Recent progress in ToF momentum microscopy
towards spin- and time resolution

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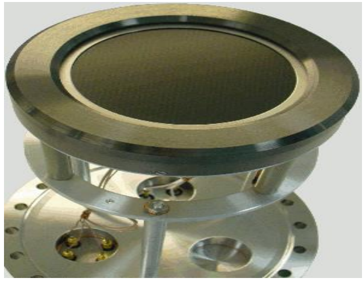
JG|U

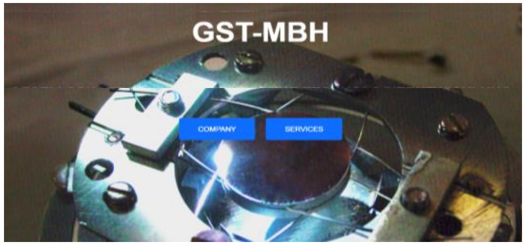
SPIN+X
SFB/TRR 173
Kaiserslautern • Mainz

SURFACE CONCEPT



JOHANNES GUTENBERG UNIVERSITÄT MAINZ

DESY



SXP @ Eu-XFEL meeting, Jan. 24, 2022

Outline

Introduction

- Multidimensional aspect of photoemission
- ToF-MM *vs* conventional ARPES

Instrument evolution

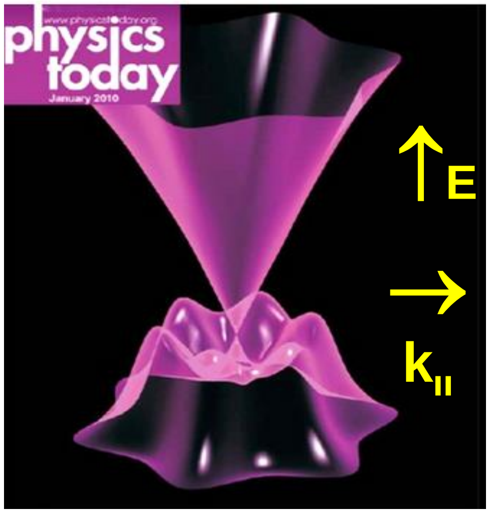
- Space-charge correction / suppression in ToF-MM
- Valence-band mapping → electronic structure
- Photoelectron diffraction XPD → geometric structure
- PEEM mode → 'sub-micron ARPES'

Spin filter

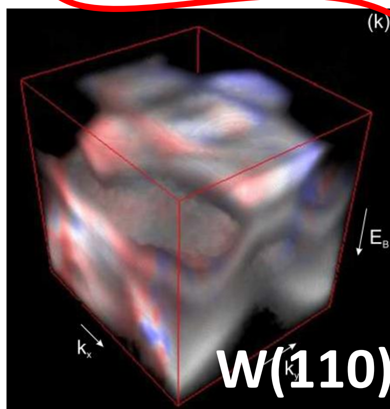
- Progress in spin filtering (high energies, larger energy band)
- First fs time resolved spin measurements (Zurich group)

Multidimensional aspect of photoemission

Static photoemission: 1 scalar and 2 vector quantities in 4D parameter space



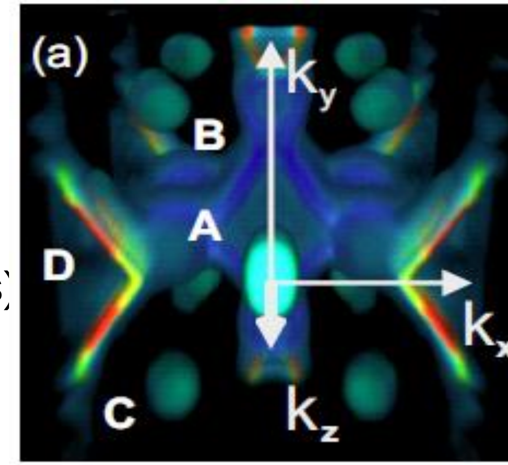
Band dispersion of topological surface state

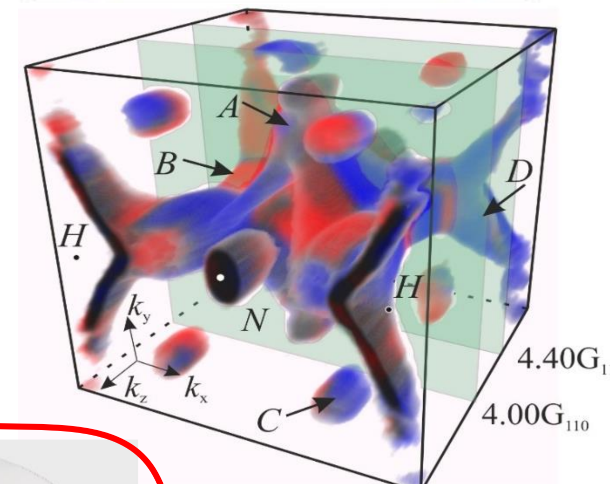


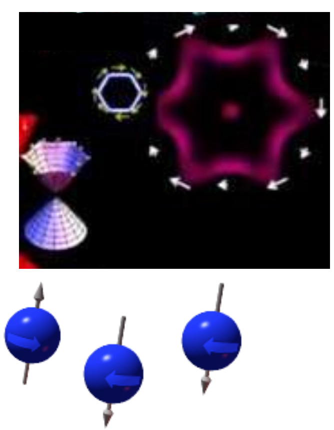
2D k-space

3D k-space

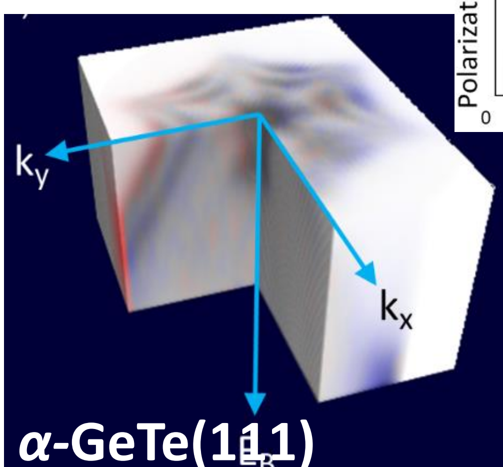
Energy isosurface →
decorated with local v_e
(one of 100 such isosurfaces)

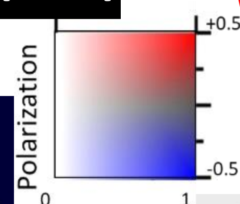


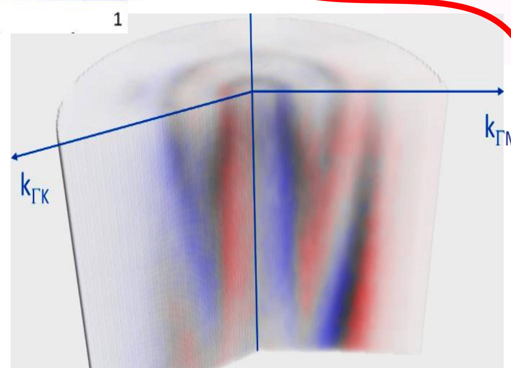




Spin texture







Fermi surface decorated with circular dichroism asymmetry
(one of 100 such isosurfaces)

Multidimensional aspect of photoemission

$$\rho_{\downarrow,\uparrow}\,(k_x,\ k_y,\ k_z,\ E_B)$$

ρ ← spectral function

P_x, P_y, P_z ← spin-polarization texture

$A_{C(L)DAD}$ ← circular (linear) dichroism in photoemission

$(k_x,\ k_y,\ k_z,\ E_B)$

Static photoemission: 1 scalar and 2 vector quantities in 4D parameter space

In tr photoemission additional „coordinates“ are important:

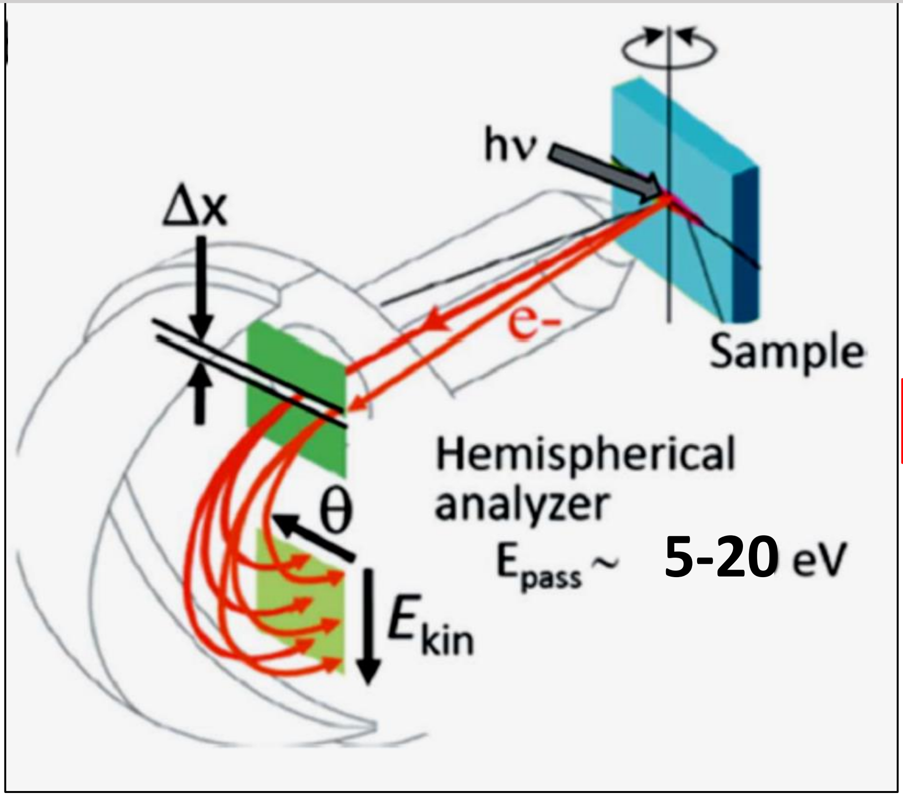
Lattice, electron and spin temperature T_l, T_e, T_s

Pump-probe delay τ

Pump fluence …

tr photoemission: complex multidimensional scenario

Dimensionality of recording



I (E_{kin},θ_x)

θ_y via scanning; k_z via hν scan

Kreios, Astraios:

I (E_{kin},k_x)

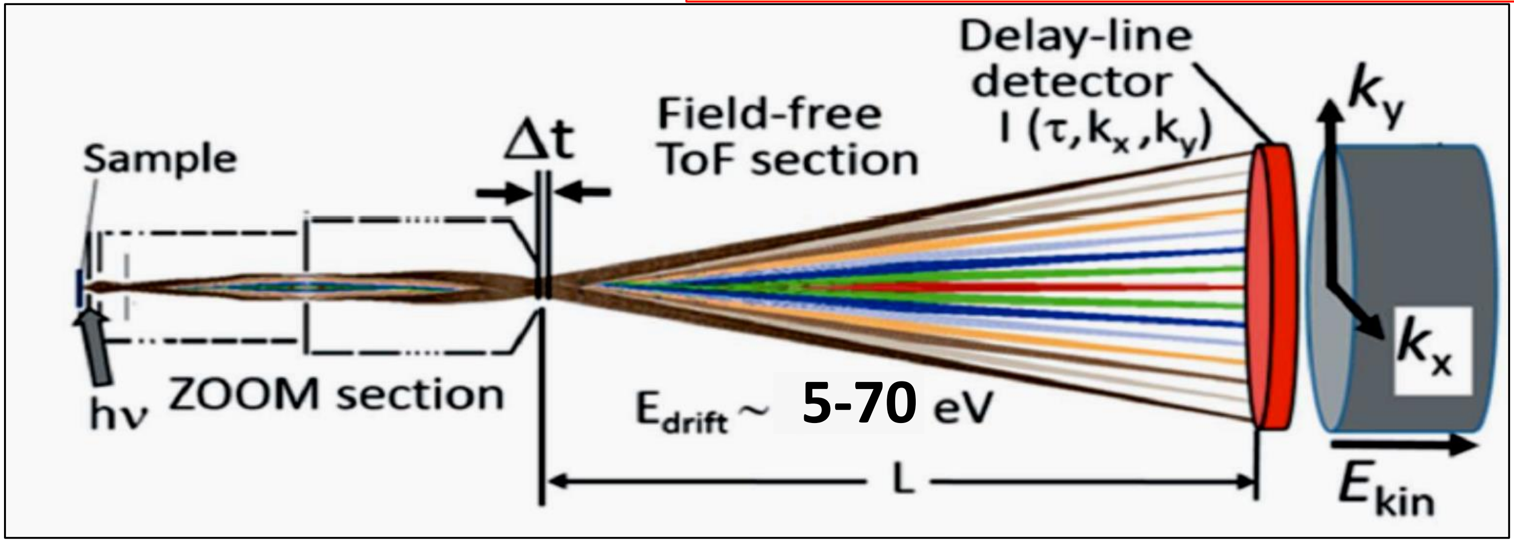
k_y via scanning; k_z via hν scan

mechanical slit Δx

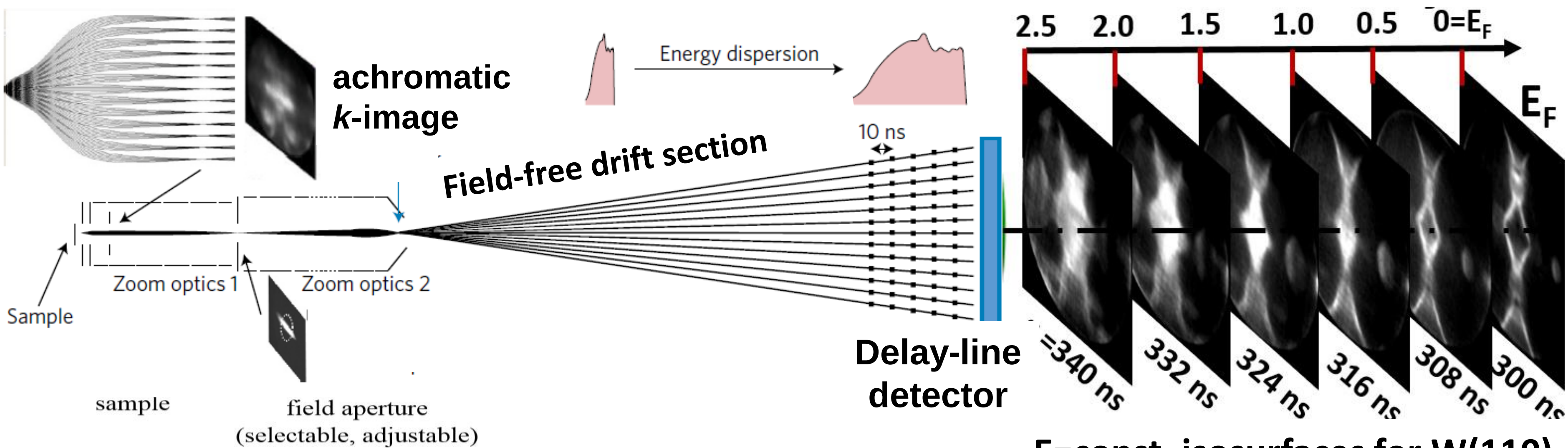
I (E_{kin},k_x,k_y)

k_z via hν scan or via large k-fields

no mechanical slits, just time „slit" Δt



The Mainz solution: ToF MM

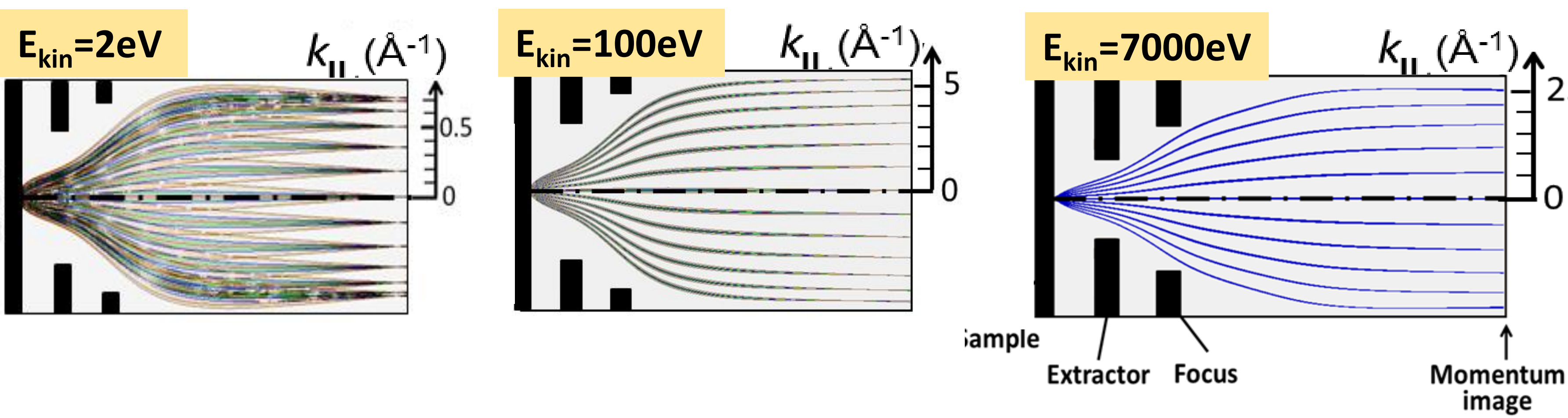


E=const. isosurfaces for W(110)

PEEM-mode FoV: 8-800 μm

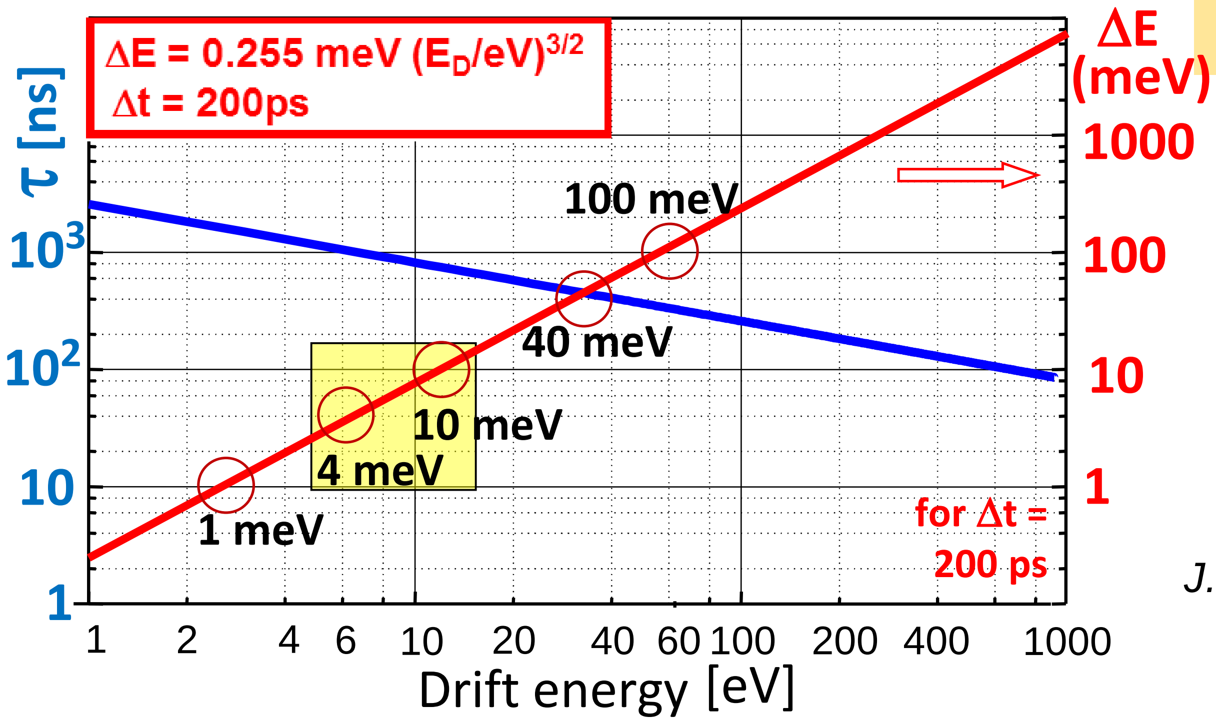
k-mode FoV: 1-6 $Å^{-1}$

From threshold to HAXPES:

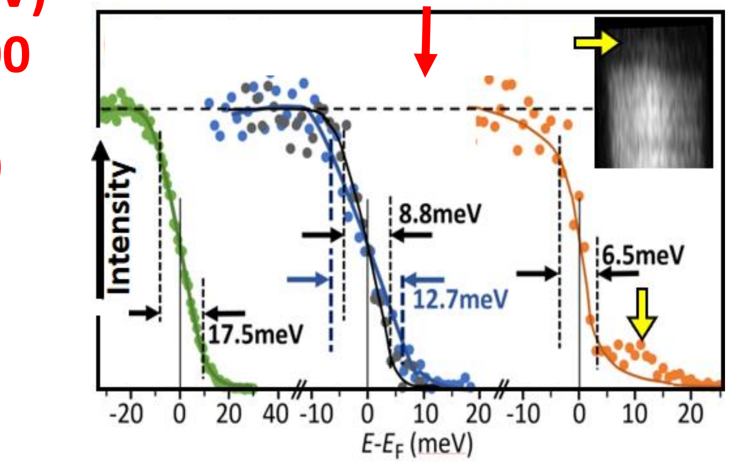


ToF MM key features: **Energy resolution**

Dispersion of ToF section



Resolution measured in the lab:
~ 9 meV (ToF)
~ 4 meV (ToF & HSA hybrid)

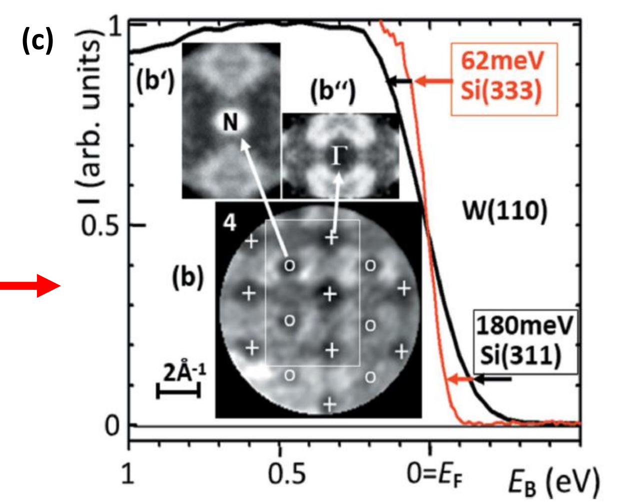


J. Synchr. Radiation **28**, 1891 (2021)

Resolution measurements @ P22:
Si(311): 180 meV FWHM @ 5980 eV
Si(333): 62 meV FWHM „

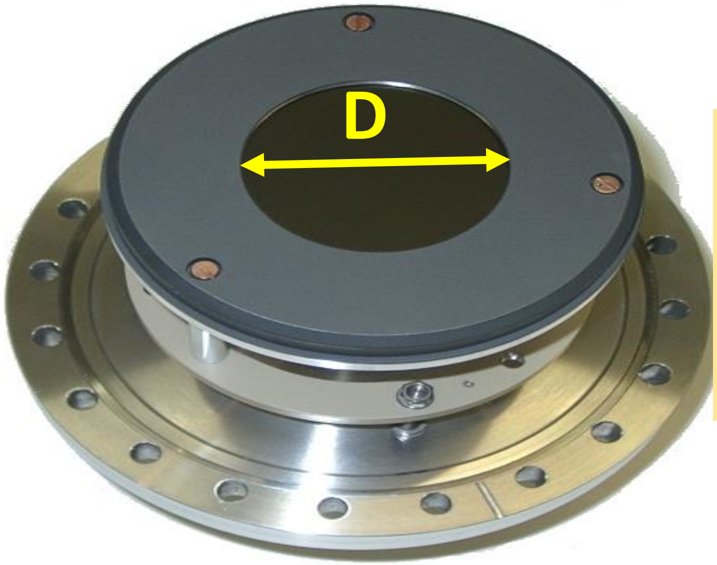
ToF resolution 40 meV

Resolving power: 10^5



J. Synchr. Radiation **26,** 1996 (2019)

ToF MM key features: **Detector, *k*-resolution**



<u>*k*-resolution:</u>

Given by no. of points along image diameter D

$\geq$ 400 resolved points: : $\leq$0.01 Å^{-1} for dia. 4 Å^{-1}

0.03 Å^{-1} for dia. 12 Å^{-1}

4-Quadrant Delayline Detector

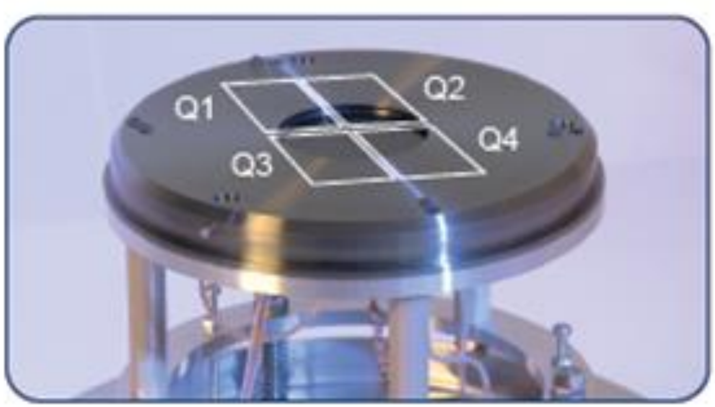
With the 4Q-detector, Surface Concept created a detector with a highly increased multihit capability. With a separation of the delayline into four quadrants, up to 400 multihits in 1 µs can be detected (or 4 multihits with absolutely no dead-time).

<u>Delayline Detector:</u>

150 ps time resolution

40 µm spatial resolution

8 Mcps



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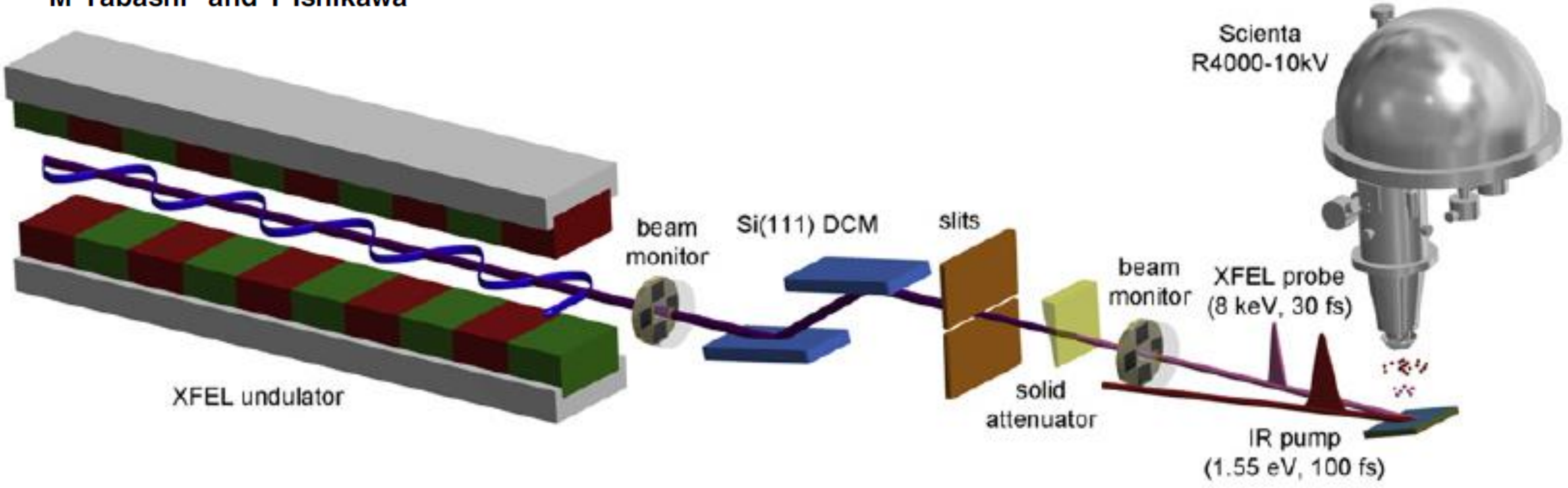
“Space charge is the main obstacle of fs photoemission”

New Journal of Physics **16** (2014) 123045
doi:10.1088/1367-2630/16/12/123045

Time-resolved HAXPES at SACLA: probe and pump pulse-induced space-charge effects

L-P Oloff[1,2], M Oura[2], K Rossnagel[1,2], A Chainani[2], M Matsunami
R Eguchi[2,4,5], T Kiss[6], Y Nakatani[6], T Yamaguchi[6], J Miyawaki[7],
M Taguchi[2,9], K Yamagami[6], T Togashi[8], T Katayama[8], K Ogawa[2]
M Yabashi[2] and T Ishikawa[2]



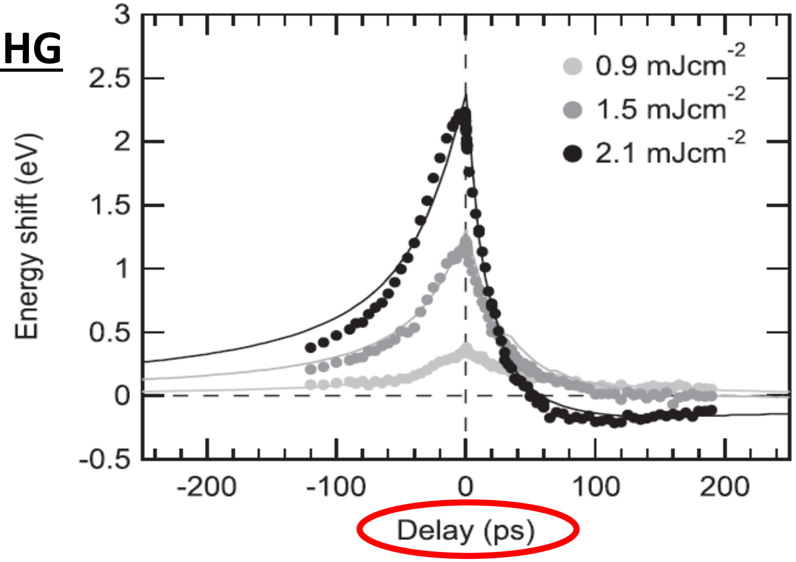
"Space charge is the main obstacle of fs photoemission"

Pump laser-induced space-charge effects in HHG-driven time- and angle-resolved photoelectron spectroscopy

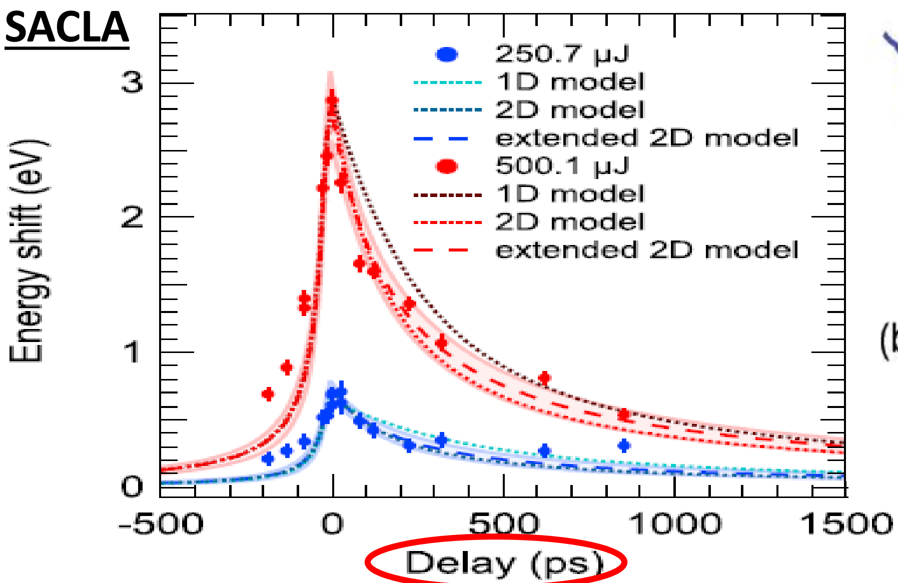
JOURNAL OF APPLIED PHYSICS **119**, 225106 (2016)

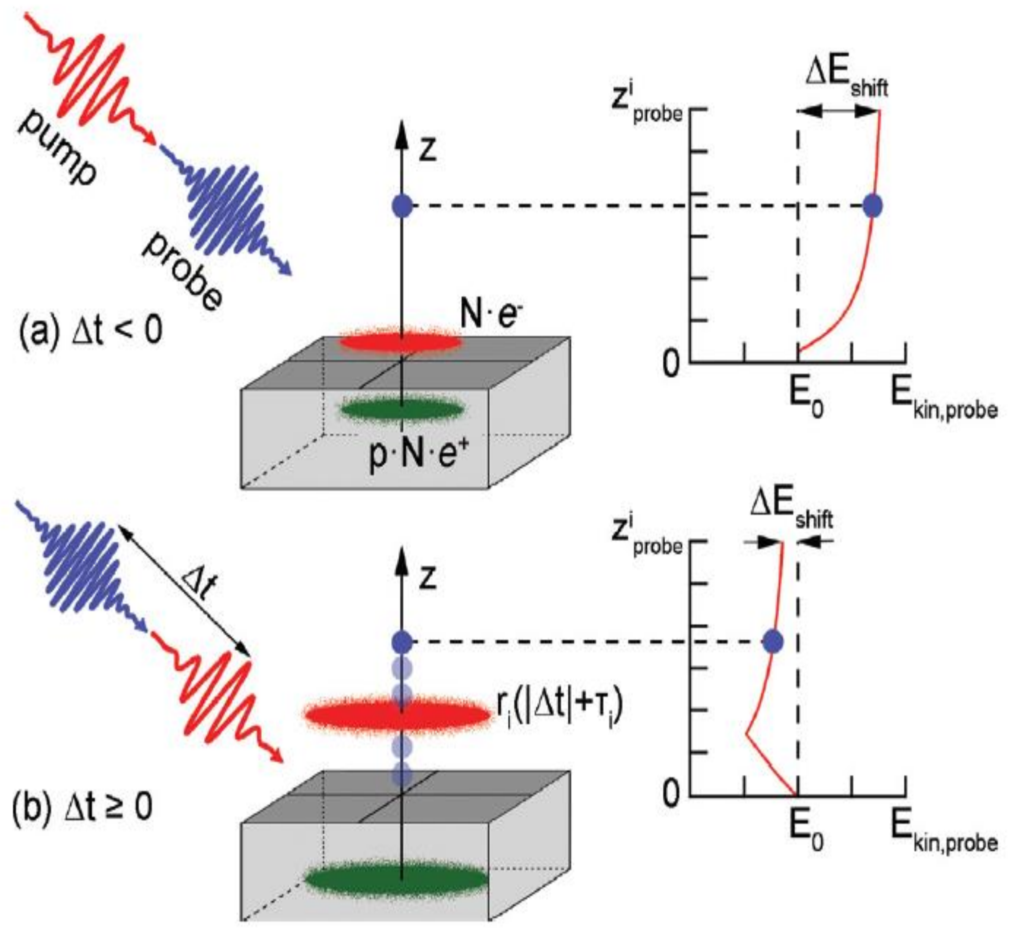
L.-P. Oloff,[a)] K. Hanff, A. Stange, G. Rohde, F. Diekmann, M. Bauer, and K. Rossnagel

HHG

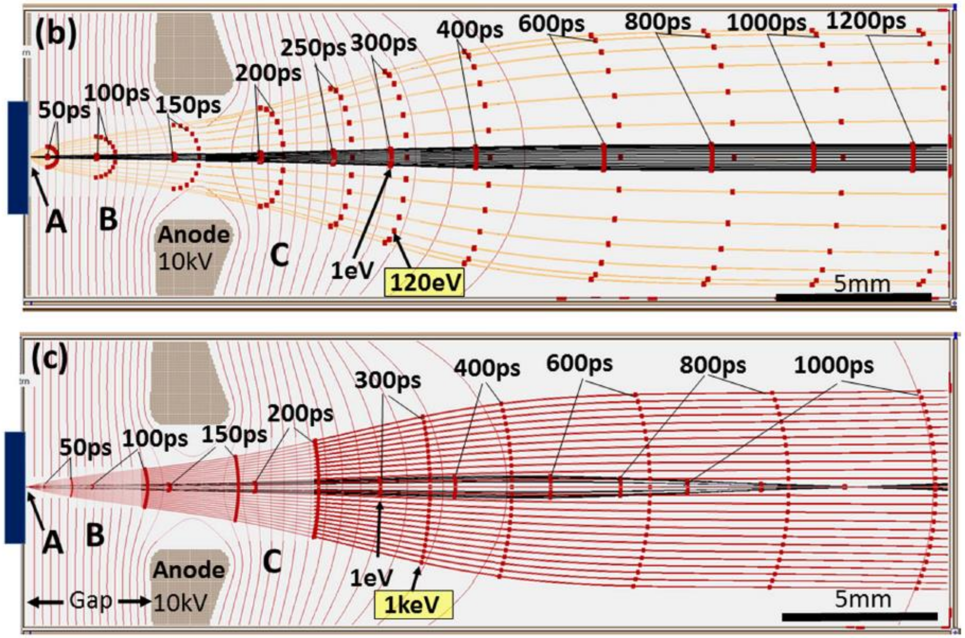


SACLA

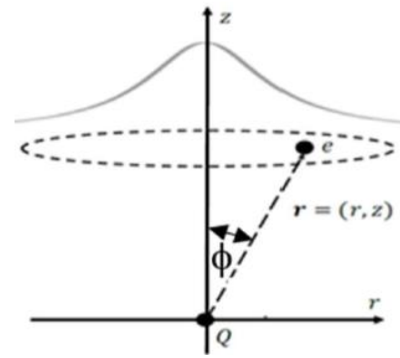


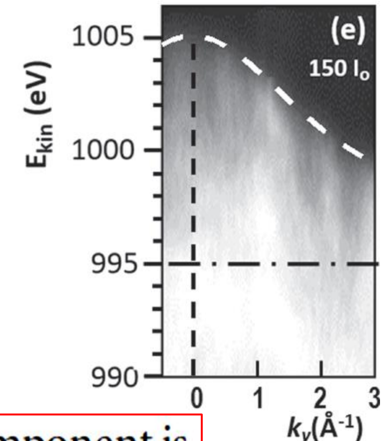


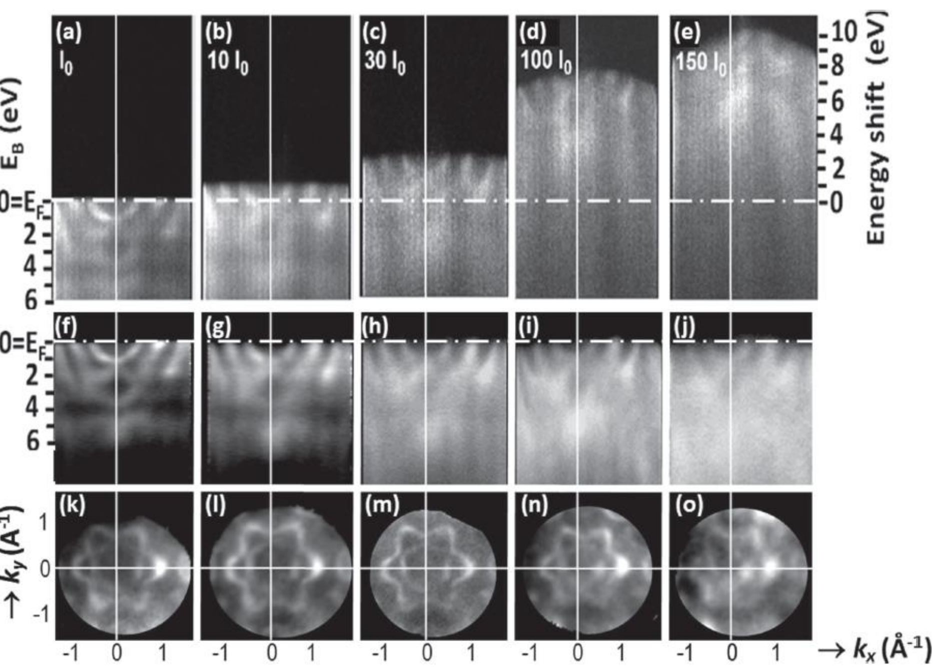
Refinement of the Oloff et al. space-charge model: **LONG-RANGE REGIME**



B. Schönhense et al.,
New J. Phys. **20**, 033004 (2018)
semi-analytical model







The axial force component is

$$F_z = F_{\mathrm{Coulomb}} \cos\phi = \frac{Qe}{4\pi\epsilon_0}\frac{1}{(r^2+z^2)}\frac{z}{\sqrt{r^2+z^2}}$$

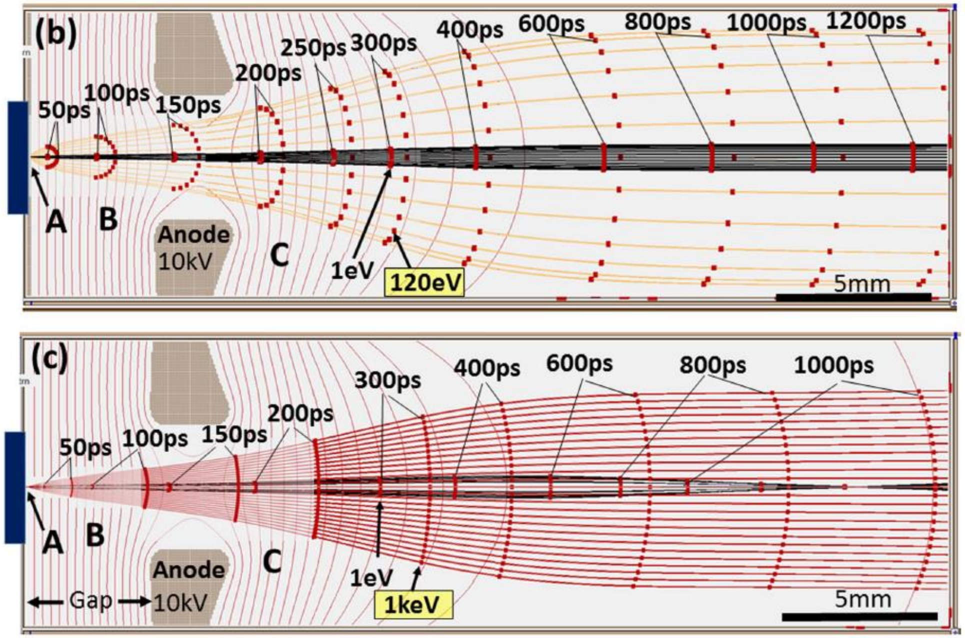
$$\propto \frac{1}{(r^2+z^2)^{3/2}} \propto \frac{1}{(1+\tan^2\phi)^{\frac{3}{2}}}.$$

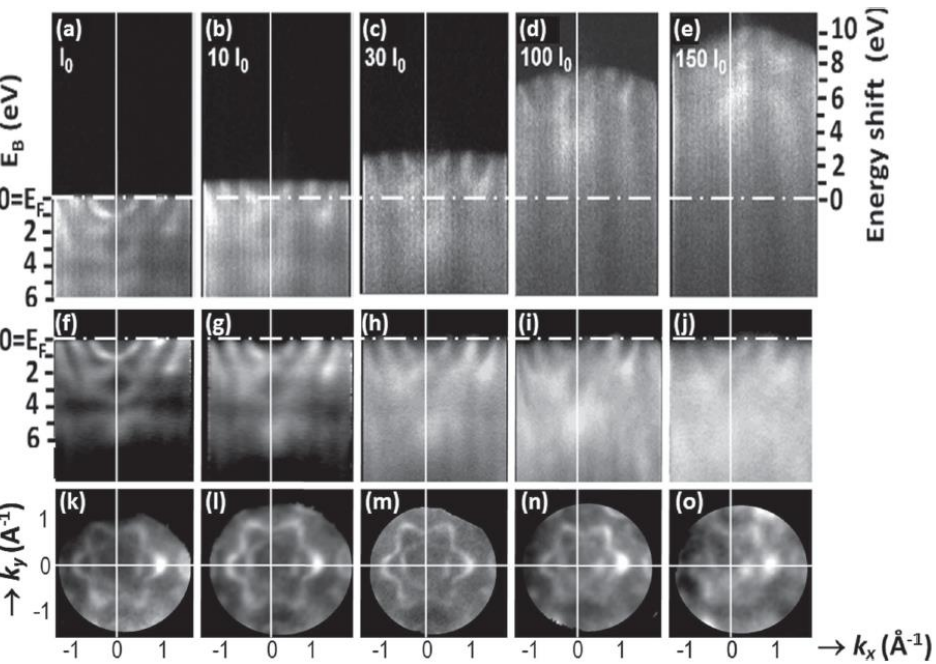
Correction algorithm

generic functional form of the space-charge effect
is then fitted to the observed data

$$\Delta t_{\mathrm{F}}(x, y) \propto \frac{Q}{(b^2+(x-x_0)^2+(y-y_0)^2)^{3/2}}.$$

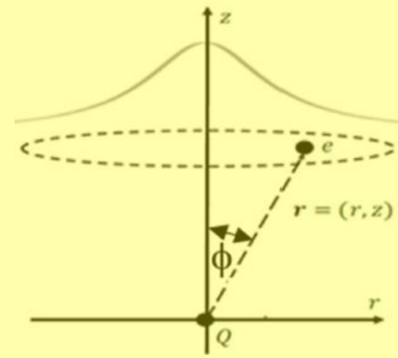
CORRECTION of space-charge shift/broadening by data processing

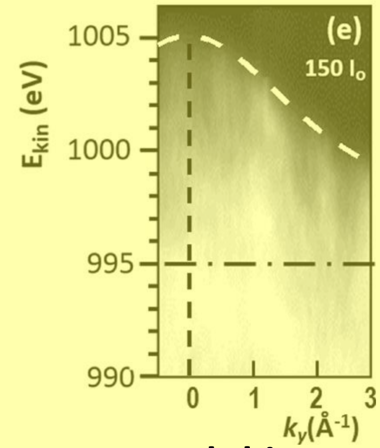




Space-charge correction:
a posteriori data treatment

B. Schönhense et al.,
New J. Phys. **20**, 033004 (2018)





Most of our published data are corrected this way

$$F_z = F_{\text{Coulomb}} \cos\phi = \frac{Qe}{4\pi\epsilon_0} \frac{1}{(r^2+z^2)} \frac{z}{\sqrt{r^2+z^2}}$$

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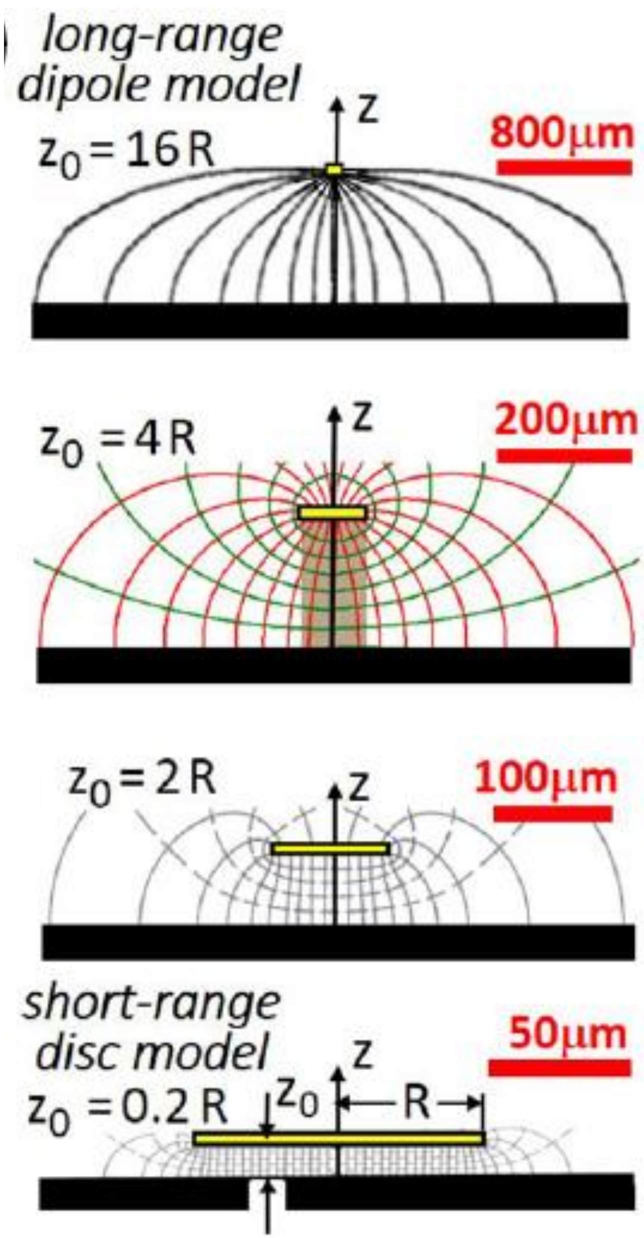
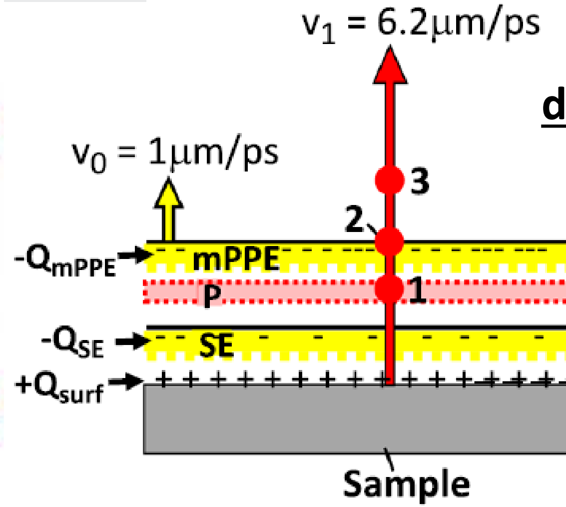
Refinement of the Oloff et al. space-charge model: **SHORT-RANGE REGIME**

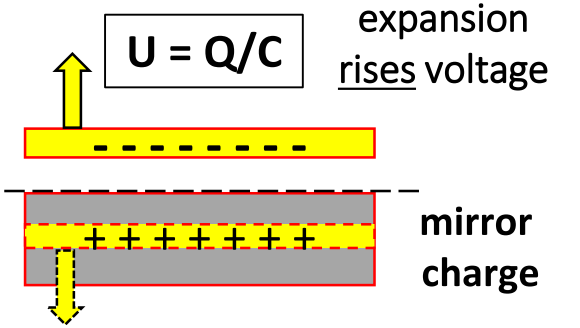


E_{kin} = 3 eV for mPPE and SE (yellow)
E_{kin} = 107 eV for photoelectrons (red).

Rev. Sci. Instrum. **92,** 053703 (2021)



disc model ("expanding capacitor")



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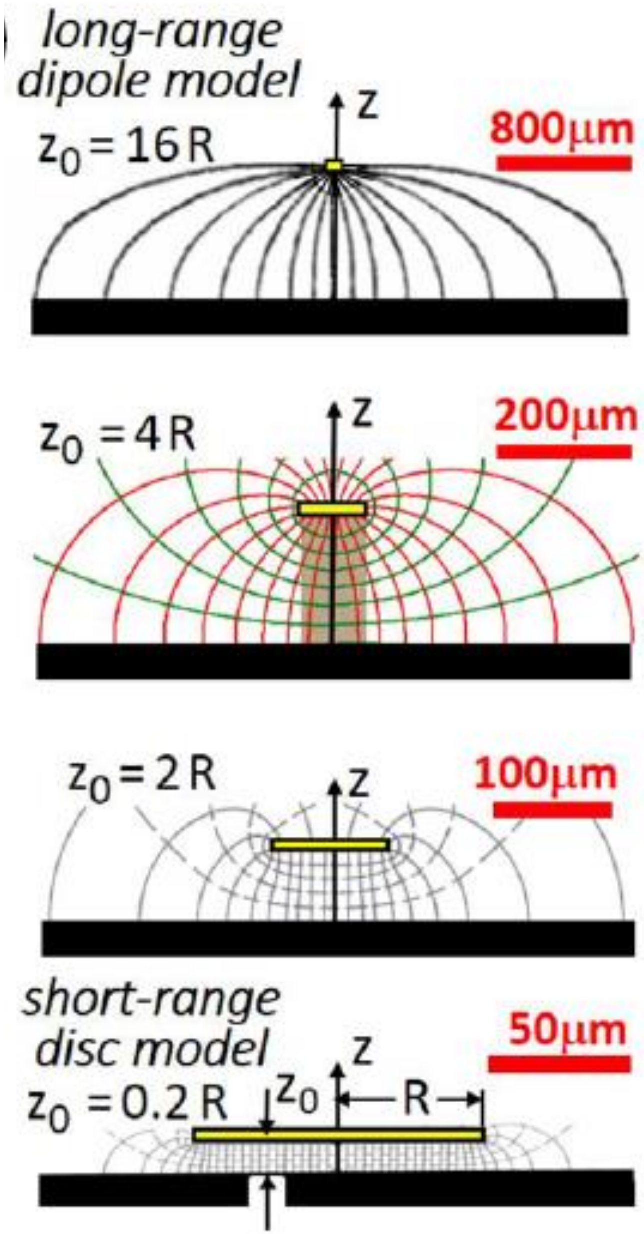
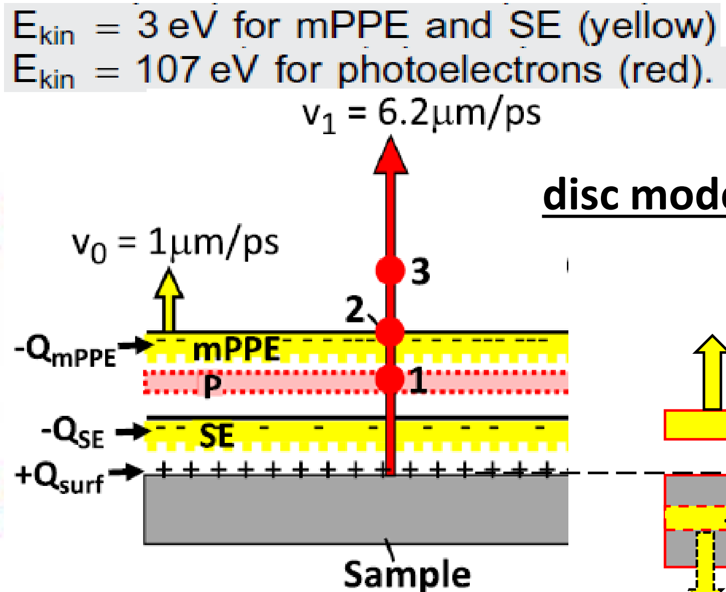
Rev. Sci. Instrum. **92,** 053703 (2021)





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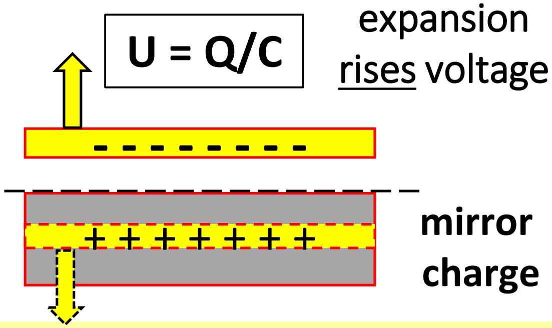
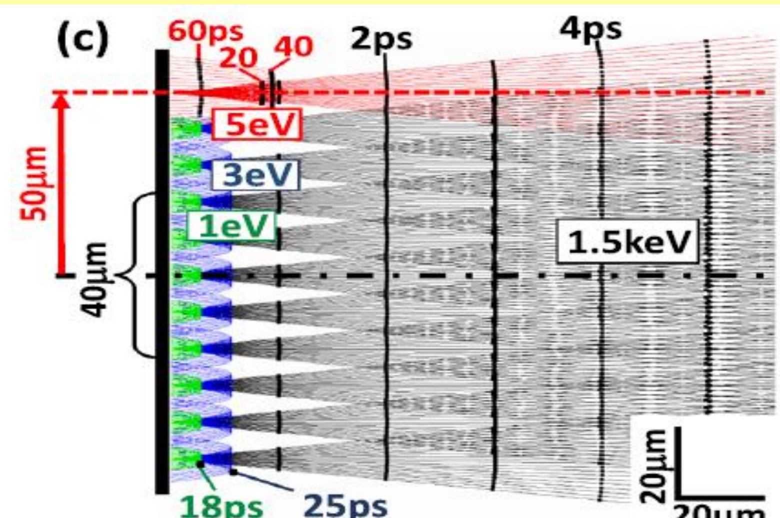


The main space-charge shift/broadening originates from the ***pushing effect of slow electrons close to the surface*.**

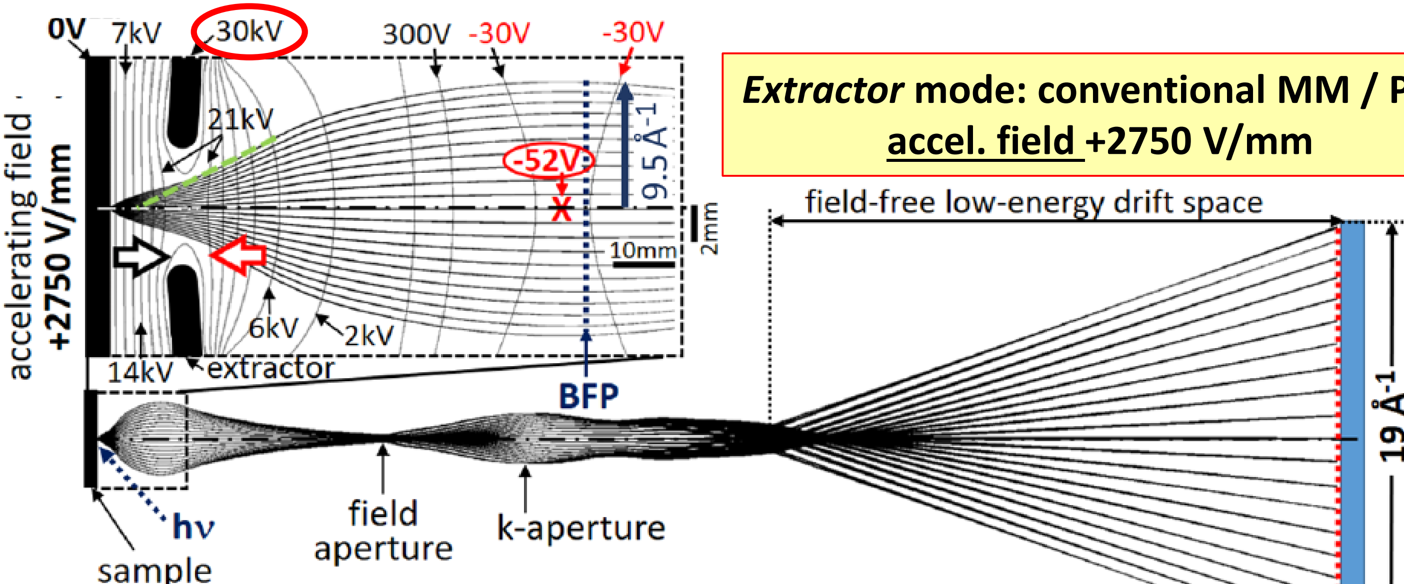
Is it possible to peel these electrons from the beam?



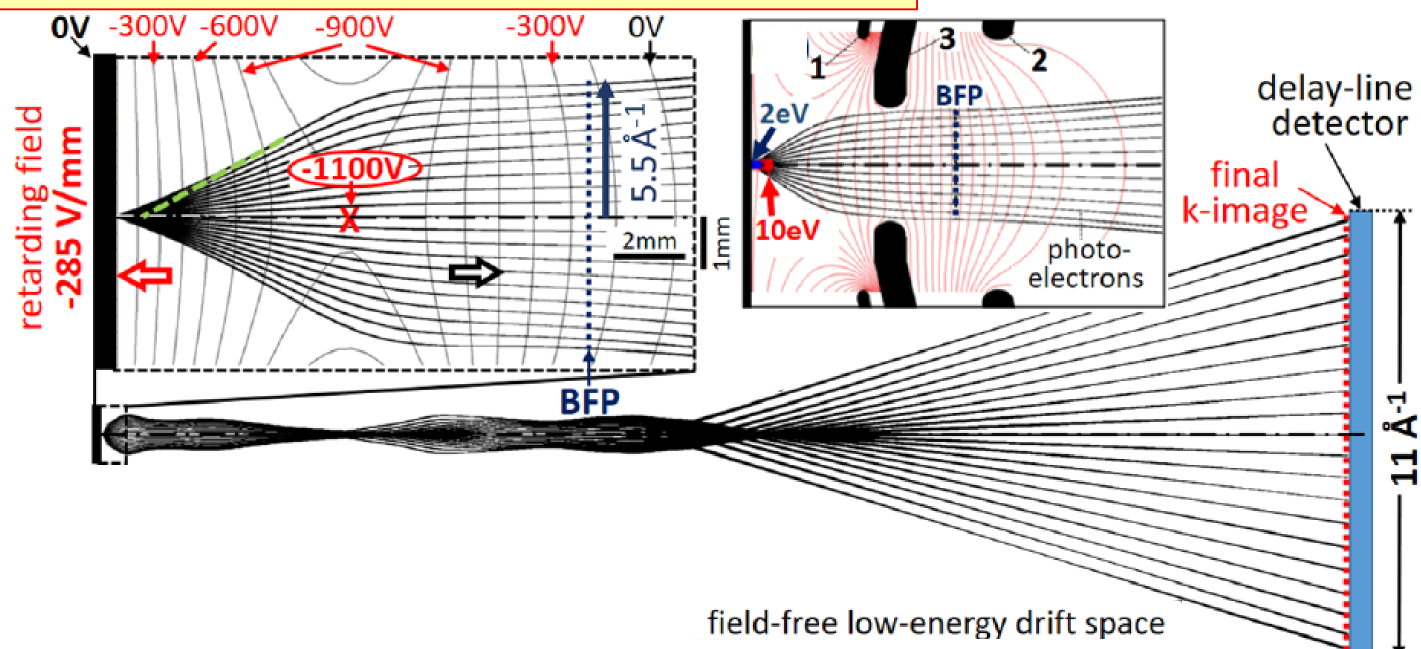
SUPPRESSION of space-charge effects by retarding front lens

***Extractor* mode: conventional MM / PEEM mode**
accel. field +2750 V/mm

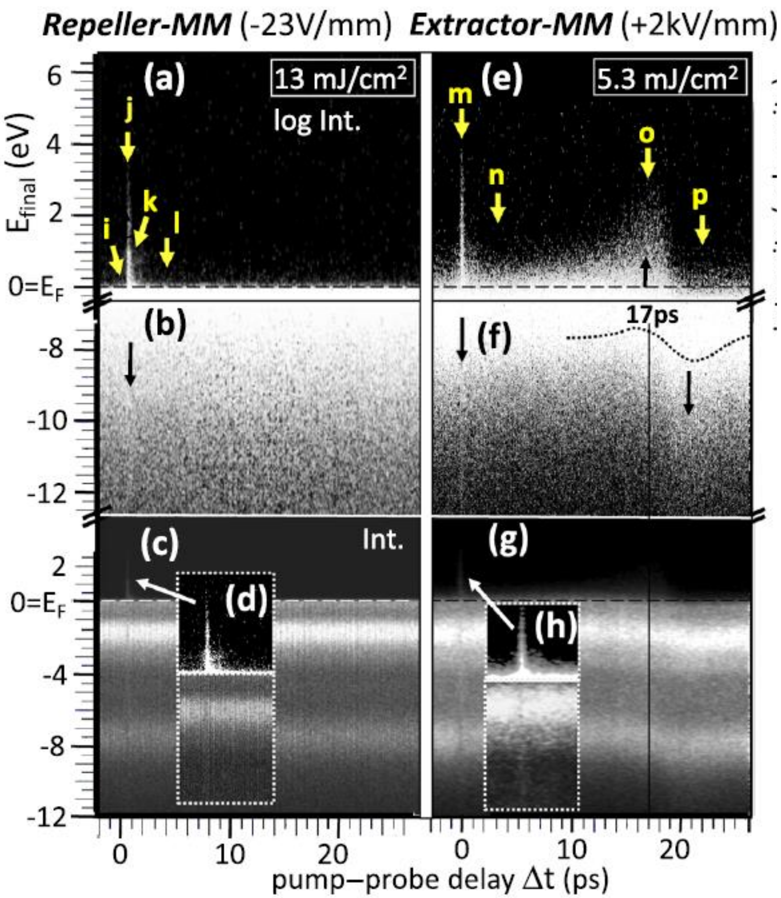
Rev. Sci. Instrum. **92,** 053703 (2021)



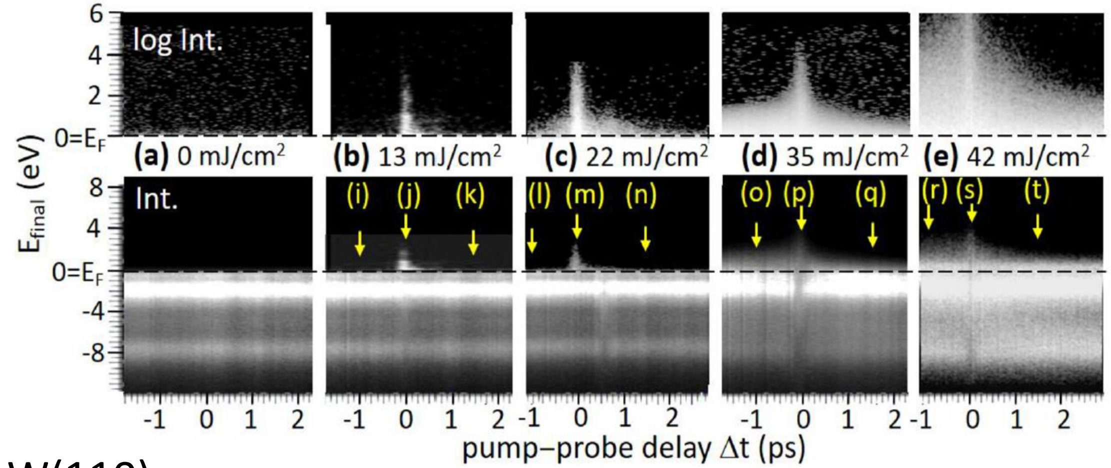
***Repeller* mode: strong retarding field at sample due to decelerating lens mode** **retarding field -285 V/mm**



SUPPRESSION of space-charge effects by retarding front lens: first experiments @ FLASH

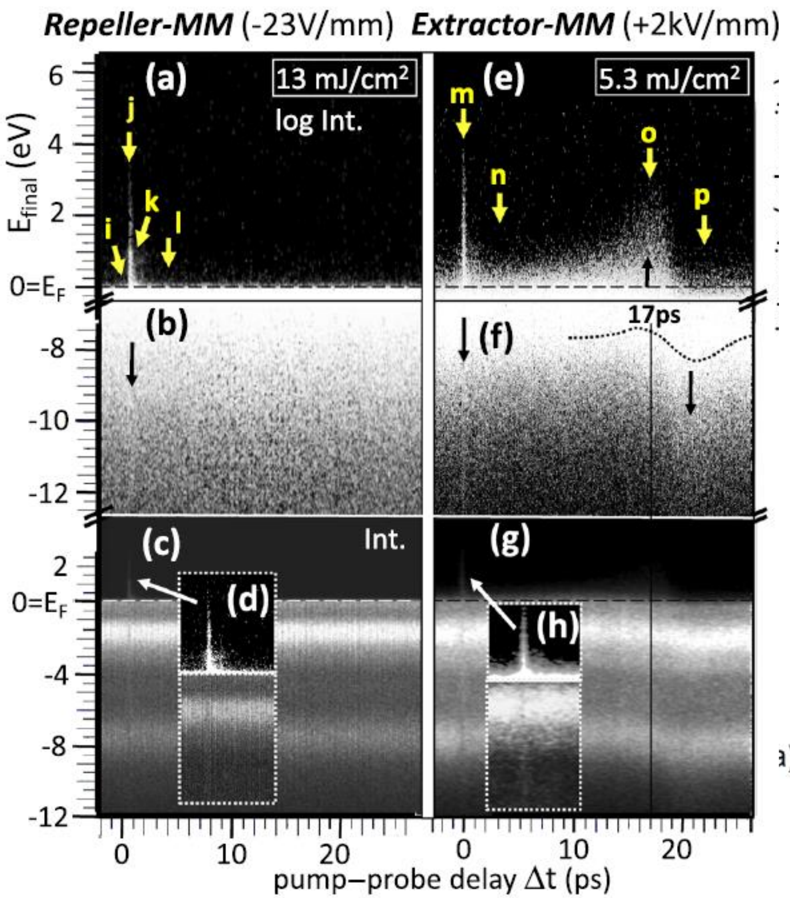


Rev. Sci. Instrum.
92, 053703 (2021)

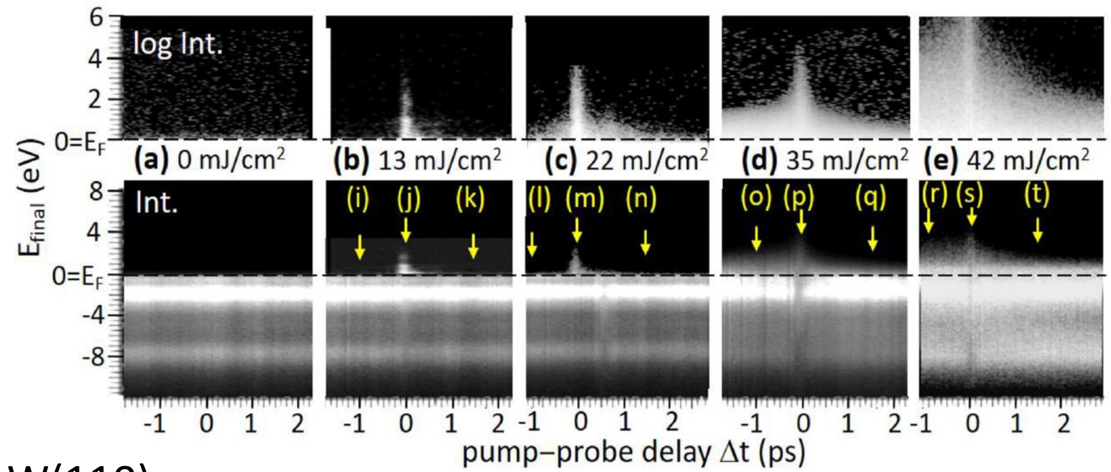


W(110)

SUPPRESSION of space-charge effects by retarding front lens: first experiments @ FLASH



Rev. Sci. Instrum. **92,** 053703 (2021)



F. Pressacco et al., *Nat. Commun.* **12**, 5088 (2021)

W(110)

Subpicosecond metamagnetic phase transition in FeRh driven by non-equilibrium electron dynamics

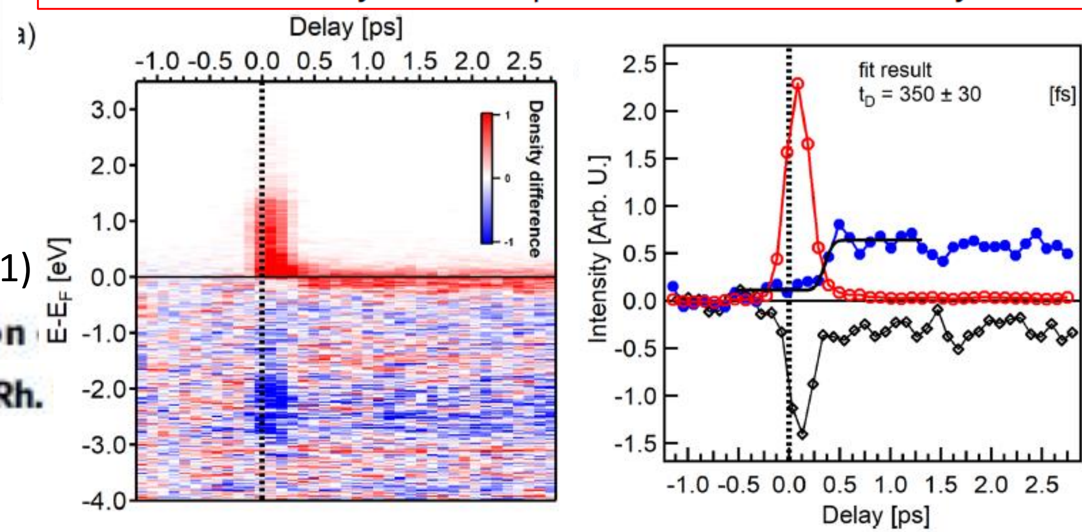


Fig. 4 Subpicosecond generation of the electronic FM order in FeRh.

Federicos talk tomorrow
MM @ PETRA & FLASH

SUPPRESSION of space-charge effects by retarding front lens: first experiments @ FLASH

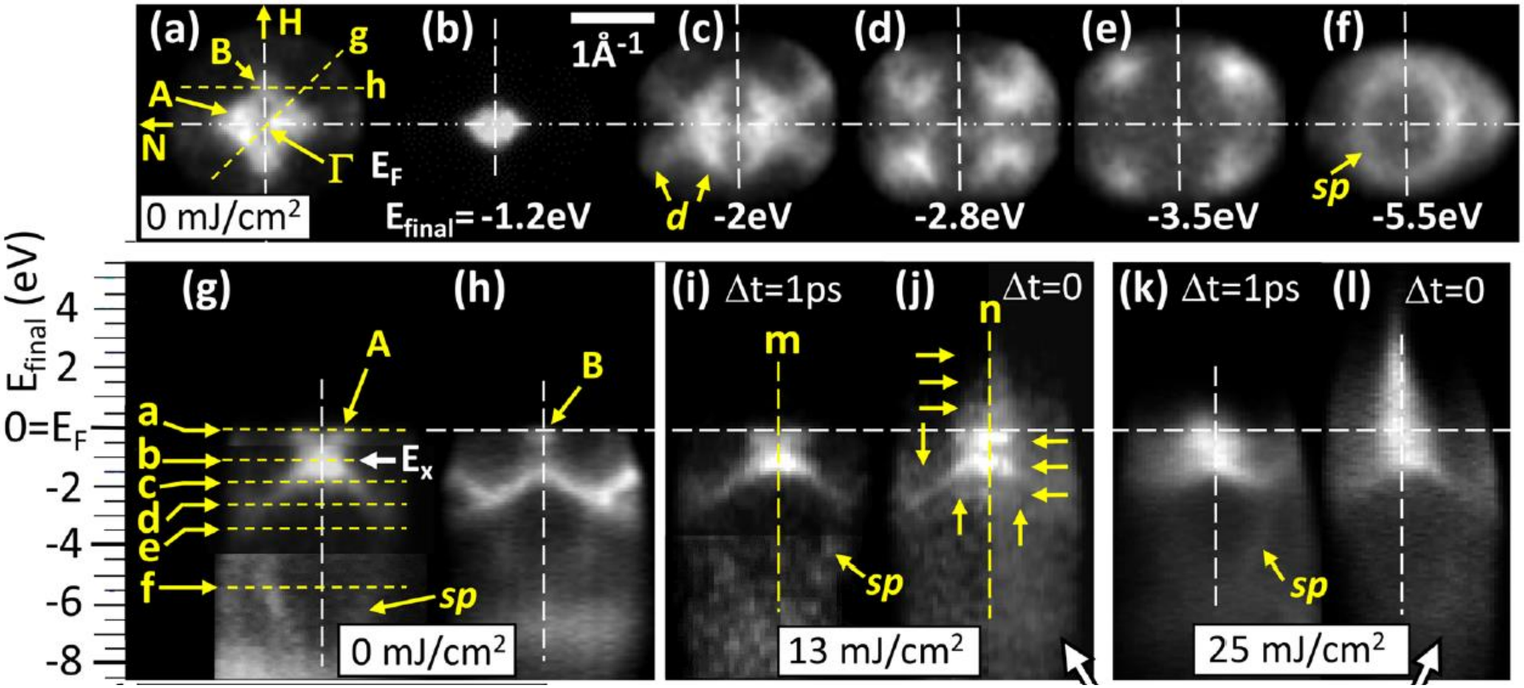


FIG. 7. Sections through the 4D (k_x, k_y, E_{final}, Δt) data arrays in the valence-band range of W(110) recorded at room temperature in the *repeller-MM* mode (field F = −21 V/mm) with $h\nu_{pump}$ = 1.2 eV (p-polarized) and $h\nu_{probe}$ = 111.6 eV. (a)–(f) k_x–k_y sections at different final-state energies as given in the panels and marked in

W(110)

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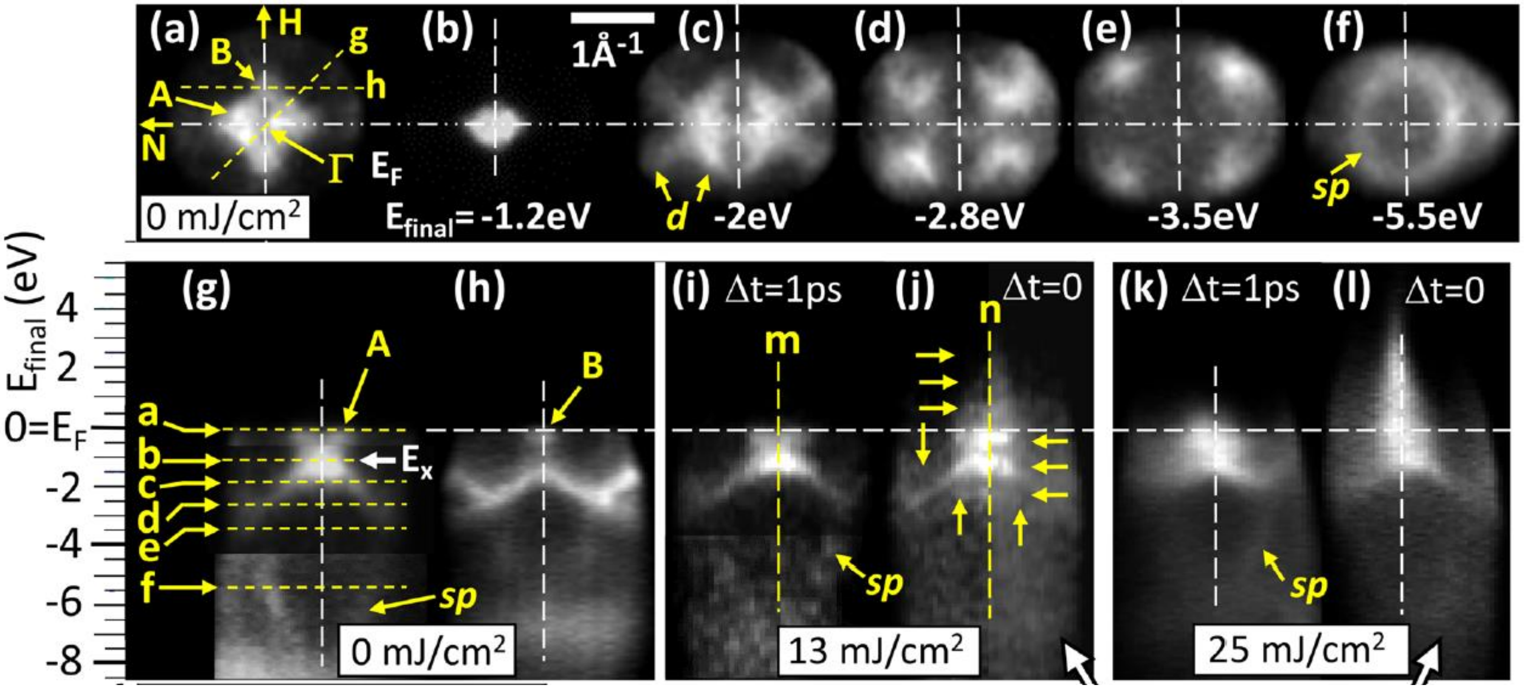
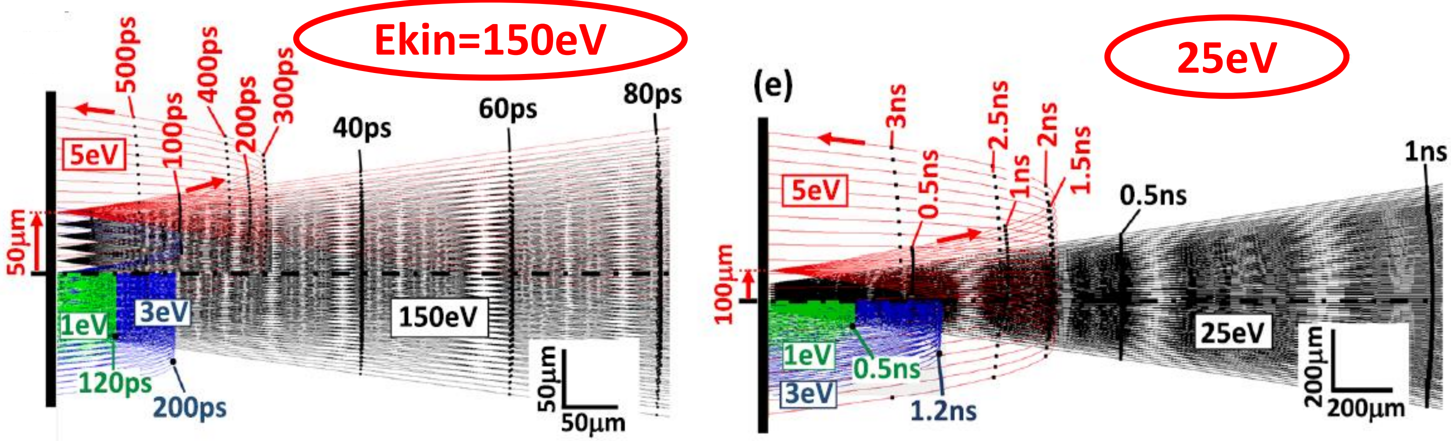


Rev. Sci. Instrum. **92,** 053703 (2021)

FIG. 7. Sections through the 4D (k_x, k_y, E_{final}, Δt) data arrays in the valence-band range of W(110) recorded at room temperature in the *repeller-MM* mode (field F = −21 V/mm) with $h\nu_{pump}$ = 1.2 eV (p-polarized) and $h\nu_{probe}$ = 111.6 eV. (a)–(f) k_x–k_y sections at different final-state energies as given in the panels and marked in



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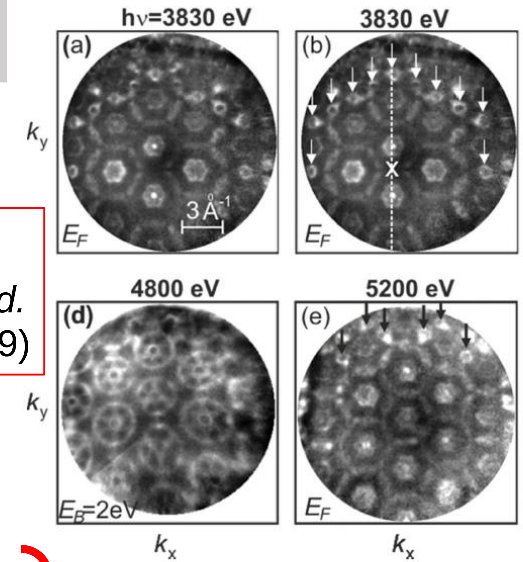
Kai's talk, Philip's talk this session, several talks tomorrow **MM @ PETRA & FLASH**

Spin filter

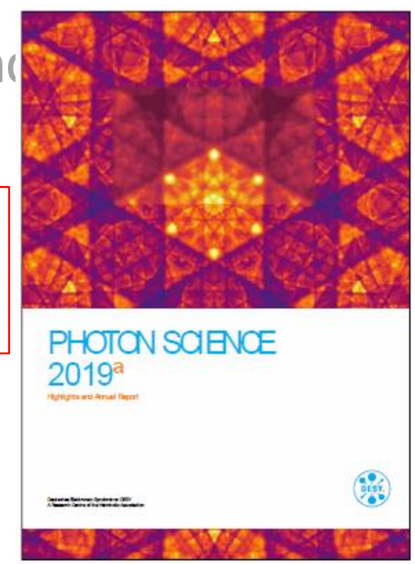
- Progress in spin filtering (high energies, larger energy band
- First fs time resolved spin measurements (Gort)

19 BZs



Re(0001)

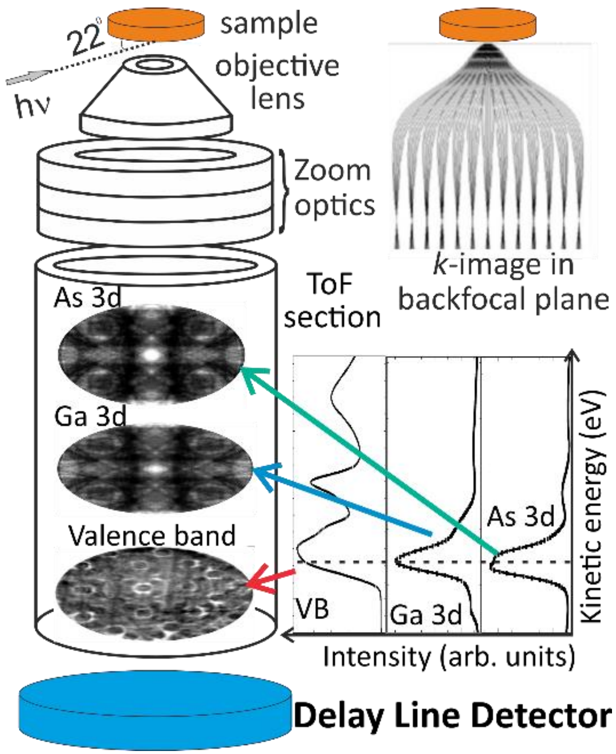
J. Synchr. Rad. **26,** 1996 (2019)



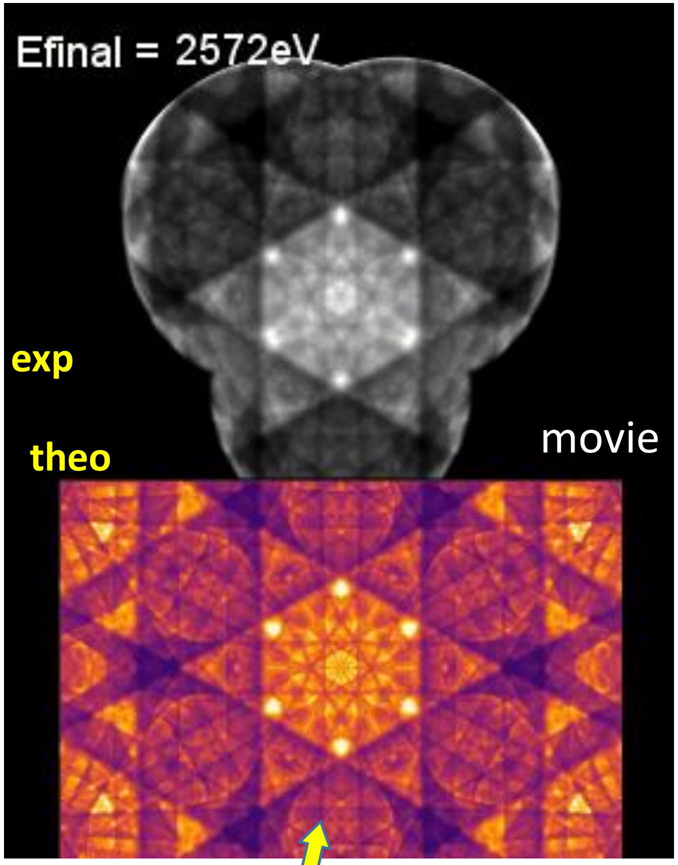
Graphite hXPD

New J. Phys. **21**, 113031 (2019)

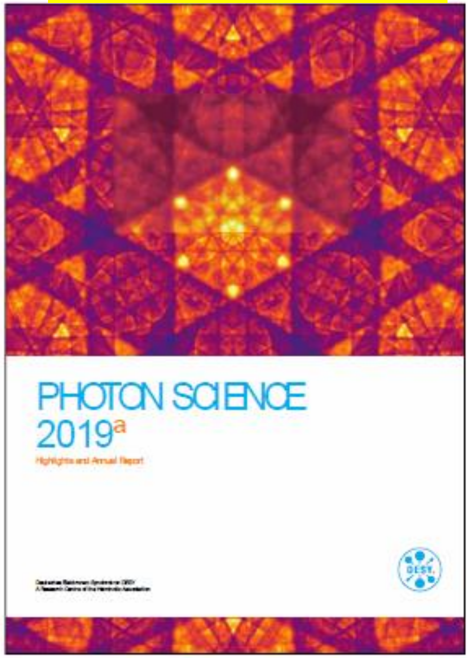
Full-field imaging hXPD



XPD graphite



DESY Highlights 2019

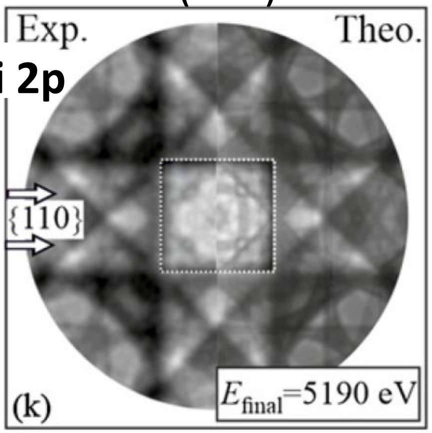


Bloch wave calculation

O. Fedchenko et al., *New J. of Phys.* **21**, 113031 (2019)

Si (100)

Si 2p



O. Fedchenko et al., *New J. of Phys.* **22**, 103002 (2020)

$G_{220} = 3.27\ \text{Å}^{-1}.$

Outline

Introduction

- Multidimensional aspect
- ToF-MM *vs* conventional ARPES

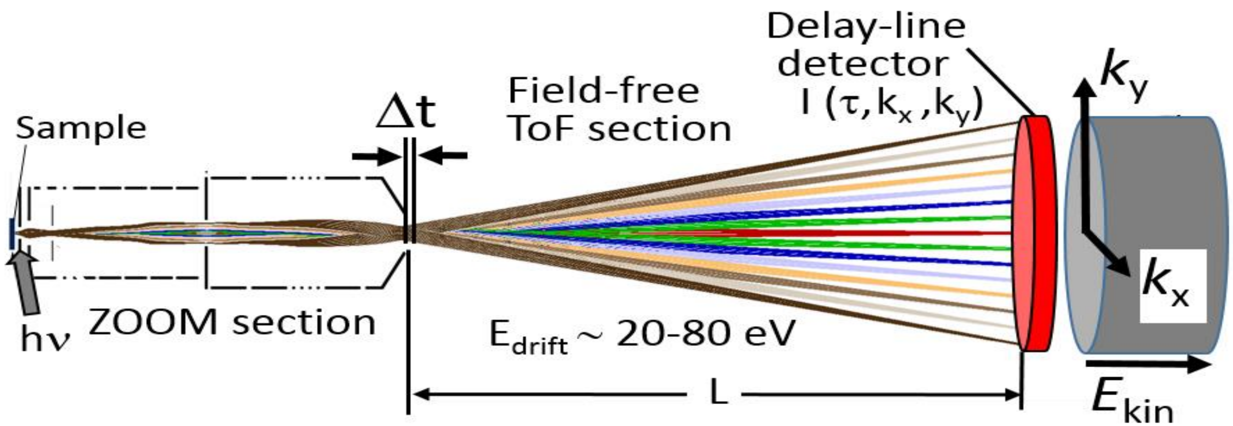
Instrument evolution

- Space-charge correction / suppression in ToF-MM
- Valence-band mapping → electronic structure
- Photoelectron diffraction XPD → geometric structure
- PEEM mode → 'sub-micron ARPES'

Spin filter

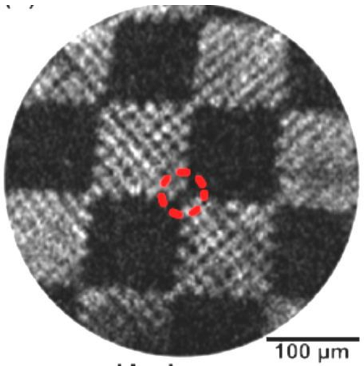
- Progress in spin filtering (high energies, larger energy band)
- First fs time resolved spin measurements (Gort)

ToF MM key features: **PEEM mode**

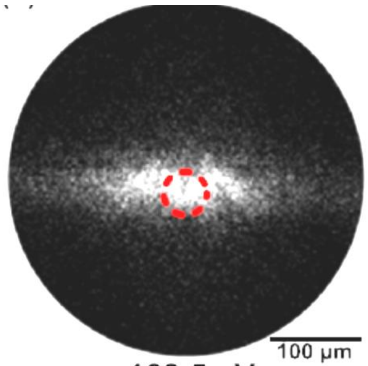


Real-Space (PEEM) images:

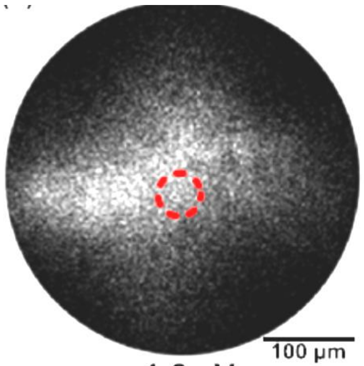
scale calibration | **footprint FEL beam** | **pump laser** | **freshly cleaved SmB_6**



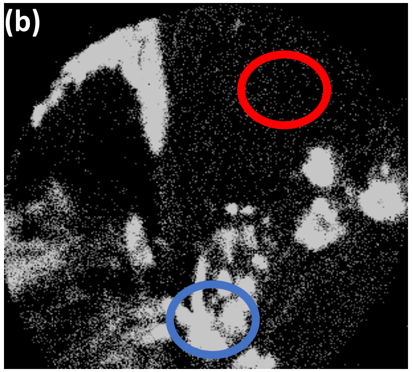
Hg lamp



109.5 eV



1.6 eV



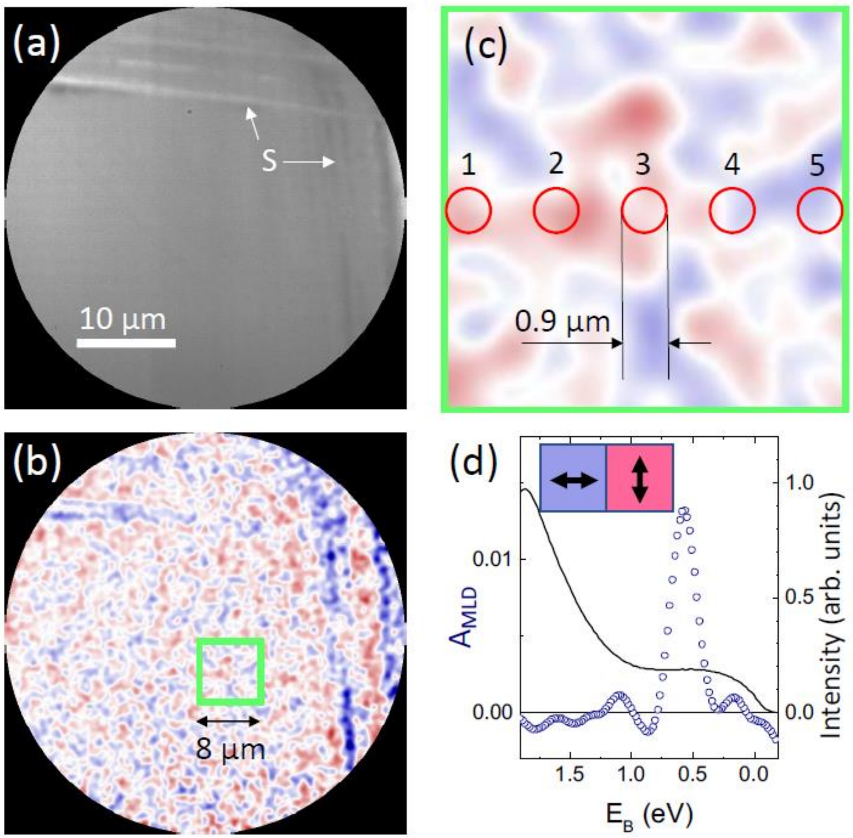
Dmitro Kutnyakhov et al.,
Rev. Sci. Instrum. **91,** 013109 (2020)

ToF MM key features: ‘**Sub-micron ARPES**’

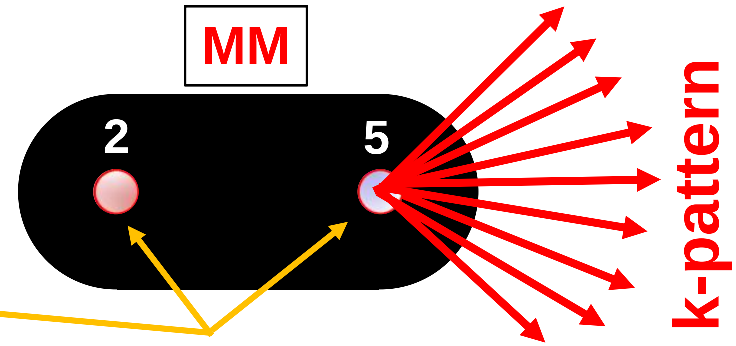
Example: Resolving micron-sized antiferromagnetic domains in Mn_2Au

PEEM

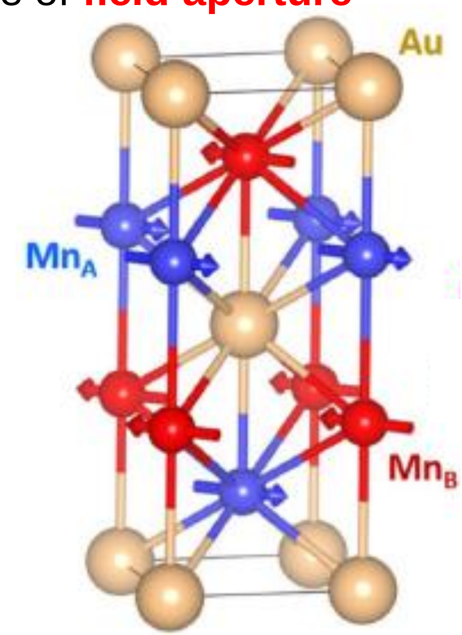
MLD contrast



MM



Positions and size of **field aperture**



Direct Observation of Antiferromagnetic Parity Violation in the Electronic Structure of Mn_2Au

O. Fedchenko, L. Smejkal et al., arXiv 2110.12186v1 (2021)

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Spin filter

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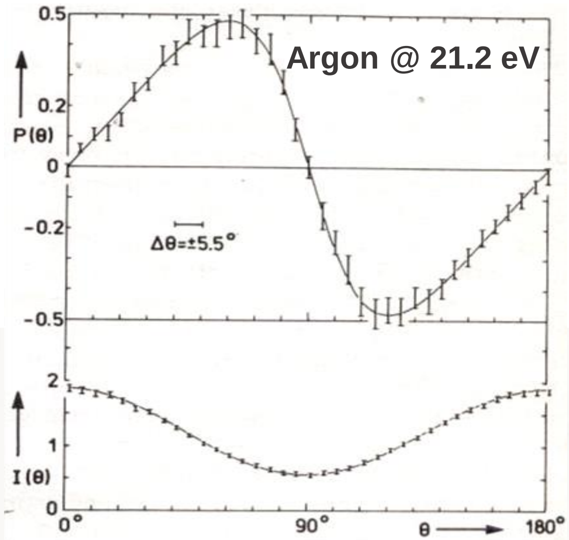
Spin-resolved experiments 4 decades ago

1980

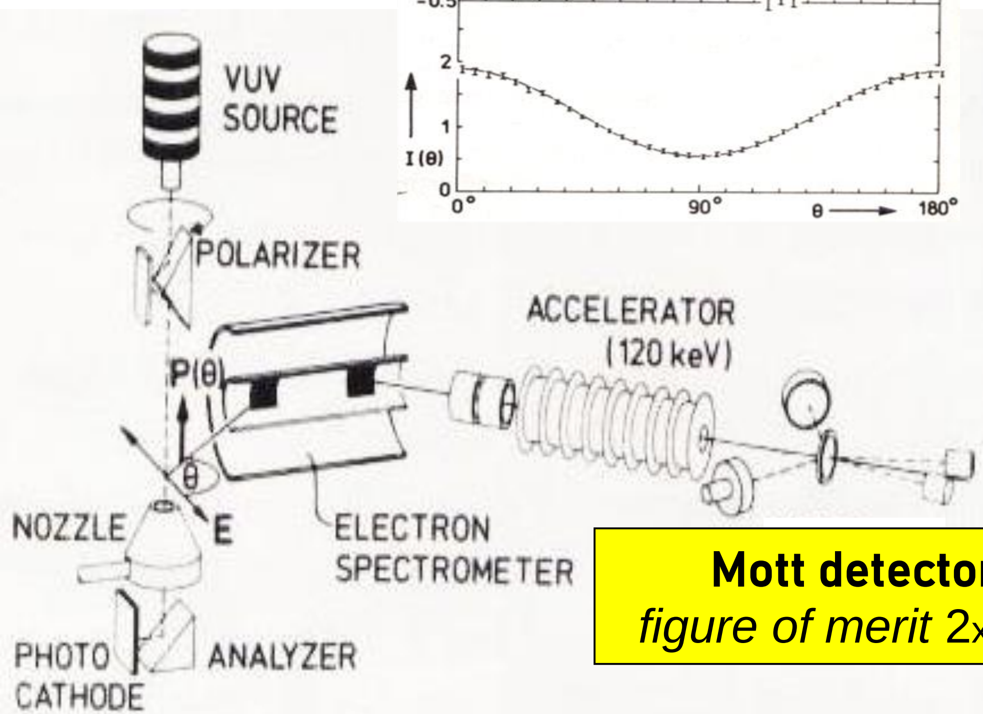
44, NUMBER 10 PHYSICAL REVIEW LETTERS

Angular Dependence of the Polarization of Photoelectrons Ejected by Plane-Polarized Radiation from Argon and Xenon Atoms

G. Schönhense
Physikalisches Institut der Universität Münster, D-4400 Münster, Germany
(Received 31 December 1979)



several weeks…



Mott detector
figure of merit 2x10^{-4}

1982

VOLUME 48, NUMBER 9 PHYSICAL REVIEW LETTERS

Photoelectron Polarization in Hg $6s^2$ Subshell Ionization with Unpolarized Light: New Aspect of the Fano Effect

G. Schönhense and U. Heinzmann
Fritz-Haber-Institut der Max-Planck-Gesellschaft, D-1000 Berlin 33, West Germany
J. Kessler
Physikalisches Institut der Universität Münster, D-4400 Münster, West Germany
N. A. Cherepkov
A. F. Ioffe Physical-Technical Institute, 194021 Leningrad, U. S. S. R.

Just 3 spin-polarization points,
3 weeks of accumulation time with the Mott

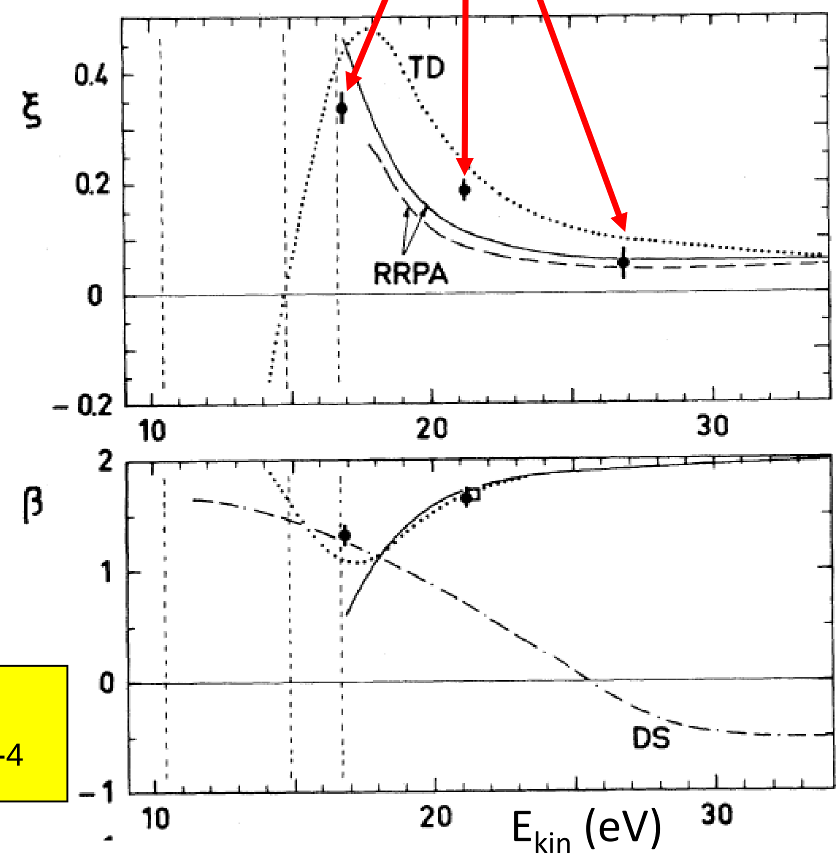
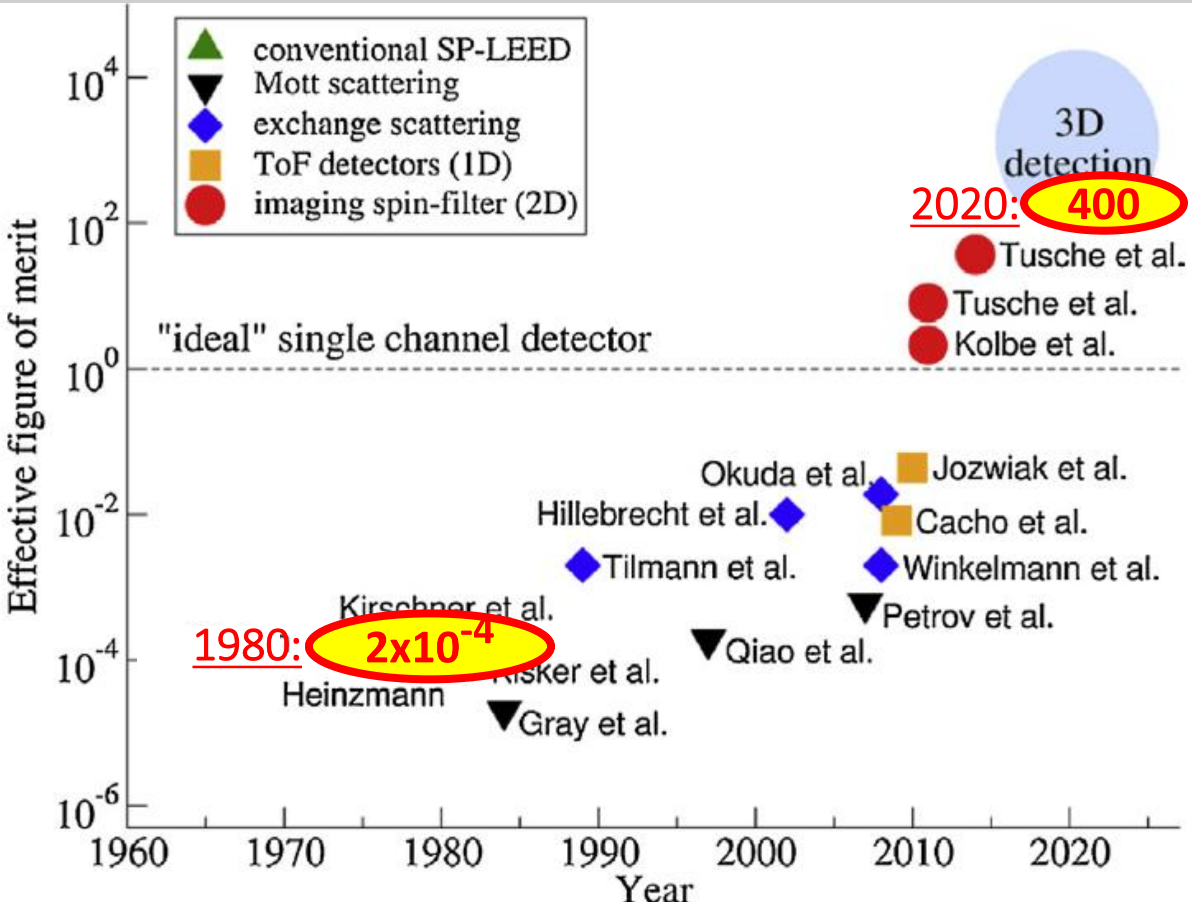


Figure of merit of spin detectors



S. Suga & C. Tusche, JESRP **200**, 119 (2015)

M. Kolbe

D. Kutnyakhov

S. Chernov

Mainz

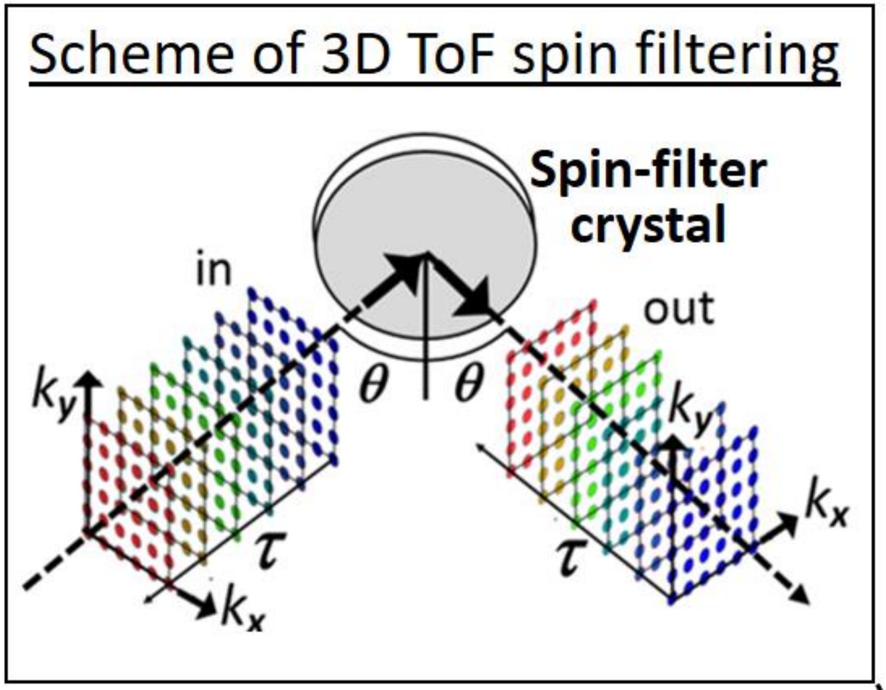
Halle

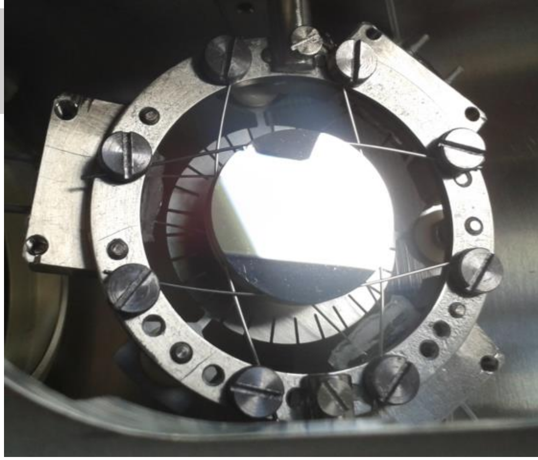
J. Kirschner

C. Tusche

D. Vasilyev

ToF MM key features: **Spin filter**





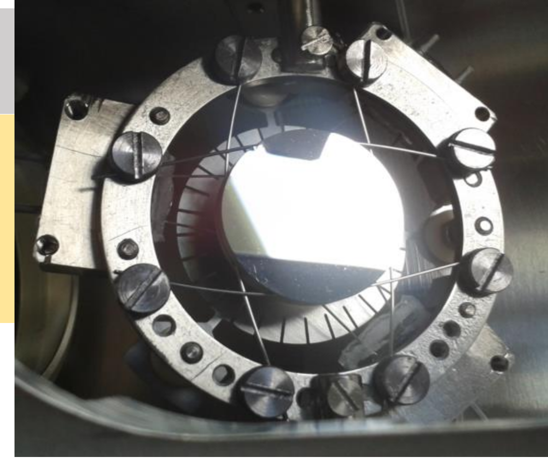
W(100) at 45° & 27eV
$FoM_{single} = 5 \times 10^{-3}$

k-resolution: 0.03 $Å^{-1}$
E-resolution: 20 meV

k-disk at E_F:
N ≈ 1700 (k_x,k_y)-pixels

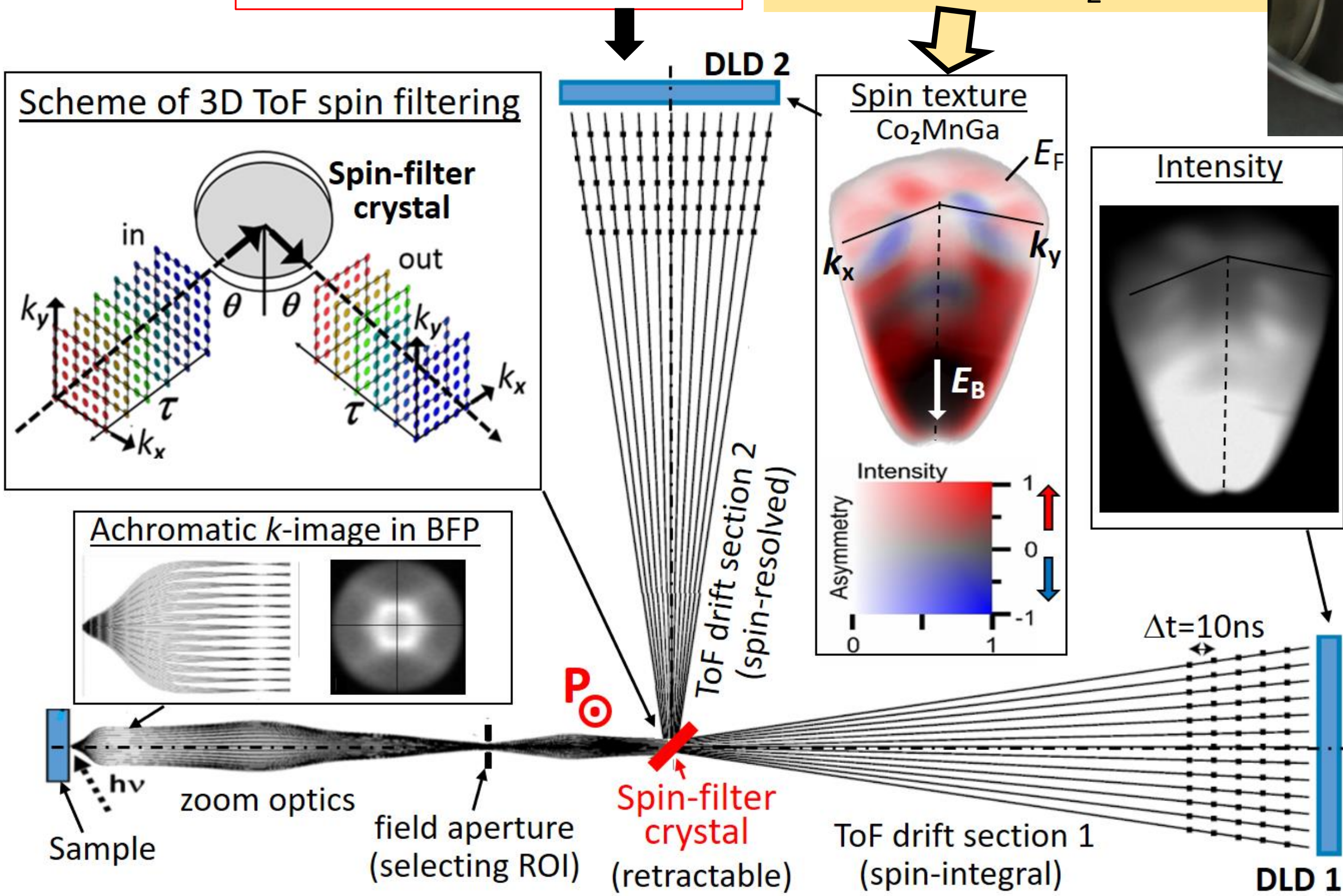
N > 10^5 resolved
(E_B,k_x,k_y)-voxels
FoM_{eff} ≈ 400

ToF MM key features: **Spin filter**



Second ToF branch captures spin-filtered image

0.5 hours: Full VB spin texture of Heusler compound Co_2MnGa

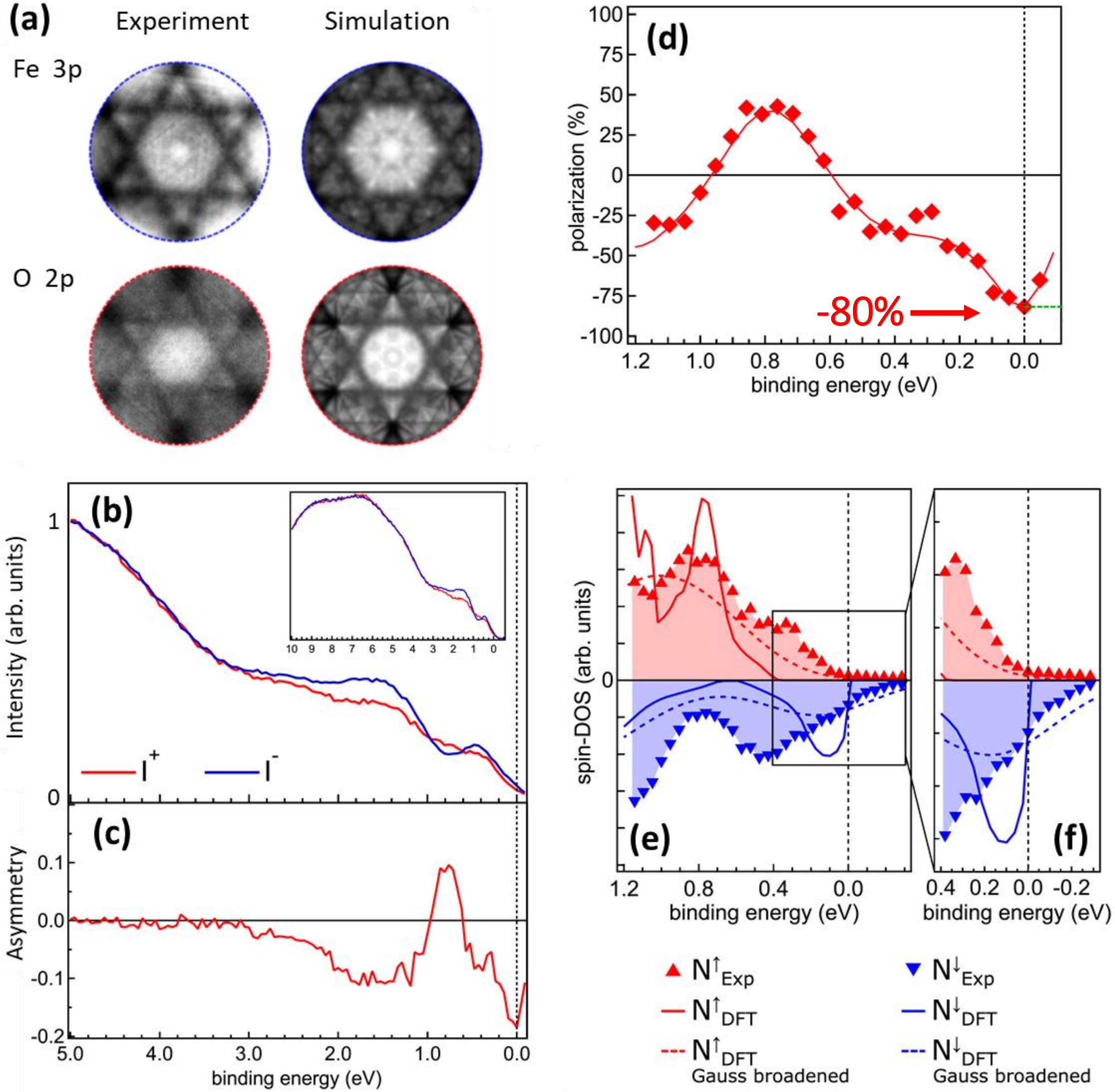


N > 10^5 resolved (E_B,k_x,k_y)-voxels

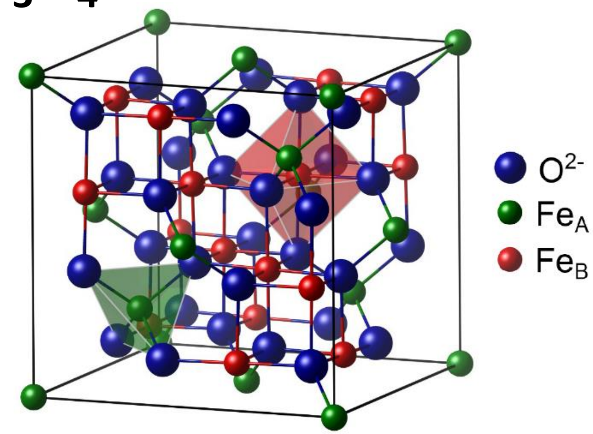
$FoM_{eff} \approx 400$

Sergey Chernov et al., *Phys. Rev*. B **103**, 054407 (2021)

ToF-MM & spin filter: True bulk spin polarization of Fe_3O_4



Fe_3O_4



hν = 5 keV

true bulk spin polarization

M. Schmitt et al., *Phys. Rev. B* **104**, 045129 (2021)

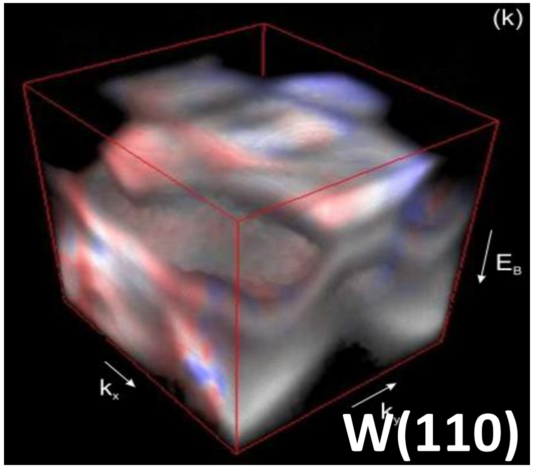
Cooperation with Würzburg University

-80% at E_F points on half-metallic ferromagnet

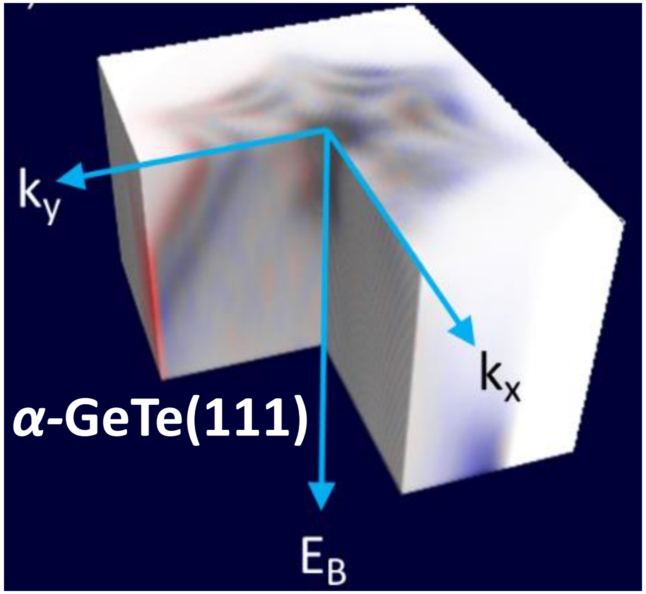
ToF MM key features: **Spin filter**

Spin texture of surface states

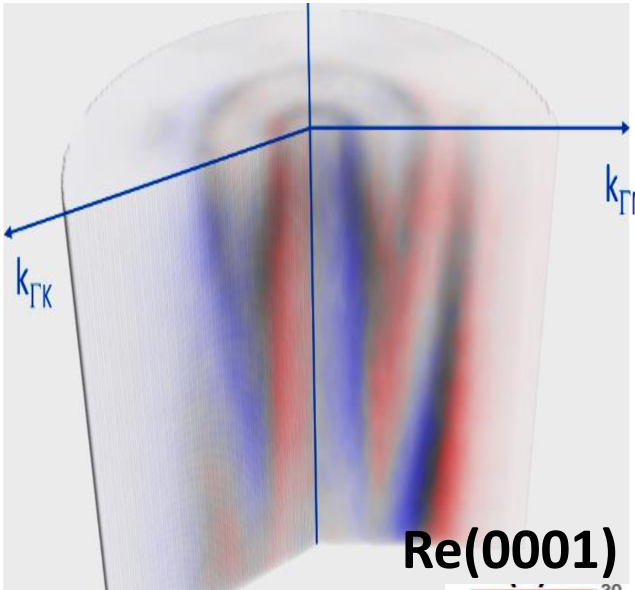
BESSY II
10m NIM



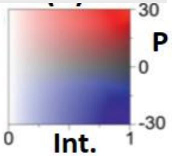
Time-reversal invariant helical Dirac state in projected bulk band gap



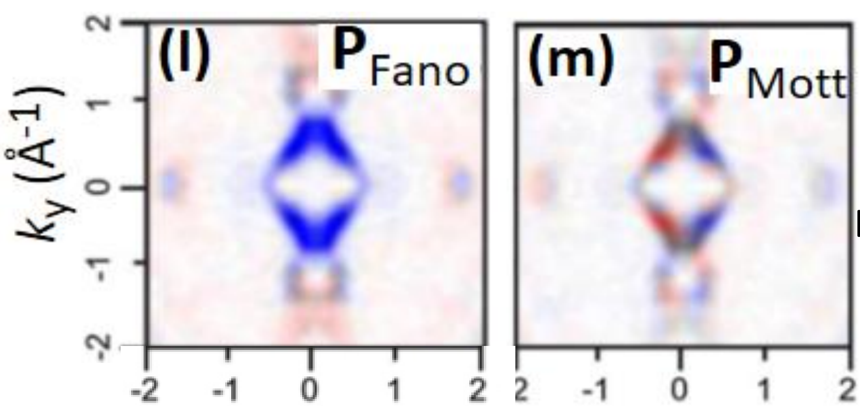
Bulk Rashba semiconductor



Tamm state with Rashba signature



Two spin components (Fano and Mott) of bulk bands



PETRA-III P04
hν=400eV
RCP, LCP

D. Vasilyev et al.,
⟵*J. Phys.* C **32**,
135501 (2020)

Spin- and time-resolved photoelectron spectroscopy and diffraction studies using time-of-flight momentum microscopes

JVST A REVIEW
Journal of Vacuum Science & Technology A

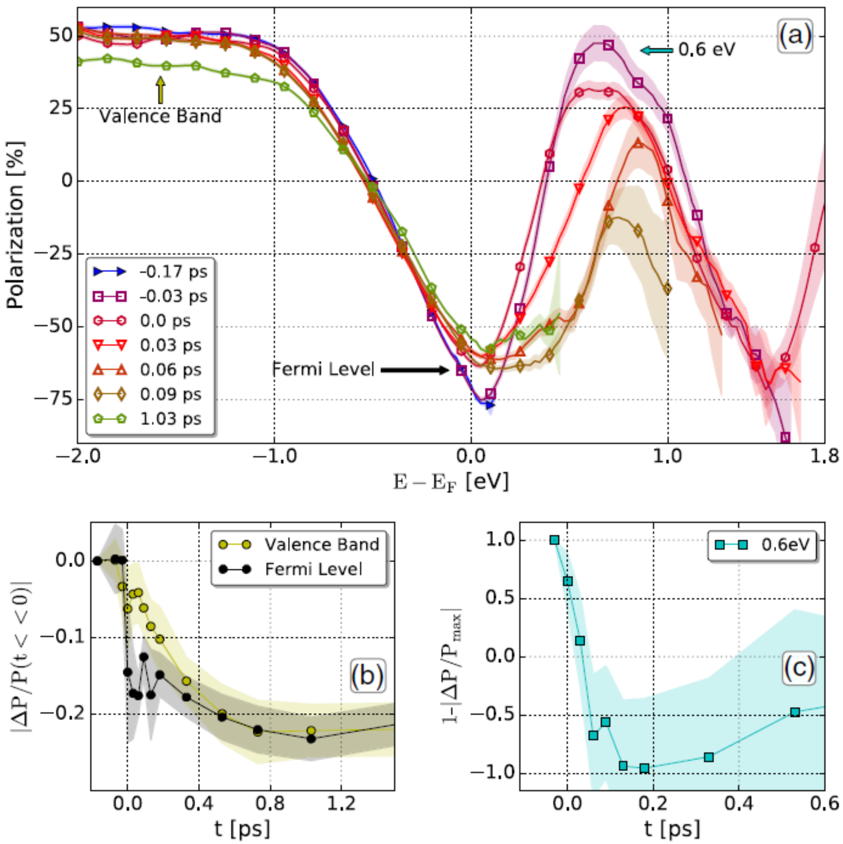
J. Vac. Sci. Technol. A **40**, (2022);
doi: 10.1116/6.0001500

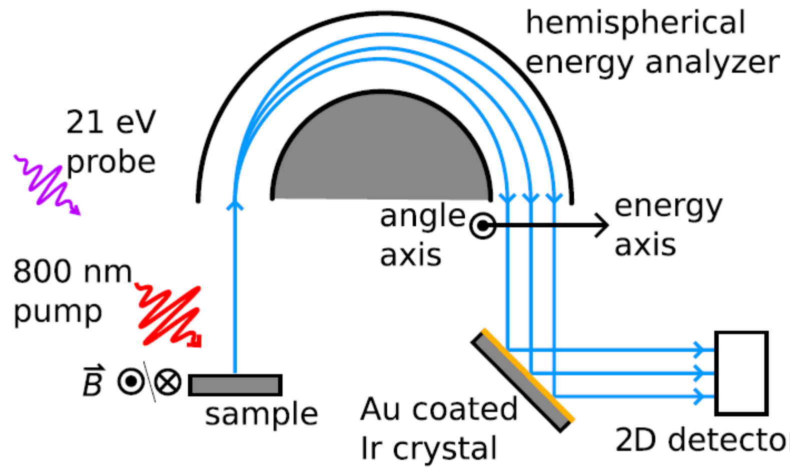
First fs time-resolved Spin-ARPES

PHYSICAL REVIEW LETTERS **121**, 087206 (2018)

Early Stages of Ultrafast Spin Dynamics in a 3*d* Ferromagnet

R. Gort,[1*] K. Bühlmann,[1] S. Däster,[1] G. Salvatella,[1] N. Hartmann,[2] Y. Zemp,[1] S. Holenstein,[3,4] C. Stieger,[5] A. Fognini,[6] T. U. Michlmayr,[1] T. Bähler,[1] A. Vaterlaus,[1] and Y. Acremann[1]





Compact setup for spin-, time-, and angle-resolved photoemission spectroscopy

K. Bühlmann et al.,
Rev. Sci. Instrum.
91, 063001 (2020)

Zürich group

k-ToF's in fs photoemission

FELs:
HEXTOF @FLASH
HEXTOF 2.0
Eu XFEL
LCLS-II

HHG sources:
8 in operation more to follow

HEXTOF @ FLASH

D. Kutnyakhov et al., ***Time- and momentum-resolved photoemission studies using time-of-flight momentum microscopy at a free-electron laser,*** Rev. Sci. Instrum. **91**, 013109 (2020); doi: 10.1063/1.5118777

M. Dendzik et al. Phys. Rev. Lett. **125**, 096401 (2020)

F. Pressacco et al., ***Subpicosecond metamagnetic phase transition in FeRh driven by non-equilibrium electron dynamics***, Nat. Commun. **12**, 5088 (2021); doi: 10.1038/s41467-021-25347-3

D. Curcio et al., ***Ultrafast electronic linewidth broadening in the C 1s core level of graphene***, Phys. Rev. B **104**, L151104 (2021); doi: 10.1103/PhysRevB.104.L161104

HHG setups

S. Beaulieu et al., ***Revealing Hidden Orbital Pseudospin Texture with Time-Reversal Dichroism in Photo-electron Angular Distributions***, Phys. Rev. Lett. **125**, 216404 (2020) [*Fritz-Haber group*]

S. Beaulieu et al., ***Ultrafast dynamical Lifshitz transition***, Science Adv. **7**, eabd9275 (2020) [*Fritz-Haber group*]

S. Beaulieu et al., M***Unveiling the Orbital Texture of 1T-TiTe$_2$ using Intrinsic Linear Dichroism in Multidimensional Photoemission Spectroscopy***, npj Quantum Materials **6**, 93 (2021); doi: 10.1038/s41535-021-00398-3 [*Fritz-Haber group & Bordeaux group*]

M. Keunecke et al., ***Electromagnetic dressing of the electron energy spectrum of Au(111) at high momenta***, Phys. Rev. B **102**, 161403 R (2020) [*Göttingen group*]

J. Madéo et al., ***Directly visualizing the momentum-forbidden dark excitons and their dynamics in atomically thin semiconductors***, Science **370**, 1199–1204 (2020) [*Okinawa group*]

M. K. L. Man et al., ***Experimental measurement of the intrinsic excitonic wave function,*** Science Adv. **7**, eabg0192 (2021) [*Okinawa group*]

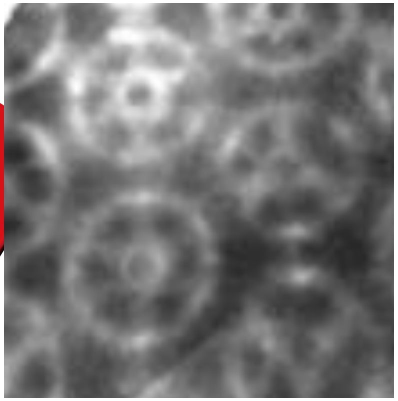
R. Wallauer et al., ***Tracing orbital images on ultrafast time scales***, Science **371**, 1056 (2021) [*Marburg group*]

R. Wallauer et al., ***Momentum-Resolved Observation of Exciton Formation Dynamics in Monolayer WS$_2$***, Nano Letters (2021); doi.10.1021/acs.nanolett. 1c01839 [*Marburg group & Regensburg group*]

….

Funding:
BMBF (several projects since 2014)
DFG through SFB TRR 173 „Spin+X“

City Arms of
Mainz am Rhein



Rhenium
(from lat. Rhenus)

Use a ToF-MM,
set $h\nu = 4400$eV,
look at $E_B = 2.0$eV,
and see the message