD deadtime

(1) multiple DLDs

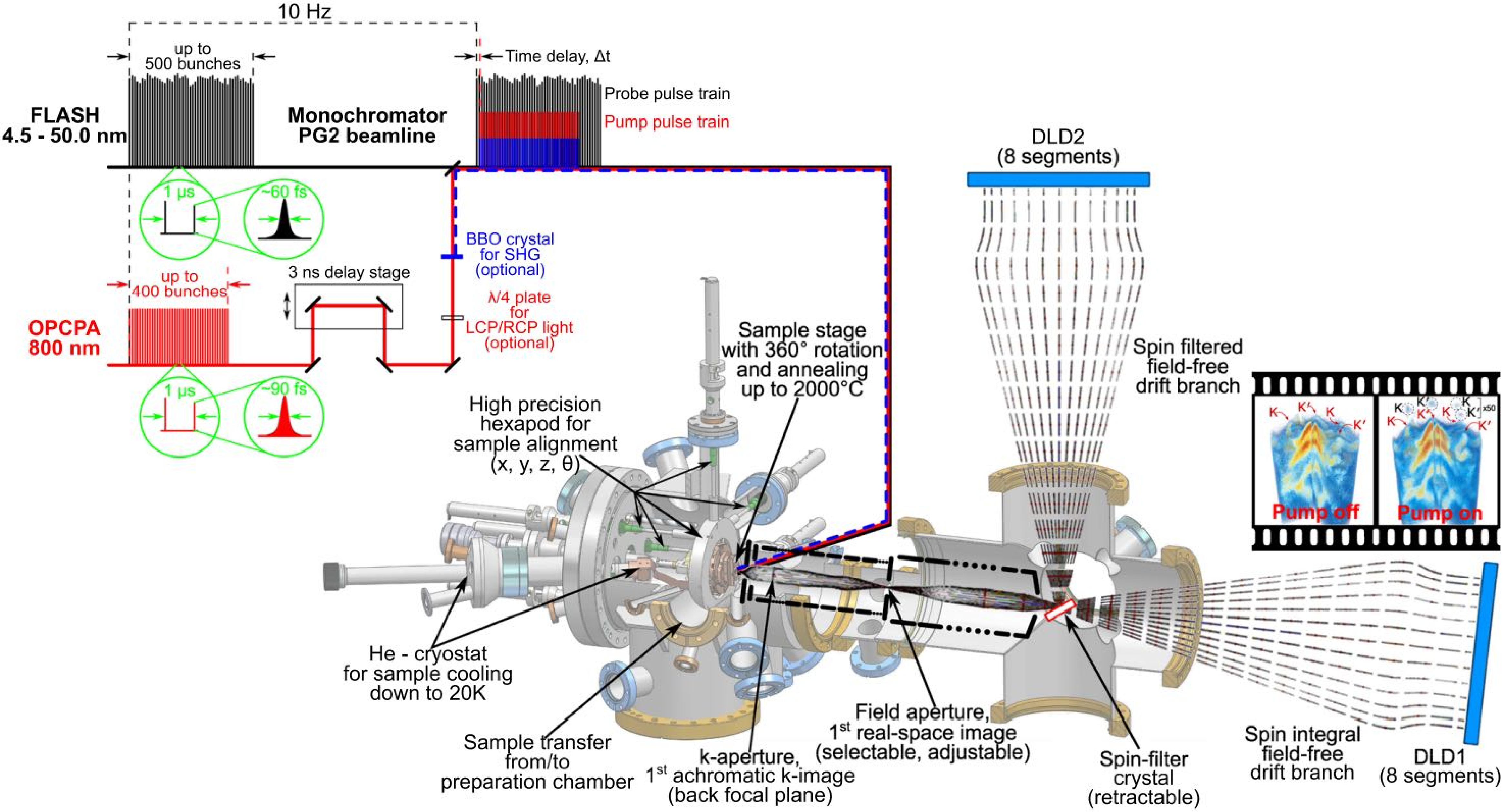
(2) RR increasing

Lifetime of MCP = 5000 h @ 10^6 cps for uniform detection but inhomogeneous degradation

Space-charge effects due to low-energy photoelectrons before entering into the electrostatic lens system

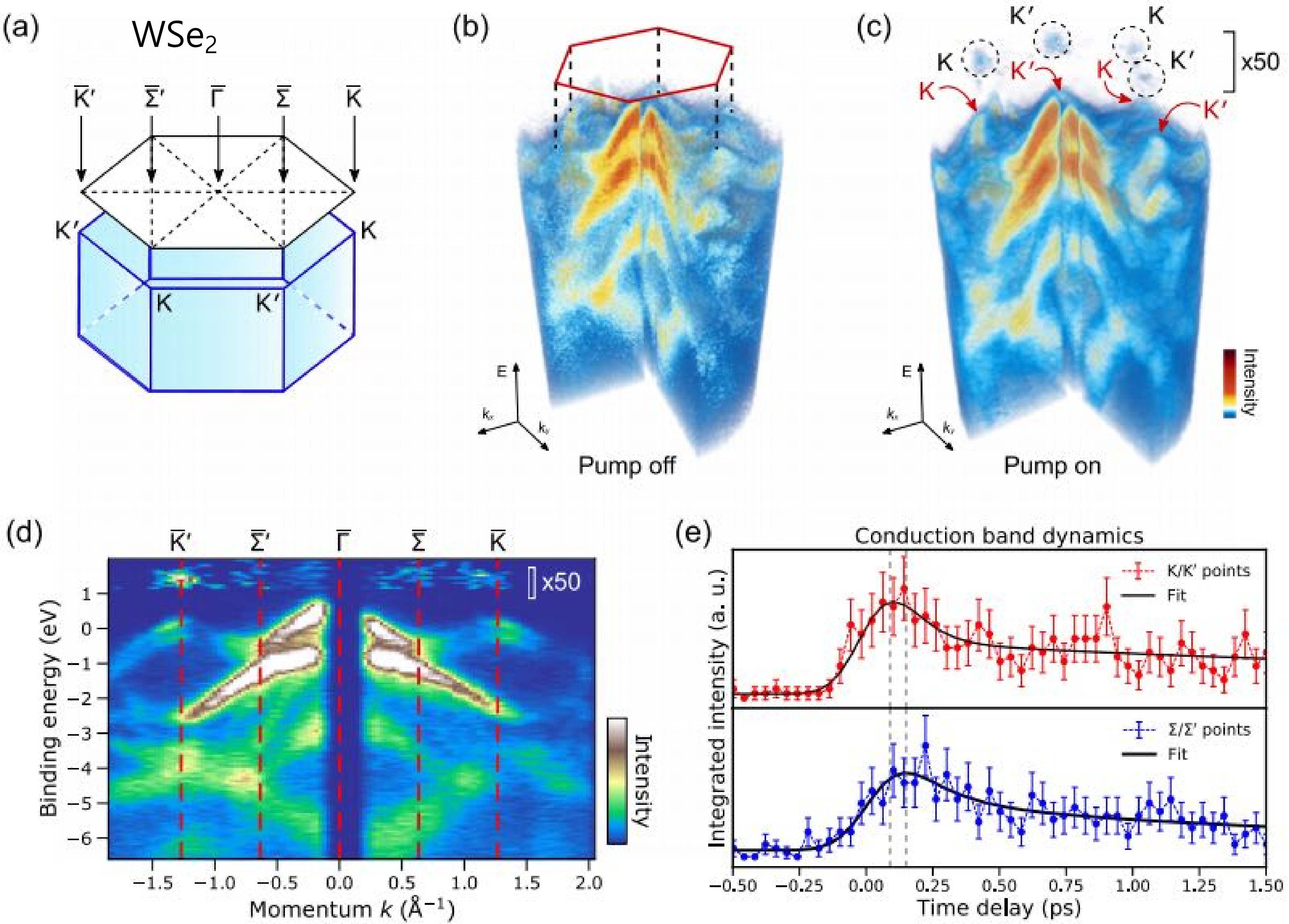
TR-ToF-PEkM @ FLASH/PG2 FEL, DESY

Kutnyakhov et al., RSI **91**, 013109 (2020)



TR-ToF-PEkM @ FLASH/PG2 FEL, DESY

Kutnyakhov et al., RSI **91**, 013109 (2020)



ΔE ~ 130 meV

Δk ~ 0.06 Å^{-1}

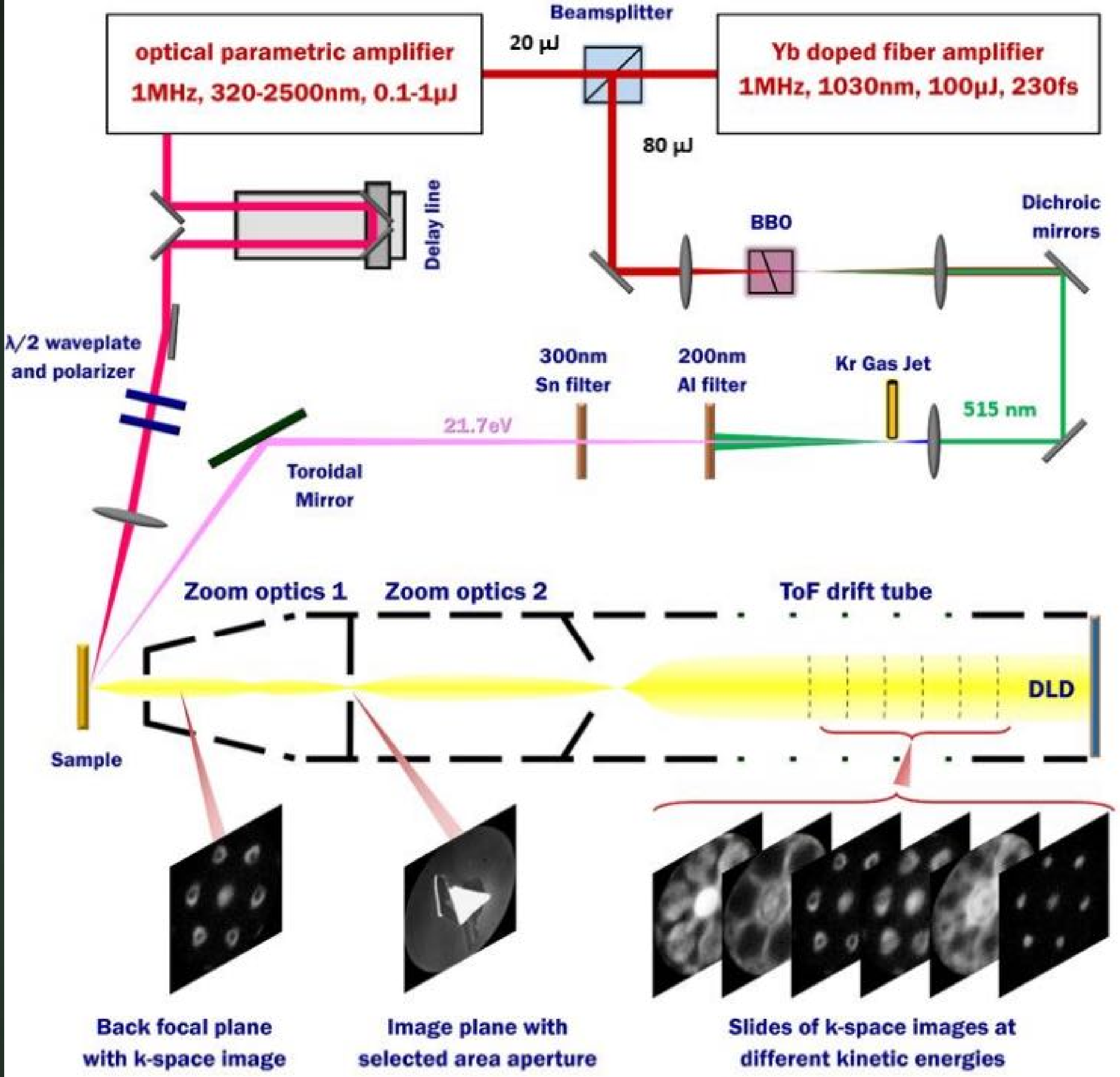
Δt ~ 150 fs

Poorer than homelab

due to space-charge effects

TR-μ-ToF-PEkM @ Okinawa

Dani group @ OIST, Science **370**, 1199 (2020)

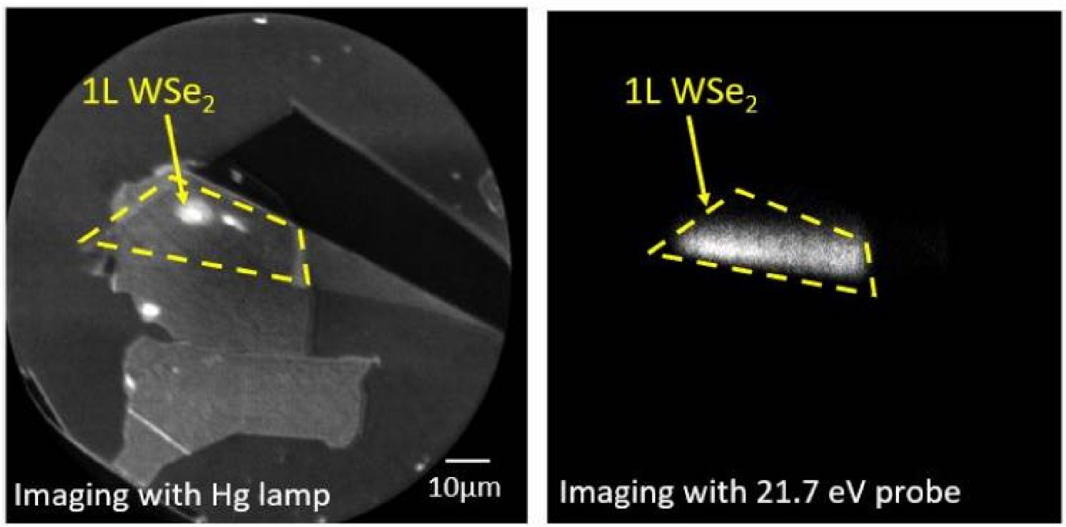


Metis 1000, SPECS GmbH

ΔE = 30 meV

Δk = 0.01 Å^{-1}

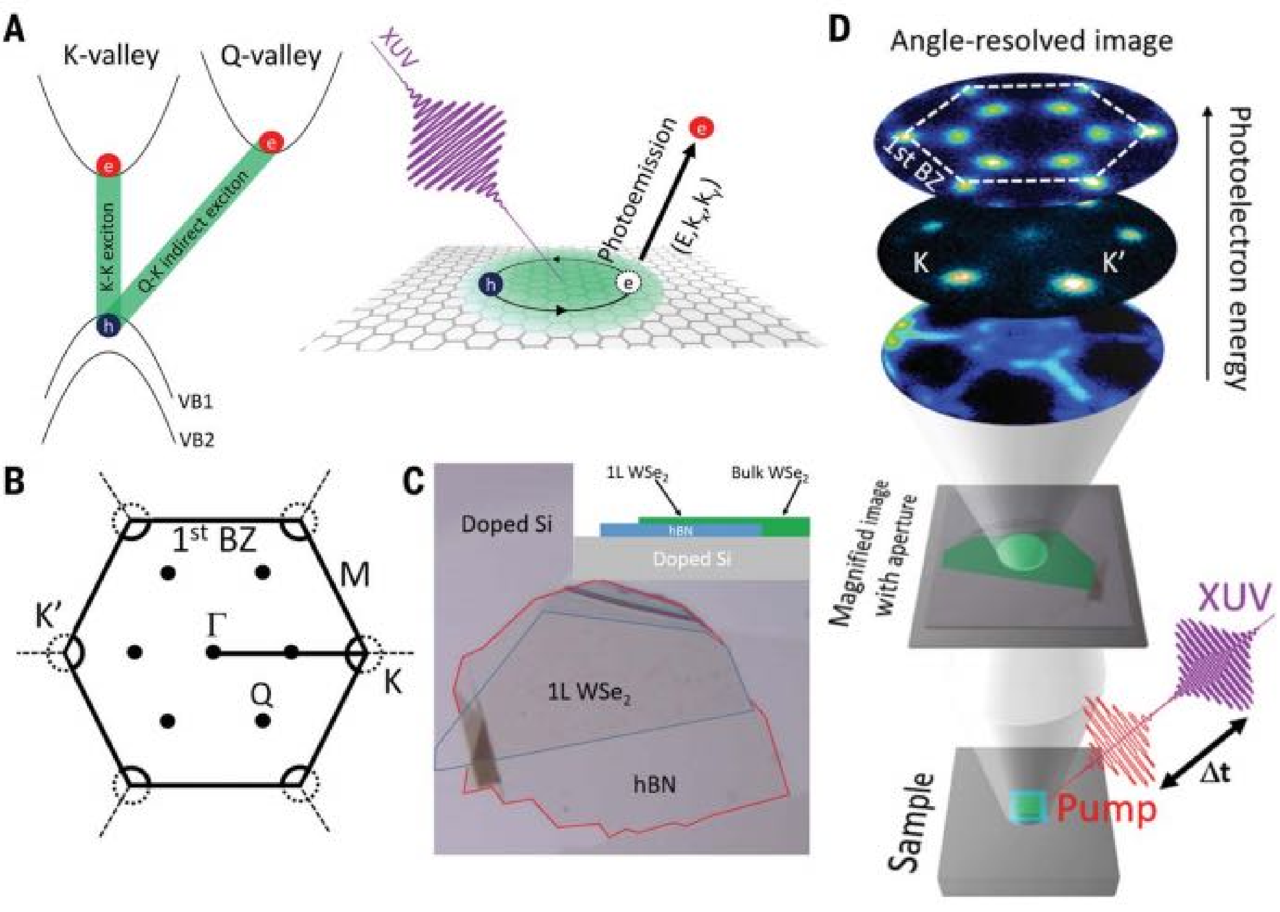
Δt = 165 fs

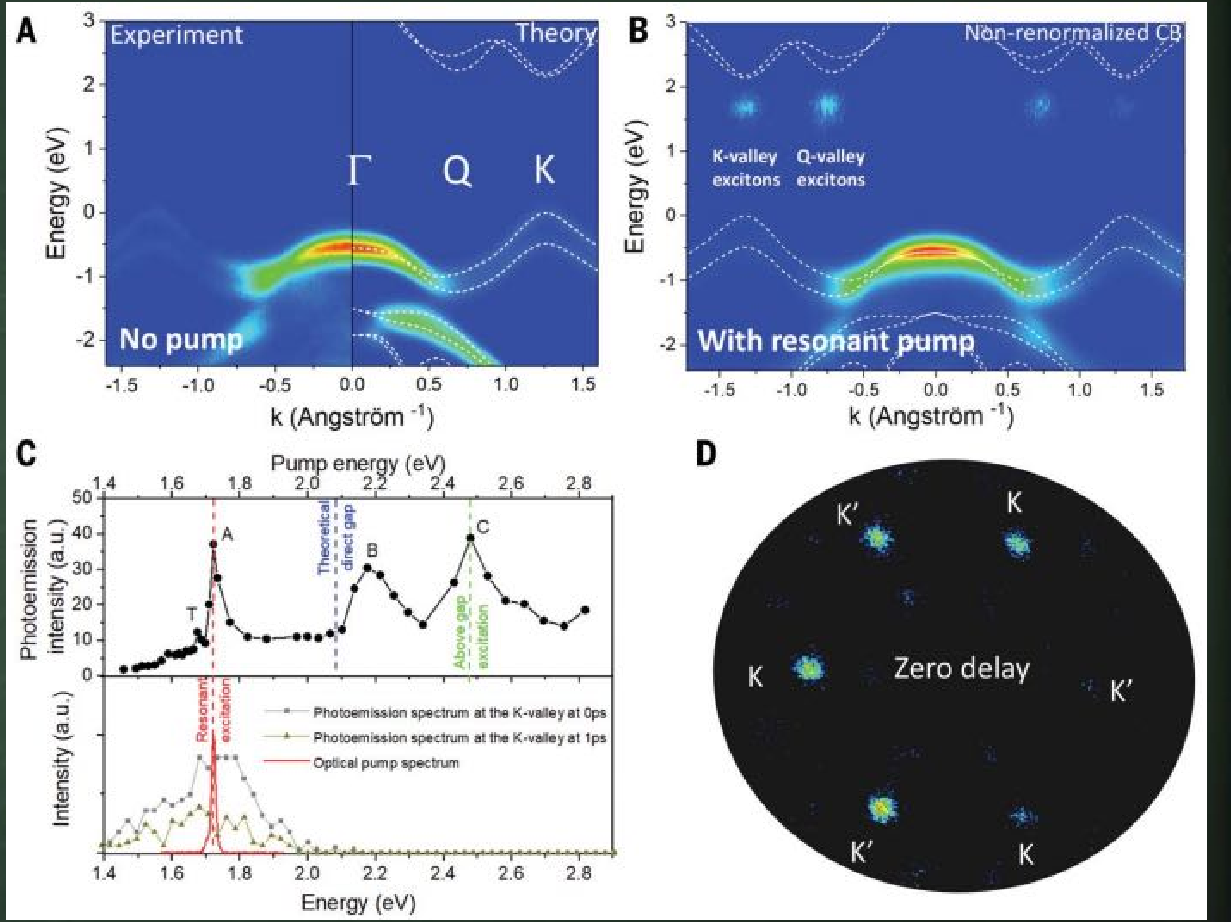


TR-μ-ToF-PEkM @ Okinawa

Dani group @ OIST, Science **370**, 1199 (2020)

Observation of ultrafast population dynamics of K- & Q-valley excitons

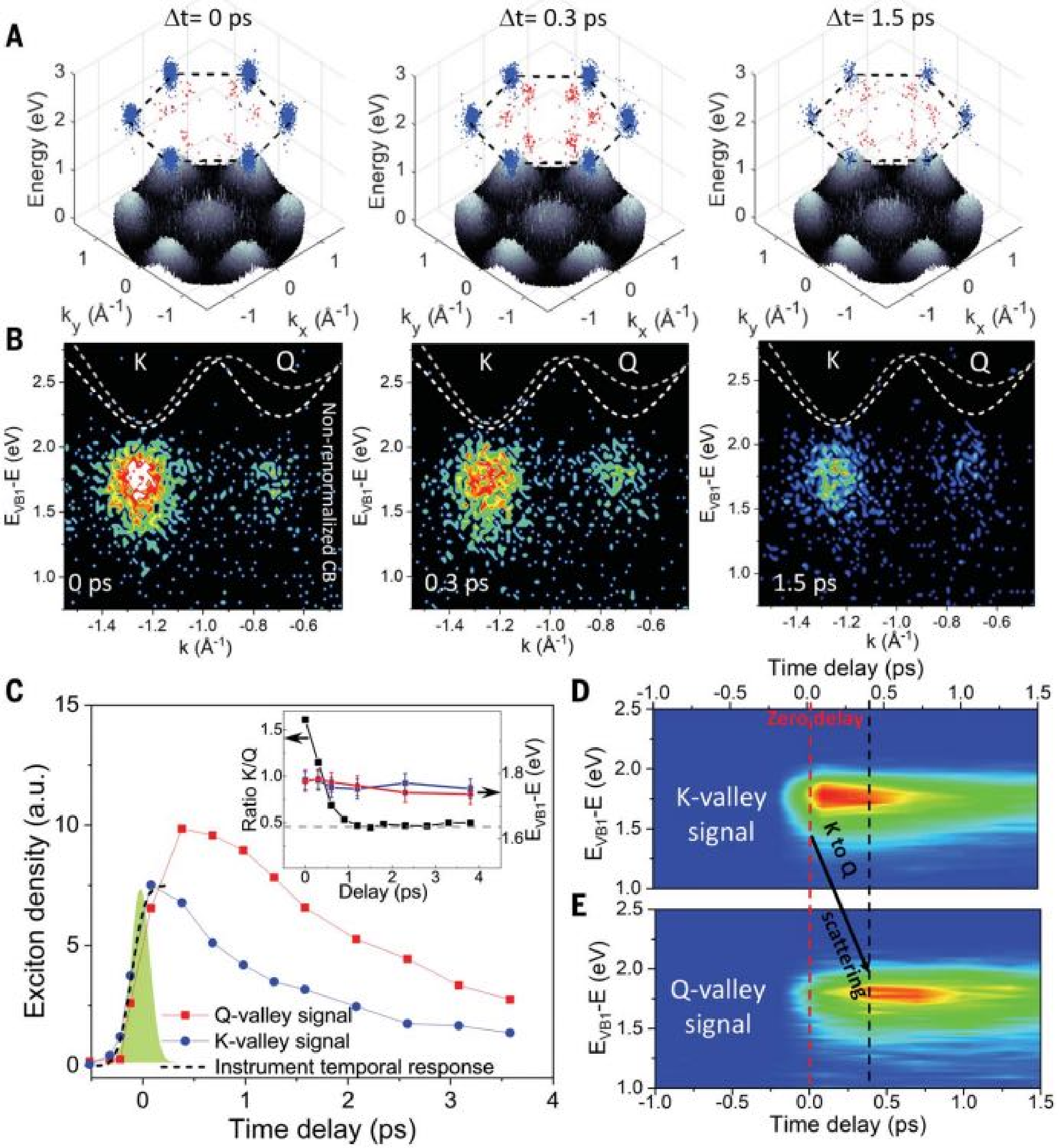




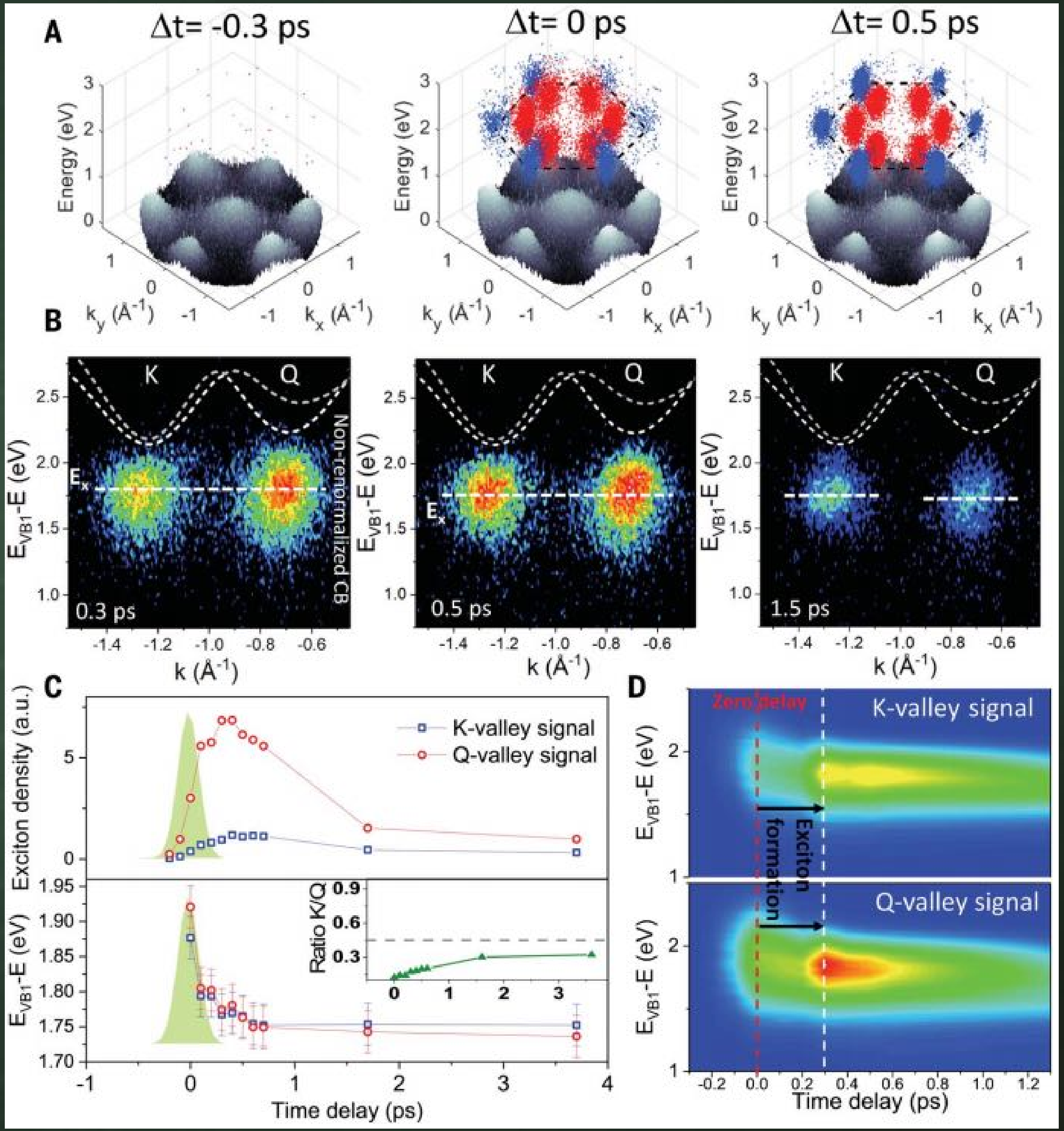
TR-μ-ToF-PEkM @ Okinawa

Dani group @ OIST, Science **370**, 1199 (2020)

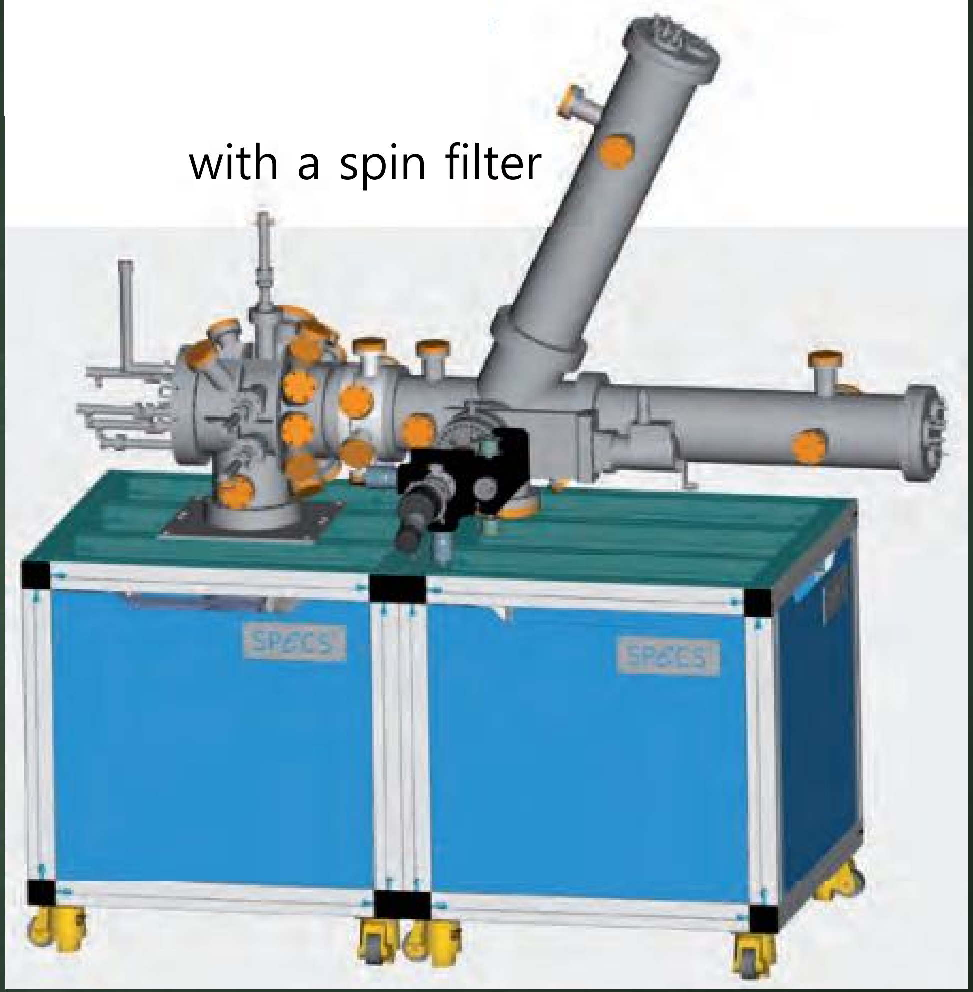
resonant pump hν = 1.72 eV



above-gap pump hν = 2.48 eV



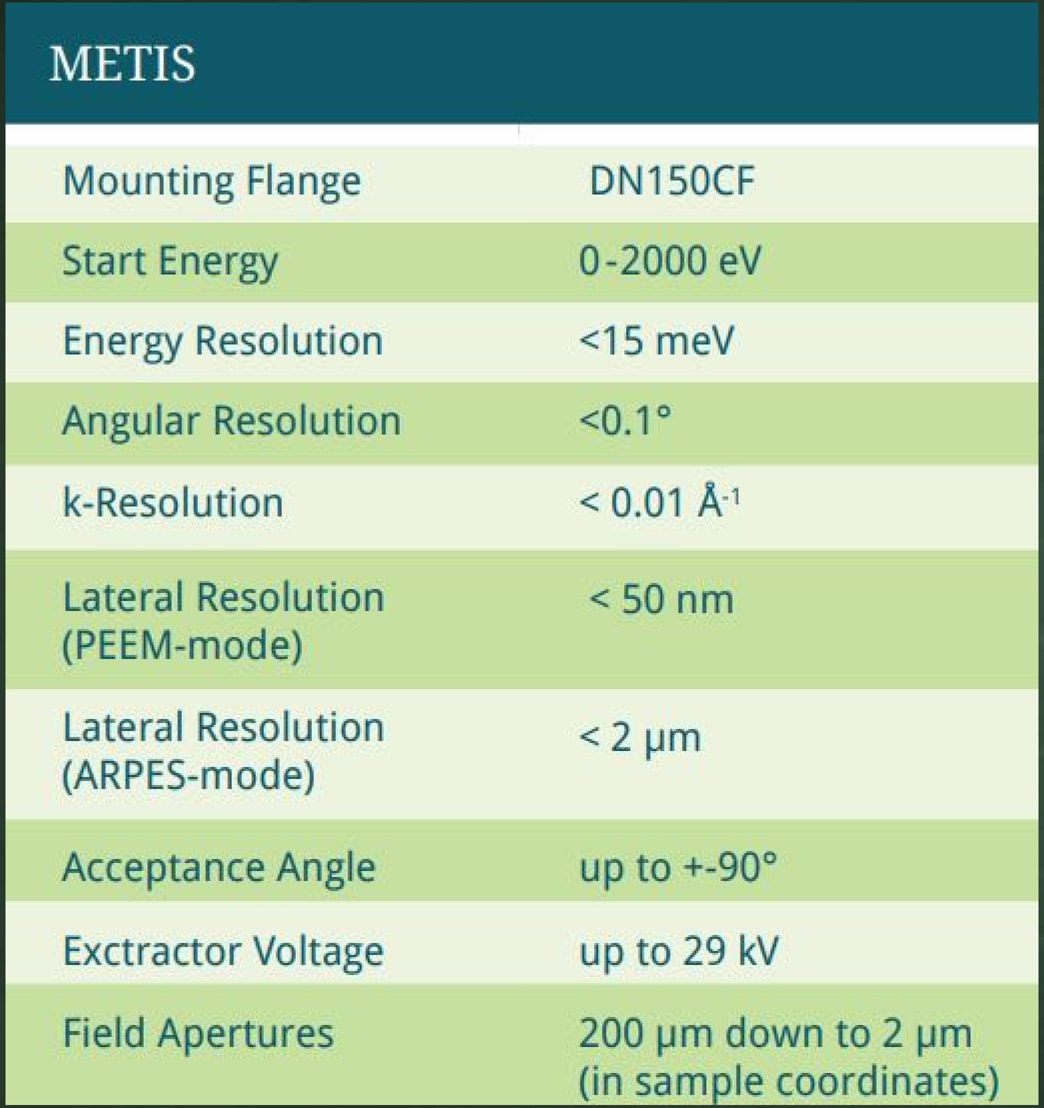
SPECS METIS 1000



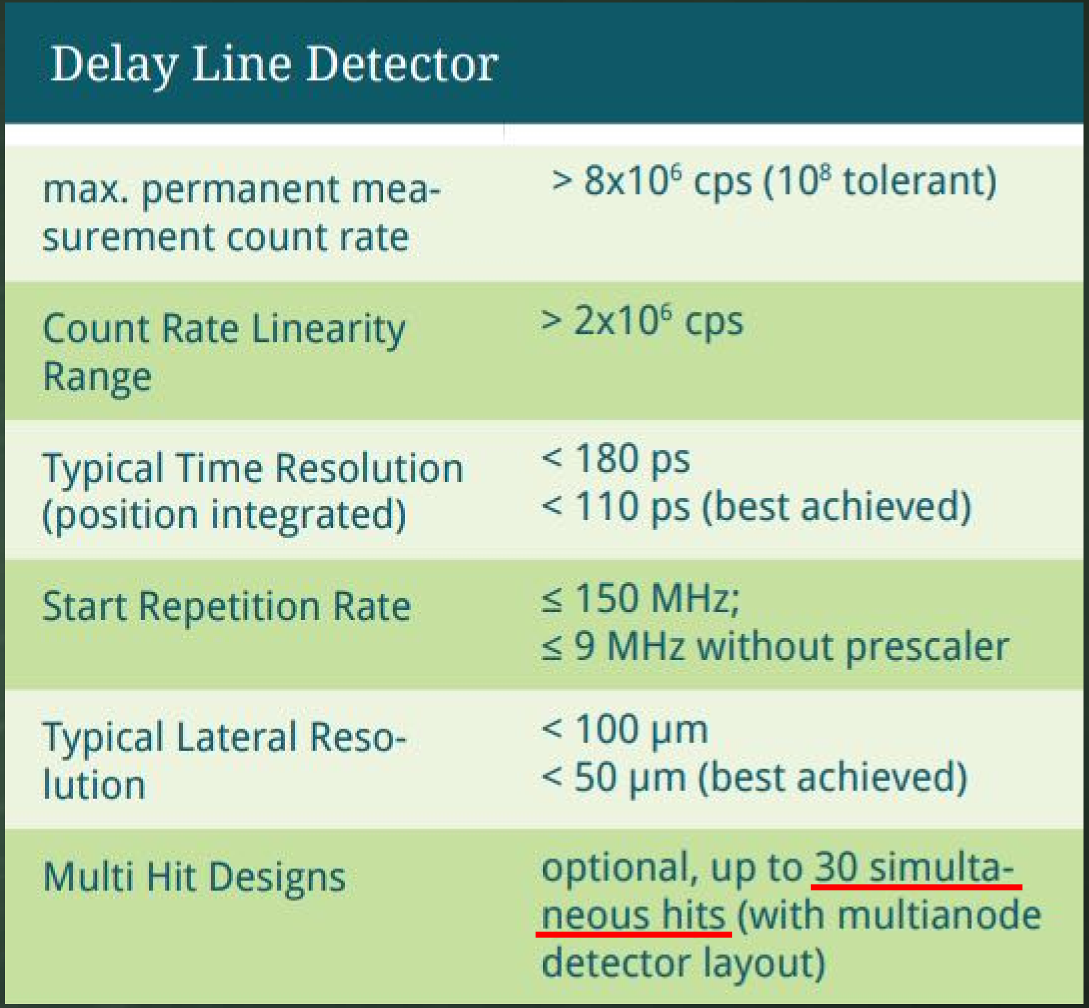
with a spin filter

https://www.specs-group.com/nc/specs/products/detail/metis-1000

SPECS METIS 1000 Specifications

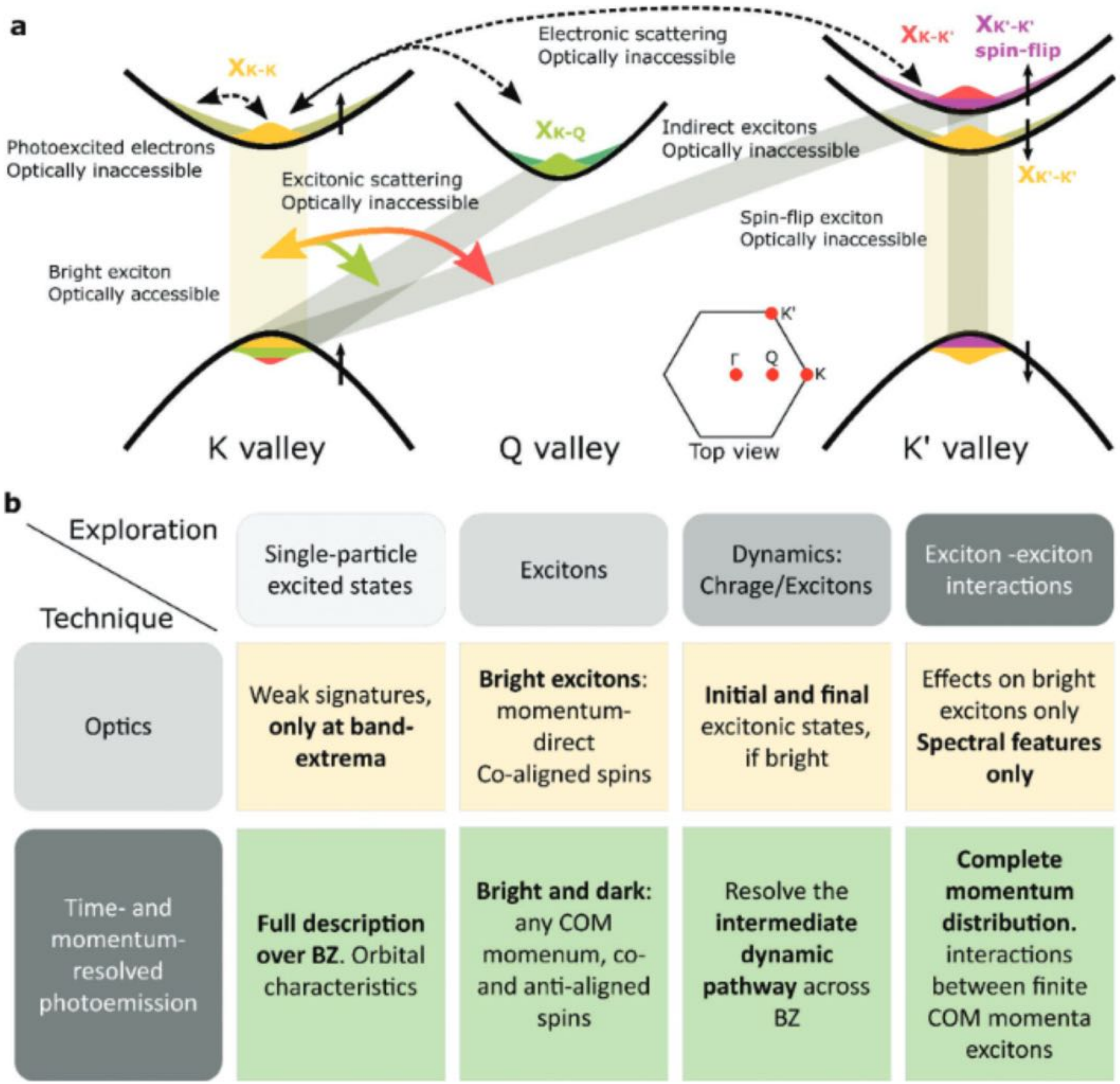


METIS	
Mounting Flange	DN150CF
Start Energy	0-2000 eV
Energy Resolution	<15 meV
Angular Resolution	<0.1°
k-Resolution	< 0.01 Å^{-1}
Lateral Resolution (PEEM-mode)	< 50 nm
Lateral Resolution (ARPES-mode)	< 2 µm
Acceptance Angle	up to +-90°
Exctractor Voltage	up to 29 kV
Field Apertures	200 µm down to 2 µm (in sample coordinates)

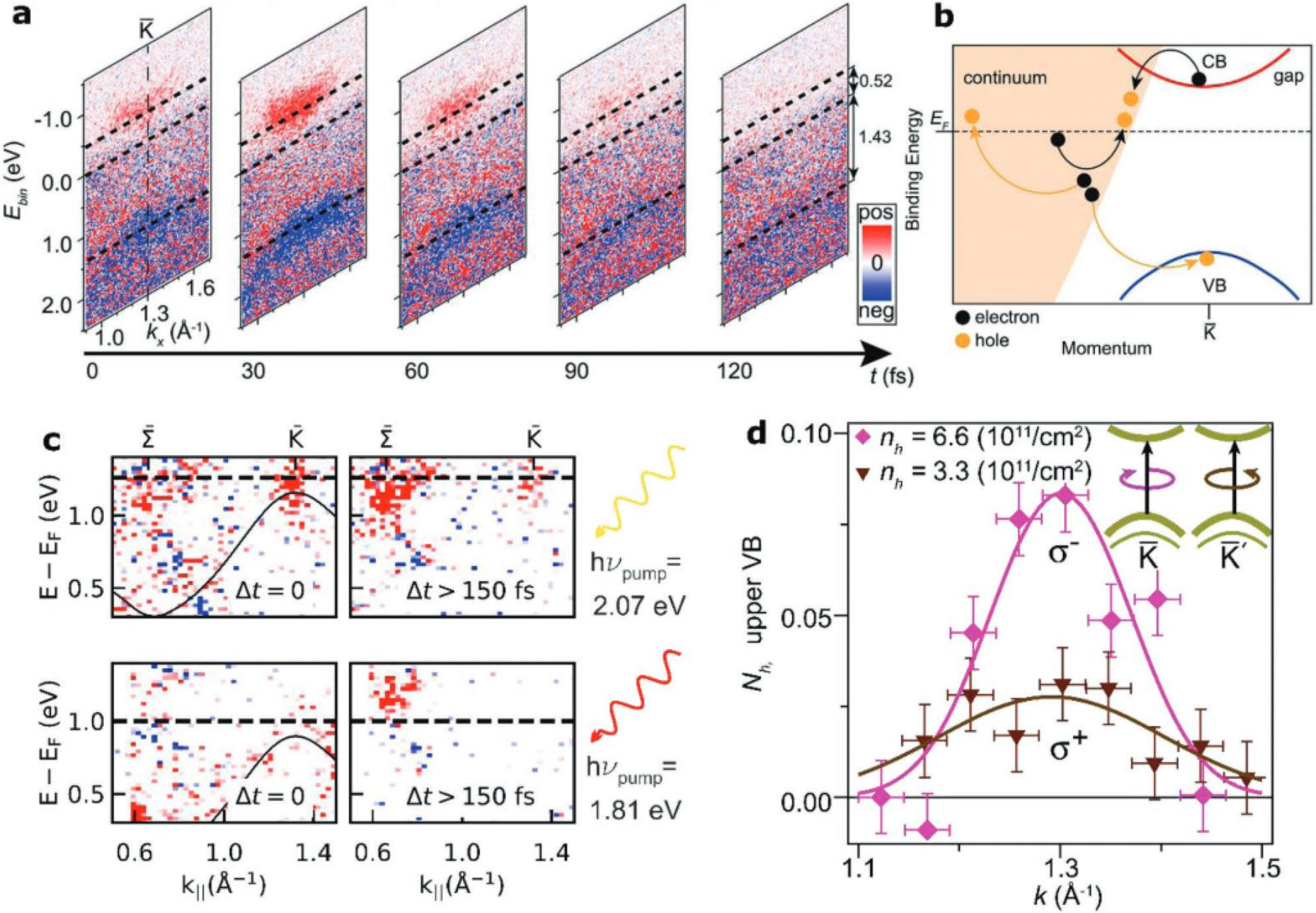


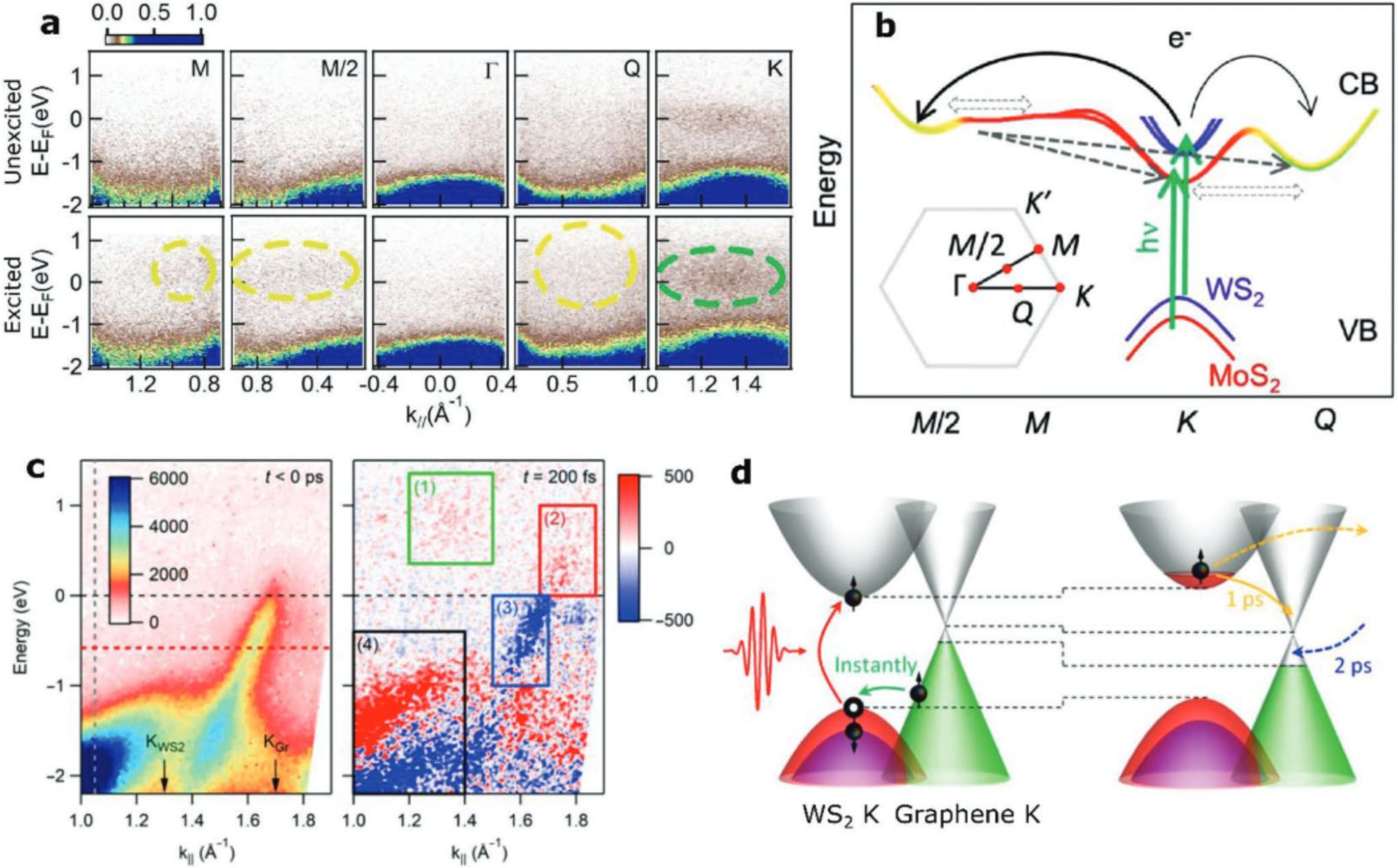
Delay Line Detector	
max. permanent measurement count rate	> 8x10^6 cps (10^8 tolerant)
Count Rate Linearity Range	> 2x10^6 cps
Typical Time Resolution (position integrated)	< 180 ps < 110 ps (best achieved)
Start Repetition Rate	≤ 150 MHz; ≤ 9 MHz without prescaler
Typical Lateral Resolution	< 100 µm < 50 µm (best achieved)
Multi Hit Designs	optional, up to 30 simultaneous hits (with multianode detector layout)

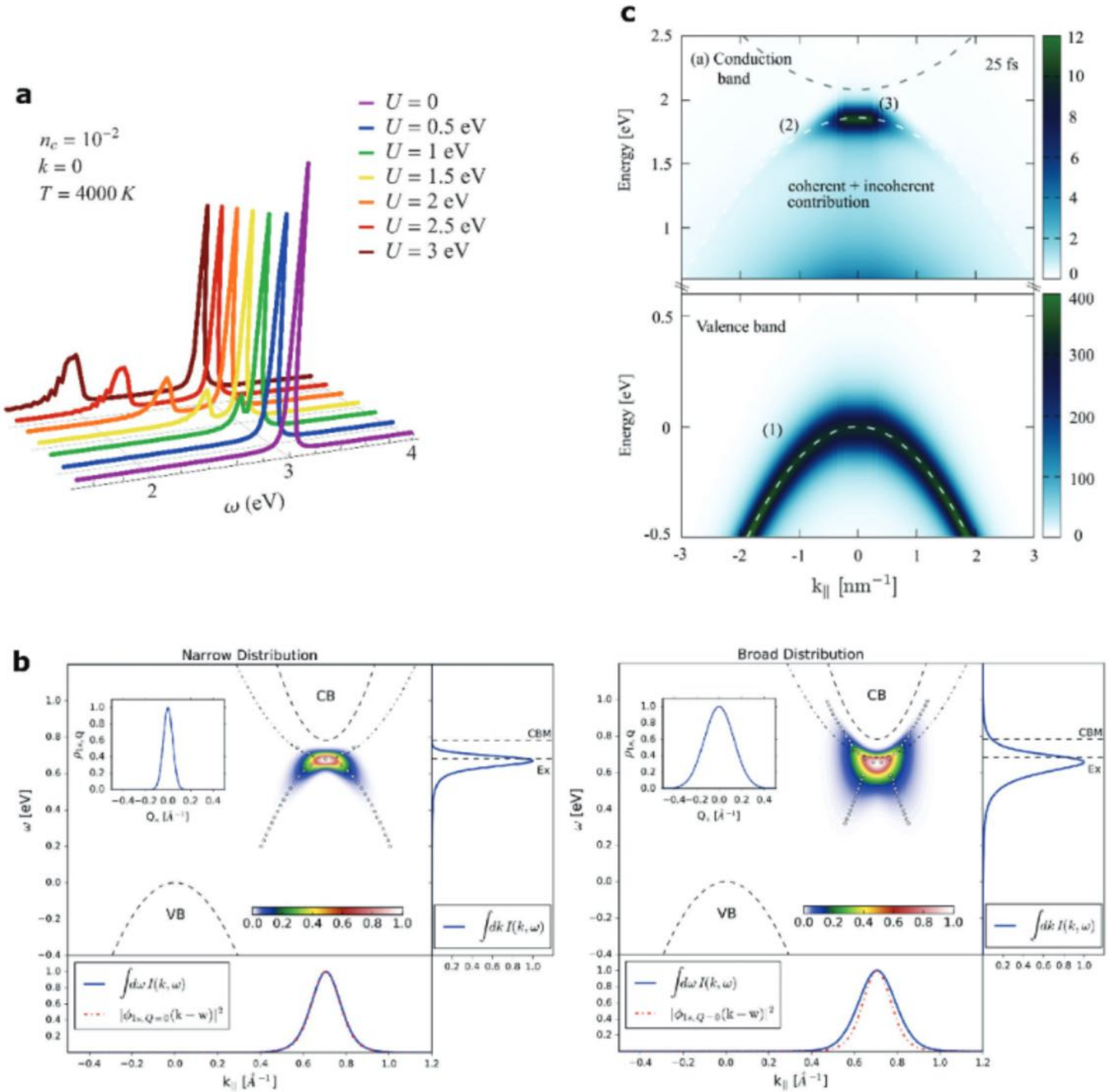
https://www.specs-group.com/fileadmin/user_upload/products/brochures/SPECS_Brochure-METIS_RZ_web.pdf

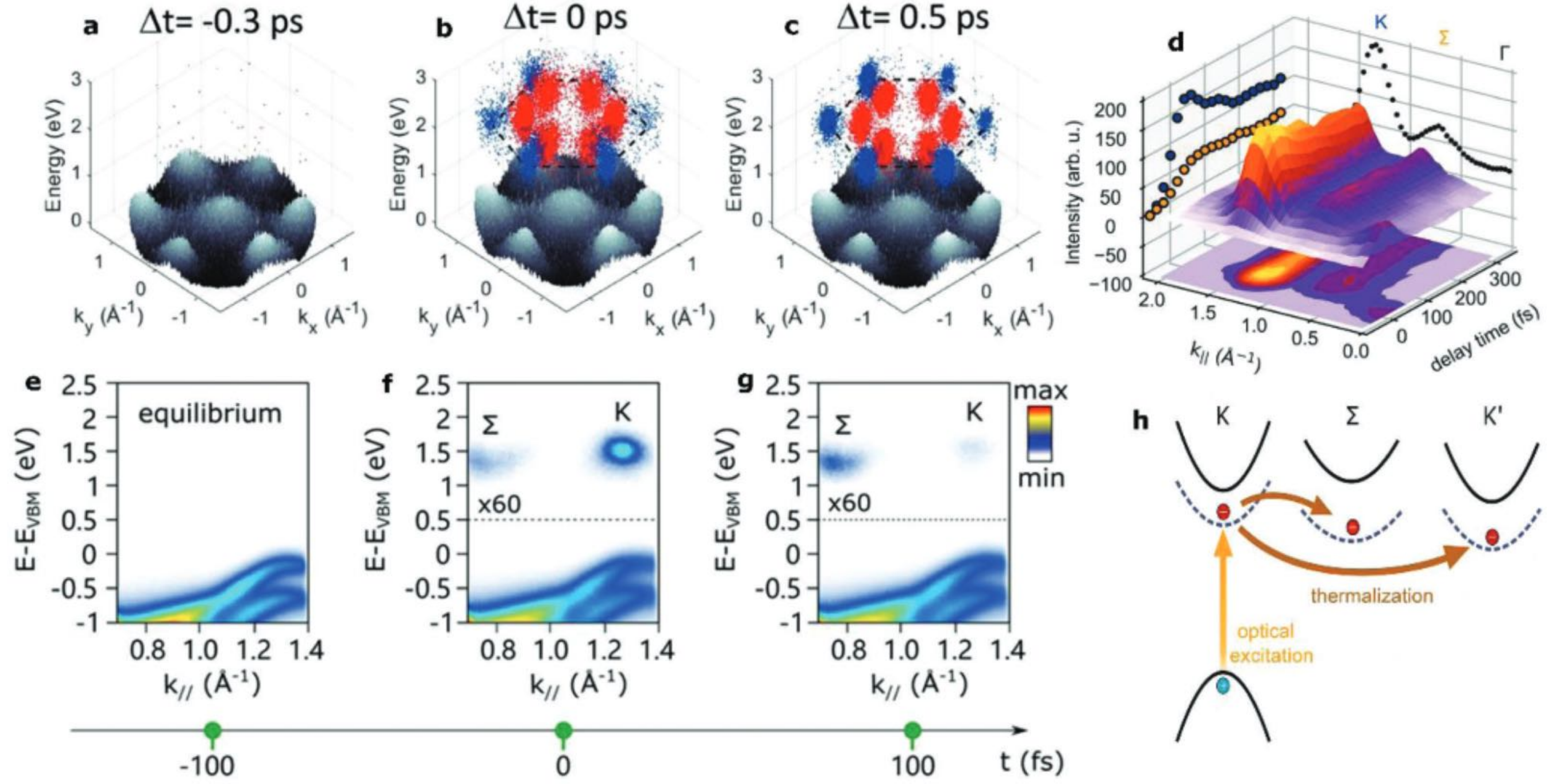


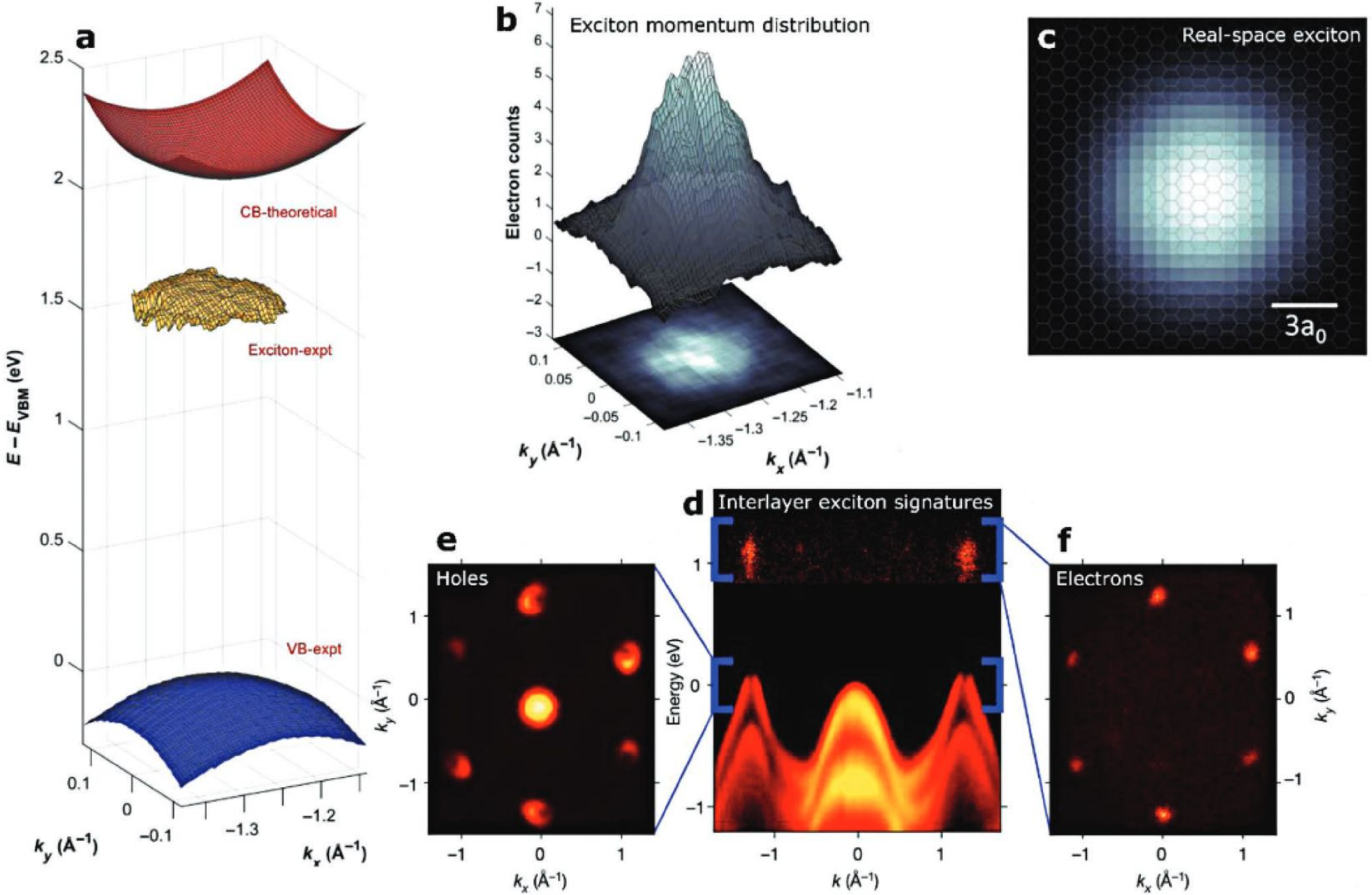
Exploration / Technique	Single-particle excited states	Excitons	Dynamics: Chrage/Excitons	Exciton -exciton interactions
Optics	Weak signatures, **only at band-extrema**	**Bright excitons**: momentum-direct Co-aligned spins	**Initial and final** excitonic states, if bright	Effects on bright excitons only **Spectral features only**
Time- and momentum-resolved photoemission	**Full description over BZ**. Orbital characteristics	**Bright and dark:** any COM momenum, co- and anti-aligned spins	Resolve the **intermediate dynamic pathway** across BZ	**Complete momentum distribution.** interactions between finite COM momenta excitons

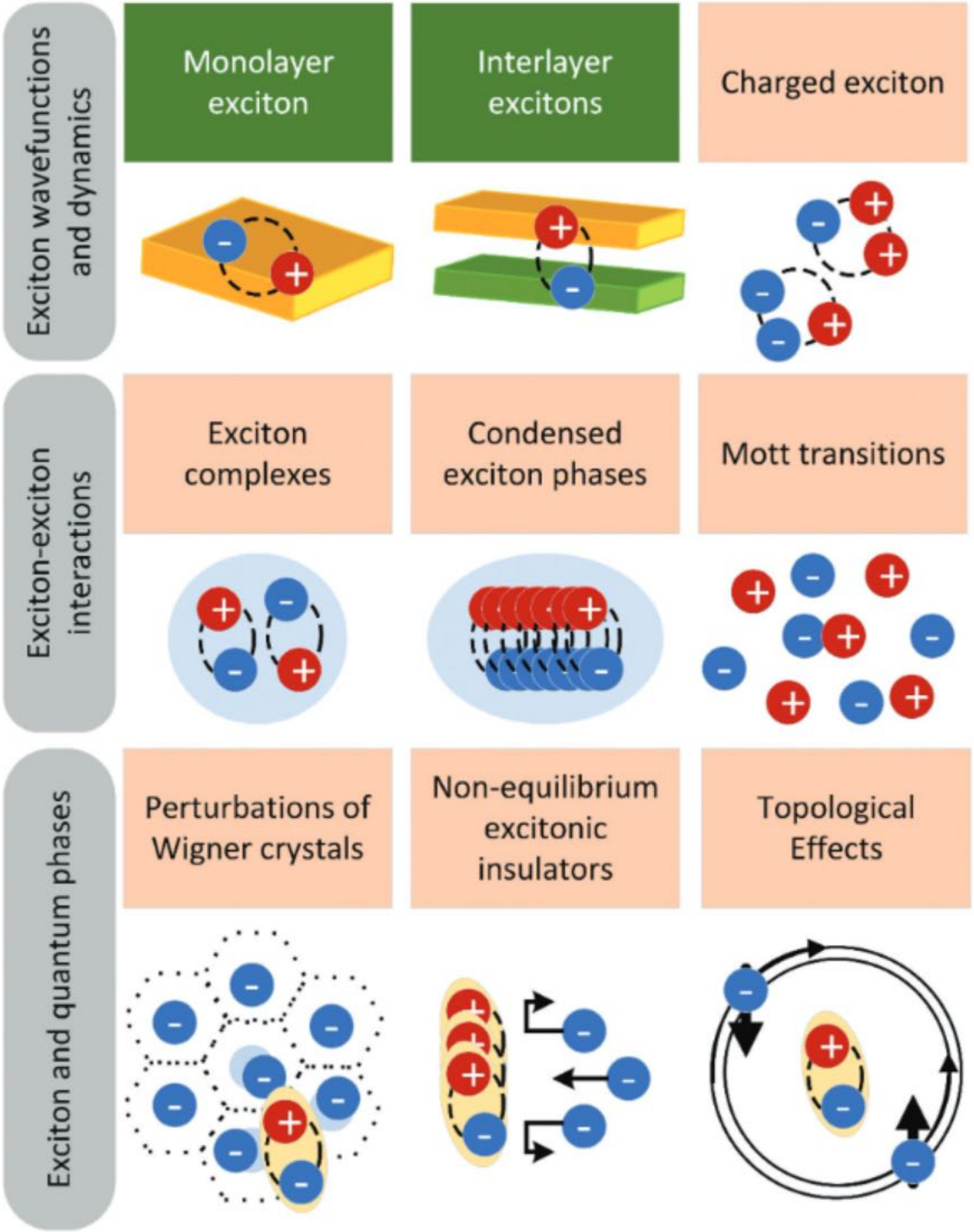

a
K̄
-1.0
0.0
1.0
2.0
E_{bin} (eV)
1.0
1.3
1.6
k_x (Å⁻¹)
0.52
1.43
pos
0
neg
0
30
60
90
120
t (fs)
b
continuum
CB
gap
E_F
Binding Energy
VB
K̄
electron
hole
Momentum
c
Σ̄
K̄
Σ̄
K̄
E − E$_F$ (eV)
1.0
0.5
Δt = 0
Δt > 150 fs
hν_{pump}=
2.07 eV
E − E$_F$ (eV)
1.0
0.5
Δt = 0
Δt > 150 fs
hν_{pump}=
1.81 eV
0.6
1.0
1.4
k$_{||}$(Å⁻¹)
0.6
1.0
1.4
k$_{||}$(Å⁻¹)
d
n_h = 6.6 (10¹¹/cm²)
n_h = 3.3 (10¹¹/cm²)
K̄
K̄′
σ⁻
σ⁺
0.10
0.05
0.00
$N_{h,}$ upper VB
1.1
1.3
1.5
k (Å⁻¹)

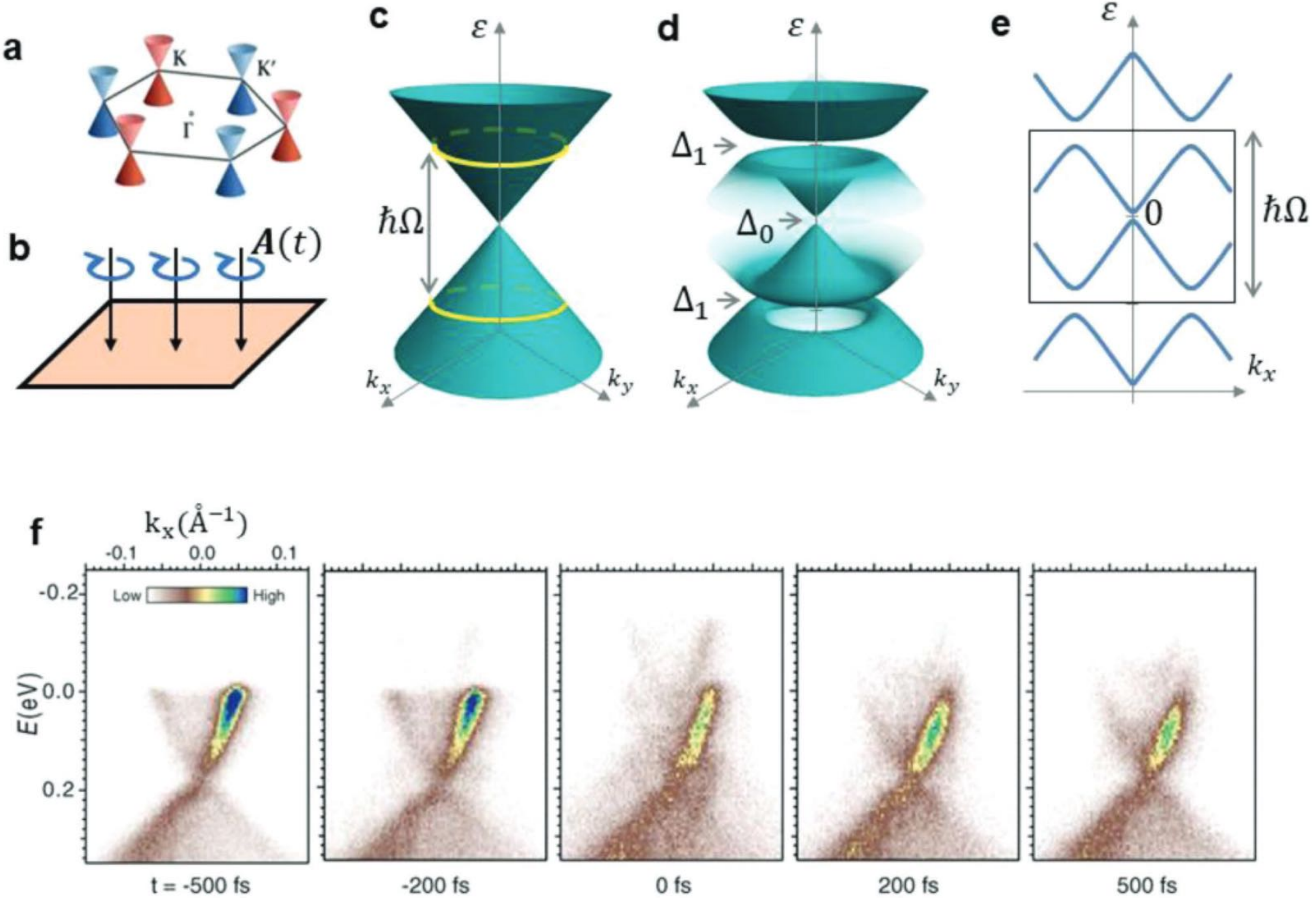

a
0.0 0.5 1.0
M
M/2
Γ
Q
K
Unexcited E-E_F(eV)
Excited E-E_F(eV)
1
0
-1
-2
1.2 0.8
0.8 0.4
-0.4 0.0 0.4
0.5 1.0
1.2 1.4
k_//(Å^-1)
b
e^-
CB
Energy
K'
M/2
M
Γ
K
Q
hν
WS2
VB
MoS2
M/2 M K Q
c
t < 0 ps
6000
4000
2000
0
Energy (eV)
K_WS2
K_Gr
1.0 1.2 1.4 1.6 1.8
k_|| (Å^-1)
t = 200 fs
(1)
(2)
(3)
(4)
500
0
-500
d
Instantly
1 ps
2 ps
WS2 K
Graphene K

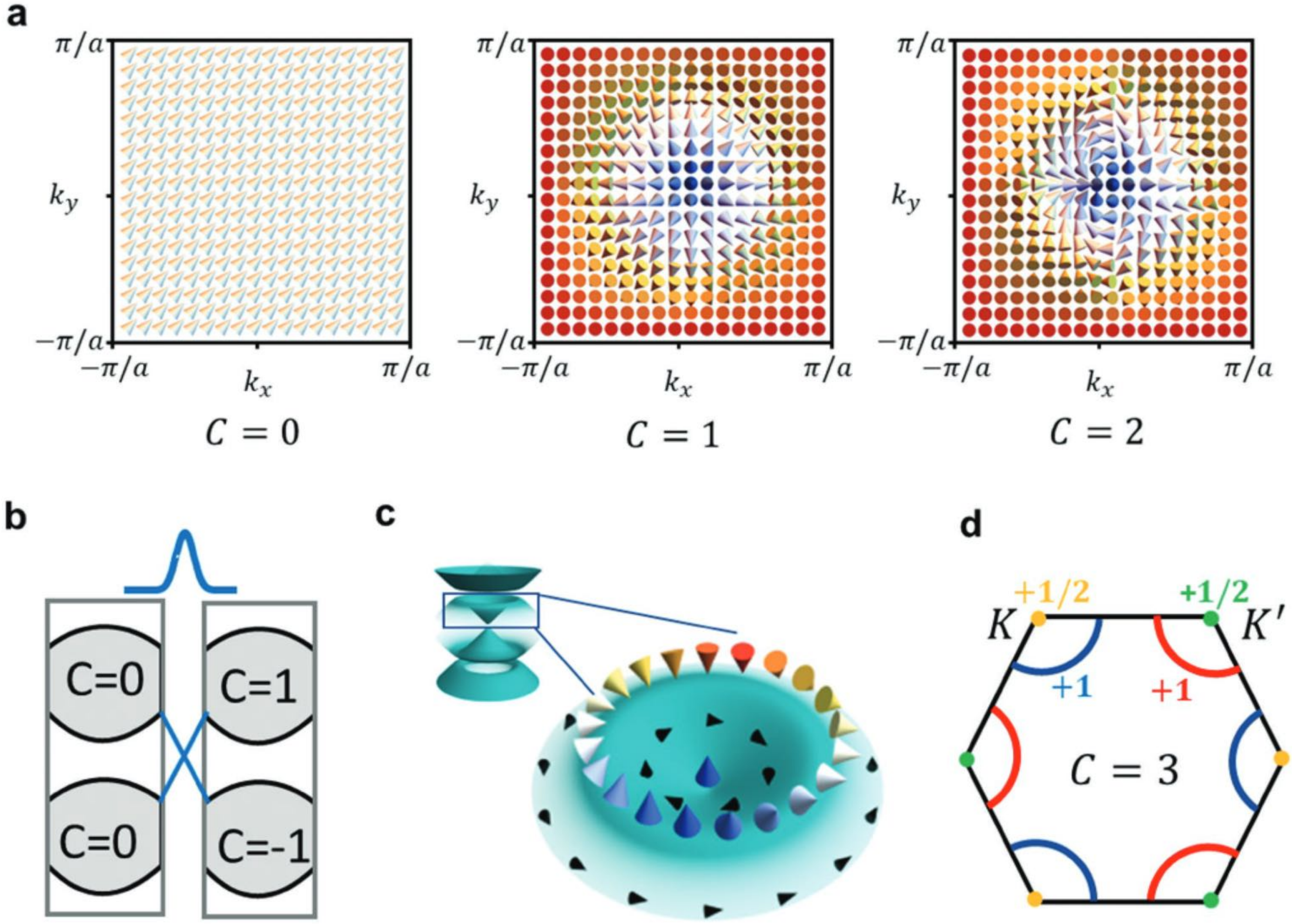

a
$n_c = 10^{-2}$
$k = 0$
$T = 4000\,K$
$U = 0$
$U = 0.5$ eV
$U = 1$ eV
$U = 1.5$ eV
$U = 2$ eV
$U = 2.5$ eV
$U = 3$ eV
2
3
4
ω (eV)
c
(a) Conduction band
25 fs
(2)
(3)
coherent + incoherent contribution
Energy [eV]
2.5
2
1.5
1
12
10
8
6
4
2
0
Valence band
(1)
0.5
0
-0.5
400
300
200
100
0
-3
-2
-1
0
1
2
3
$k_\parallel$ [nm^{-1}]
b
Narrow Distribution
Broad Distribution
CB
VB
CBM
Ex
ω [eV]
$\rho_{1s,Q}$
Q_x [Å^{-1}]
$\int dk\, I(k,\omega)$
$\int d\omega\, I(k,\omega)$
$|\phi_{1s,Q=0}(\mathbf{k}-\mathbf{w})|^2$
0.0 0.2 0.4 0.6 0.8 1.0
-0.4 -0.2 0.0 0.2 0.4 0.6 0.8 1.0 1.2
$k_\parallel$ [Å^{-1}]

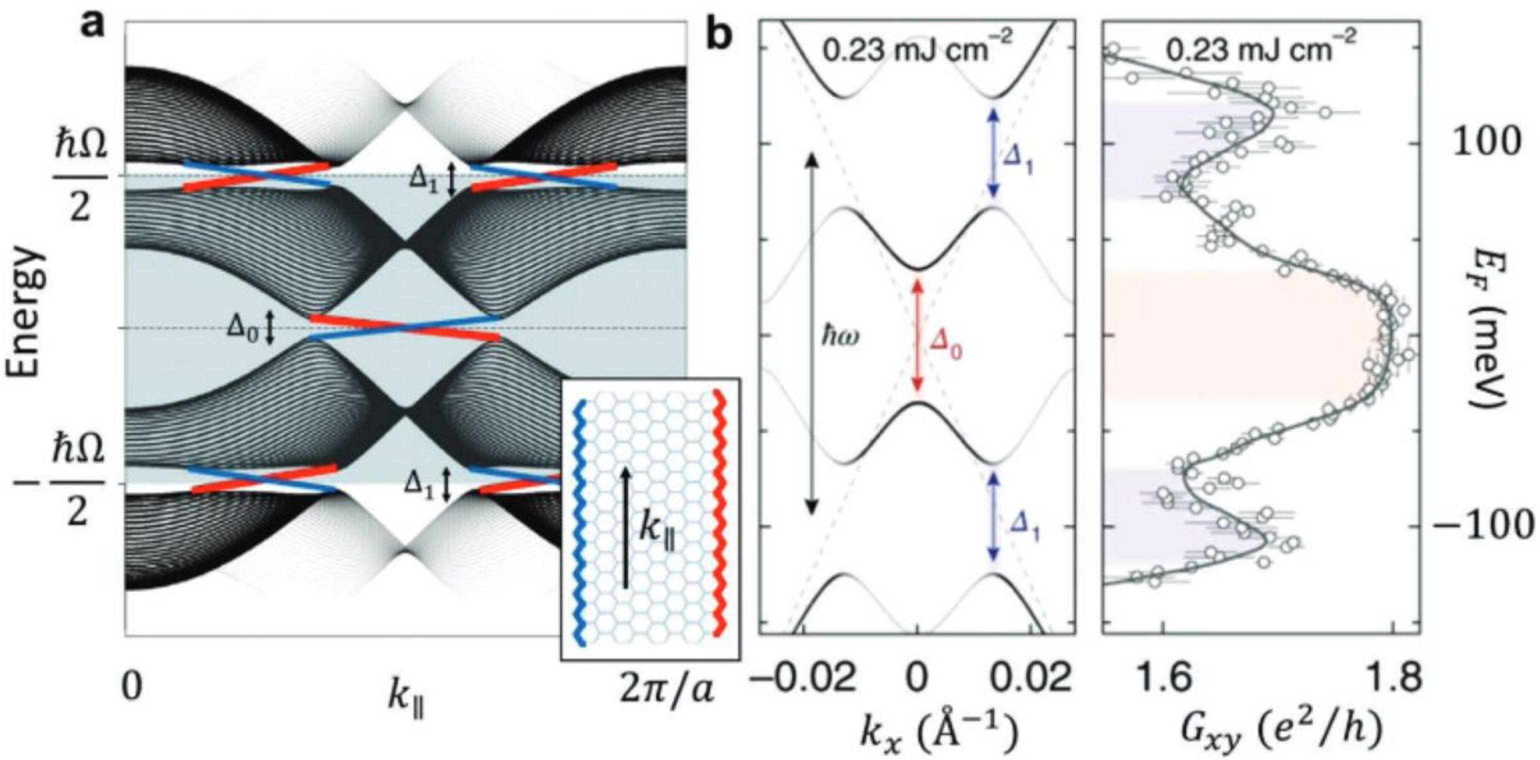

a
Δt= -0.3 ps
b
Δt= 0 ps
c
Δt= 0.5 ps
Energy (eV)
0
1
2
3
k_y (Å^{-1})
k_x (Å^{-1})
-1
0
1
d
K
Σ
Γ
Intensity (arb. u.)
200
150
100
50
0
−50
−100
$k_{//}$ (Å^{-1})
2.0
1.5
1.0
0.5
0.0
delay time (fs)
0
100
200
300
e
equilibrium
f
g
Σ
K
x60
$E-E_{VBM}$ (eV)
2.5
2
1.5
1
0.5
0
-0.5
-1
$k_{//}$ (Å^{-1})
0.8
1.0
1.2
1.4
max
min
h
K
Σ
K'
thermalization
optical excitation
-100
0
100
t (fs)


a
2.5
2
1.5
1
0.5
0
E − E_VBM (eV)
CB-theoretical
Exciton-expt
VB-expt
0.1
0
−0.1
k_y (Å⁻¹)
−1.3
−1.2
k_x (Å⁻¹)
b
Exciton momentum distribution
Electron counts
7
6
5
4
3
2
1
0
−1
−2
−3
0.1
0.05
0
−0.05
−0.1
k_y (Å⁻¹)
−1.35
−1.3
−1.25
−1.2
−1.1
k_x (Å⁻¹)
c
Real-space exciton
3a_0
d
Interlayer exciton signatures
Energy (eV)
1
0
−1
−1
0
1
k (Å⁻¹)
e
Holes
k_y (Å⁻¹)
1
0
−1
−1
0
1
k_x (Å⁻¹)
f
Electrons
k_y (Å⁻¹)
1
0
−1
−1
0
1
k_x (Å⁻¹)


Exciton wavefunctions and dynamics
Monolayer exciton
Interlayer excitons
Charged exciton
Exciton-exciton interactions
Exciton complexes
Condensed exciton phases
Mott transitions
Exciton and quantum phases
Perturbations of Wigner crystals
Non-equilibrium excitonic insulators
Topological Effects


a
K
K′
Γ
b
A(t)
c
ε
ħΩ
k_x
k_y
d
ε
Δ_1
Δ_0
Δ_1
k_x
k_y
e
ε
ħΩ
0
k_x
f
k_x(Å^{-1})
-0.1
0.0
0.1
Low
High
-0.2
0.0
0.2
E(eV)
t = -500 fs
-200 fs
0 fs
200 fs
500 fs


a
π/a
k_y
−π/a
−π/a
k_x
π/a
$C = 0$
$C = 1$
$C = 2$
b
C=0
C=0
C=1
C=-1
c
d
+1/2
+1/2
K
K′
+1
+1
$C = 3$


a
ħΩ/2
Energy
Δ₁
Δ₀
Δ₁
−ħΩ/2
k∥
0
k∥
2π/a
b
0.23 mJ cm⁻²
Δ₁
ħω
Δ₀
Δ₁
−0.02
0
0.02
k_x (Å⁻¹)
0.23 mJ cm⁻²
1.6
1.8
G_xy (e²/h)
100
−100
E_F (meV)