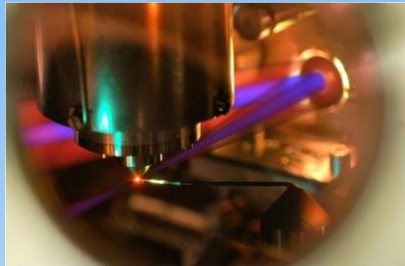


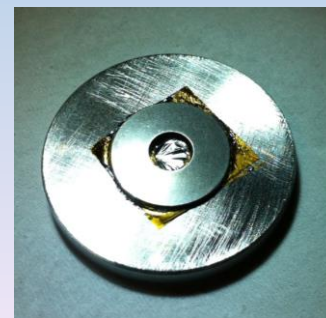
Optimization and application of attosecond multilayers



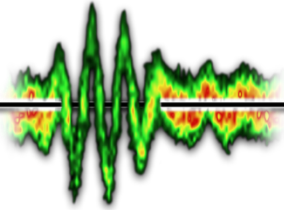
Alexander Guggenmos



Ludwig-Maximilians-University Munich, Faculty of physics, Garching, Germany
Max-Planck-Institute of Quantum Optics, Garching, Germany

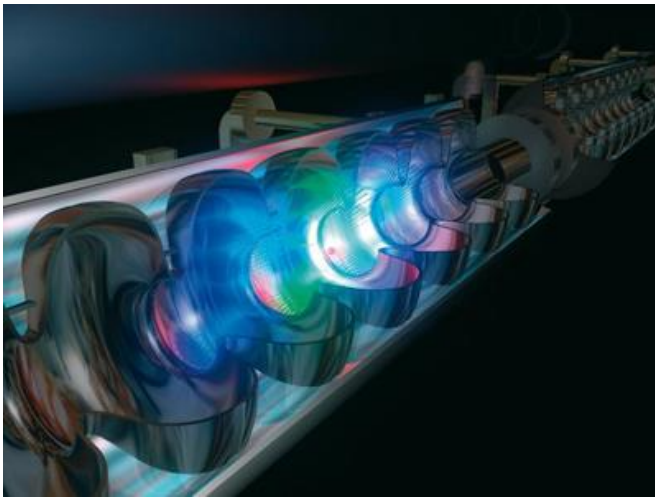


Motivation



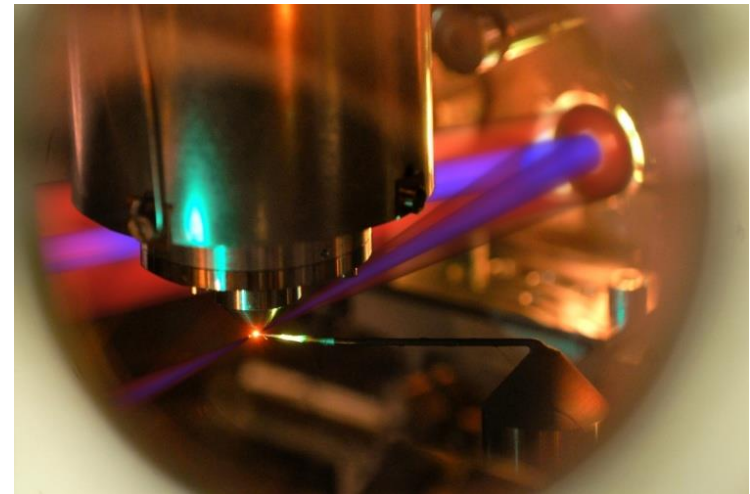
Excellent XUV/soft x-ray optics for sources emitting ultra short pulses:

(X)FEL/LCLS



grazing optics due to high intensities
High intensities in the
200 eV – keV range
Goal: shorter pulses (fs→as)

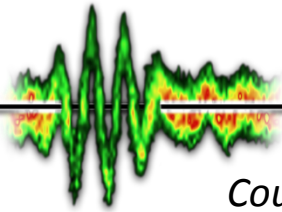
OUR FOCUS: HHG



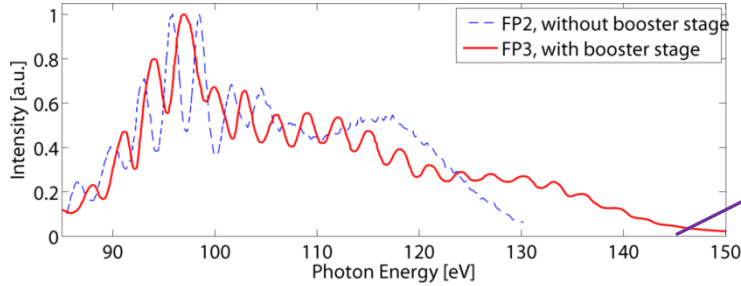
normal incidence optics possible
as pulses (requires large ΔE)
High intensities only < 200 eV
Goal: higher energies
Photon flux essential!

Both require optics for spectral filtering, phase shaping, ...

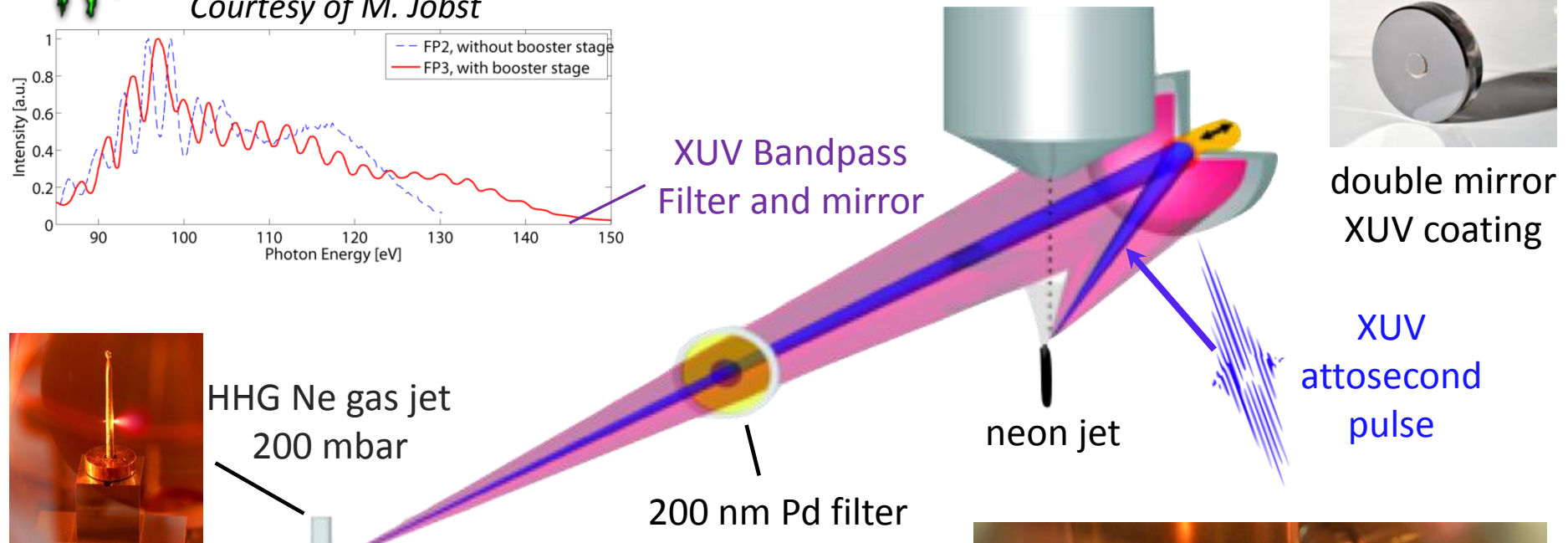
Attosecond electron streaking



Courtesy of M. Jobst



TOF electron spectrometer



HHG Ne gas jet
200 mbar

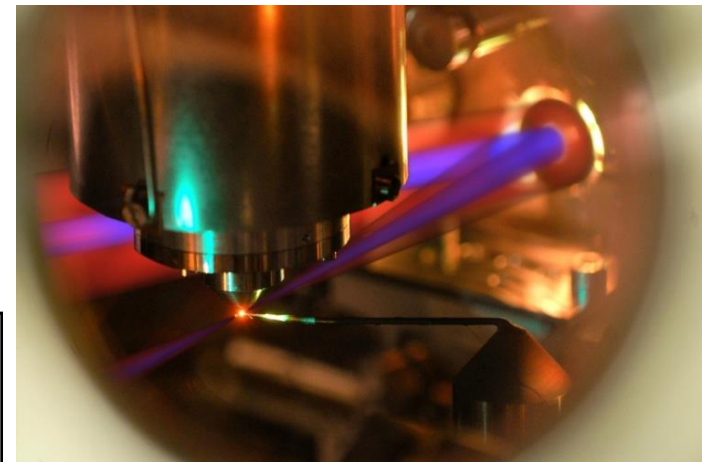
200 nm Pd filter

neon jet

double mirror
XUV coating

XUV
attosecond
pulse

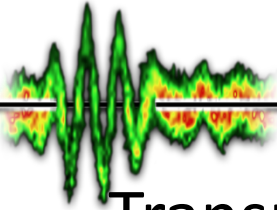
broadband, CEP stabilized
1.5 cycle NIR pulse
<math><4\text{ fs}</math>, 1.5 mJ, f=40 cm



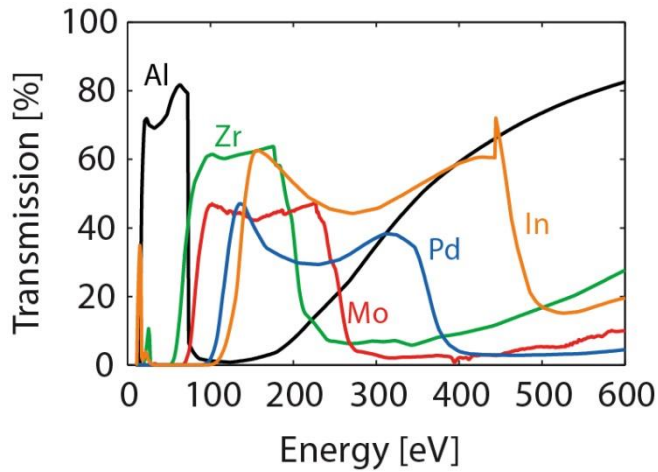
key component: optics

- to **delay** the XUV in respect to the laser pulse
- to **focus** the beams into the target
- to **shape the reflectivity and phase** of XUV

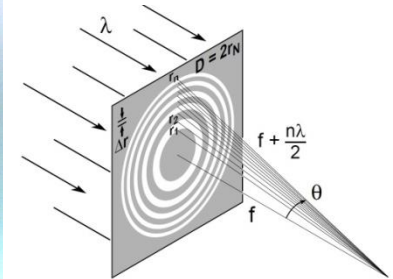
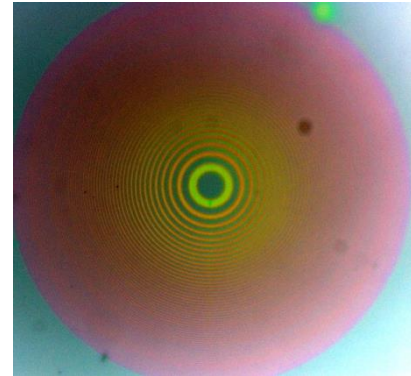
XUV attosecond optics



Transmissive optics



Diffraction optics

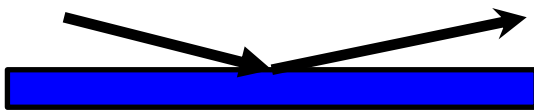


Zone plate image (light microscope)

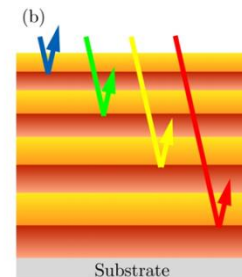
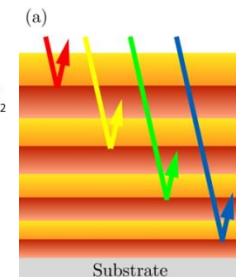
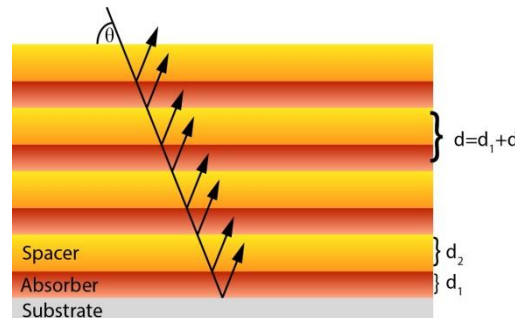
Courtesy of C. Späth

Reflective optics

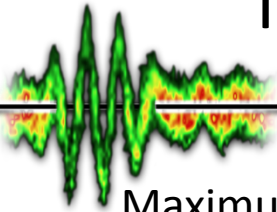
Bulk reflectors



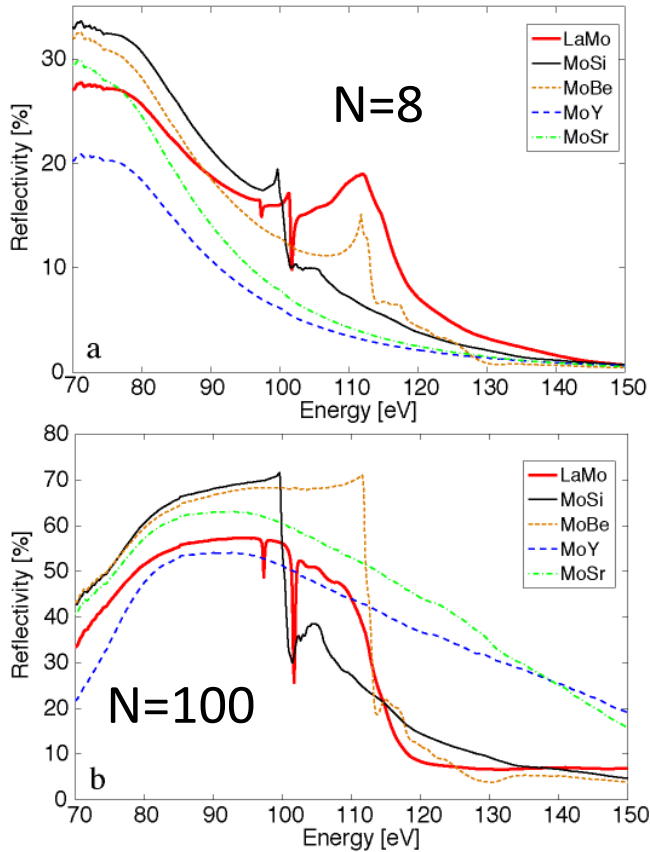
Multilayer mirrors



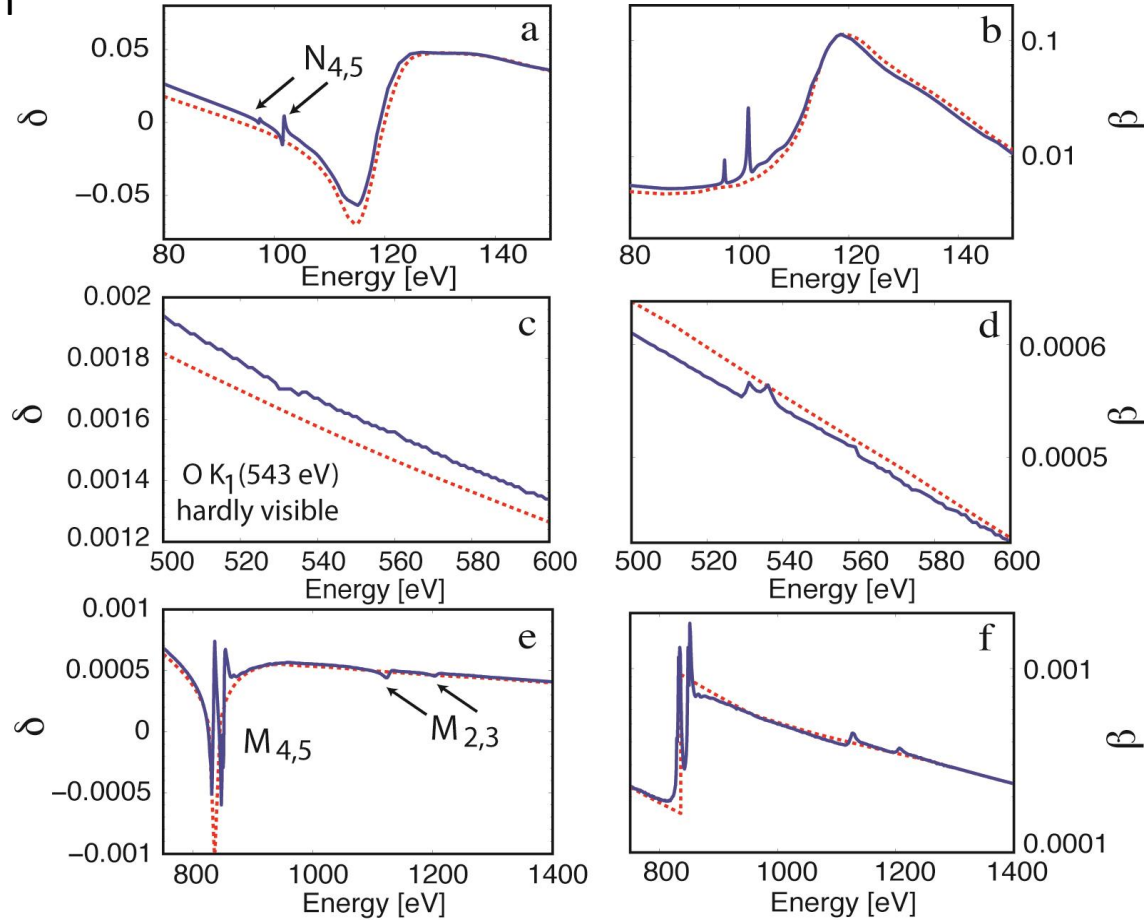
Test new material systems: LaMo (80-130 eV)



Maximum reflectivity simulation



Lanthanum: measurement of optical constants*

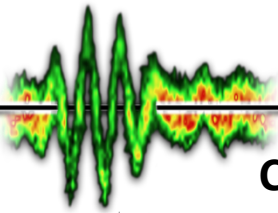


* Available at <http://www.cxro.lbl.gov/>

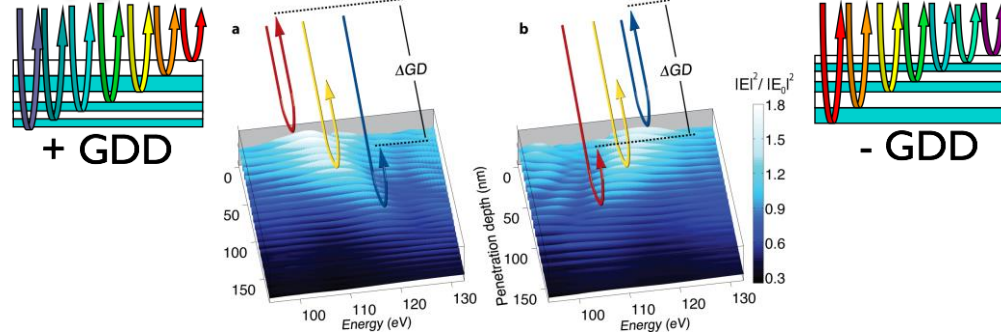
Lanthanum-molybdenum multilayer mirrors for attosecond pulses between 80 and 130 eV

Hofstetter *et al.* Optics Express **19**, 1767 (2011)

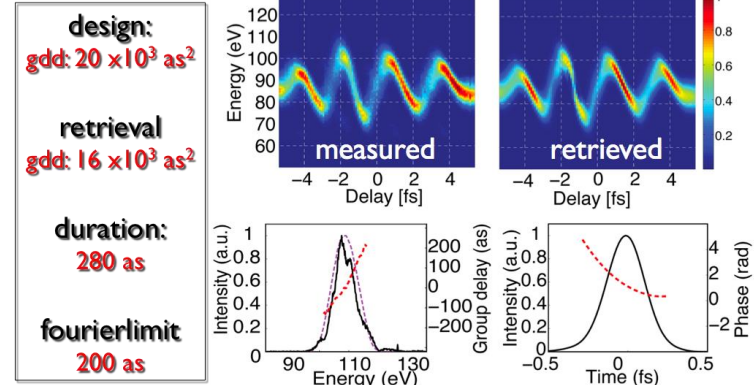
Characterization by attosecond streaking



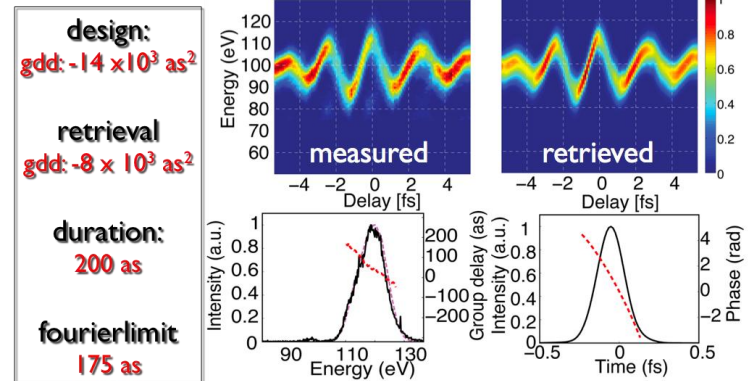
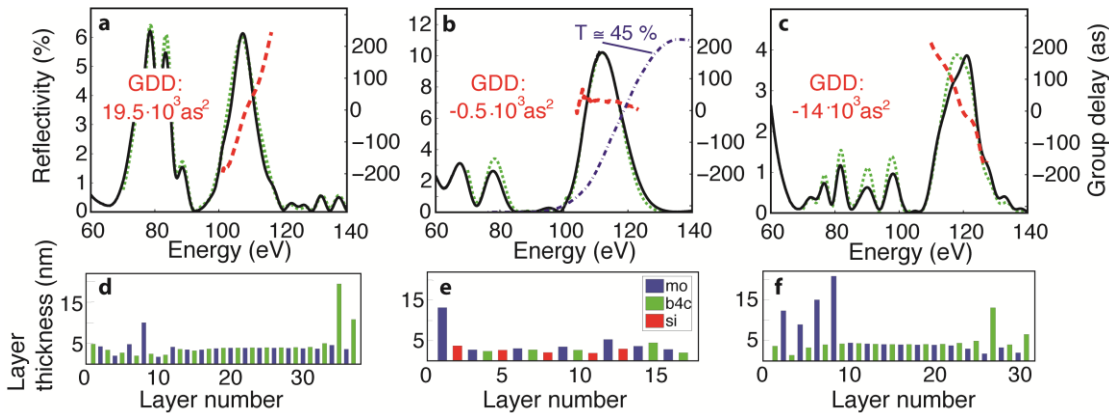
Chirped XUV multilayer mirrors



Streaking retrieval

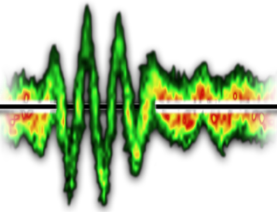


Design/Spectra



Attosecond dispersion control by extreme ultraviolet multilayer mirrors
Hofstetter *et al.* Optics Express **19**, 1767 (2011)

Attosecond Spectroscopy Requirements



Attosecond XUV/soft x-ray multilayer optics

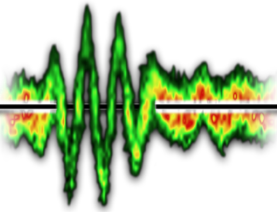


≈ 120 eV (MoSi, LaMo, MoB4C)

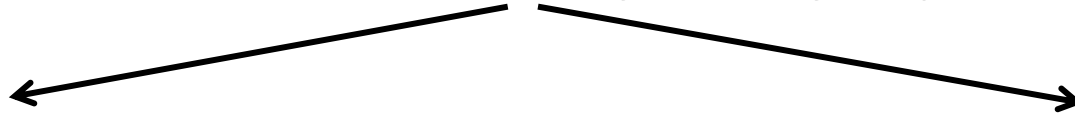
Support of attosecond beamlines
for current experiments

- Selectable central energy
20 eV – 120 eV ✓ Hofstetter *et al.*
New J. Phys. **13**, 063038 (2011)
→LaMo as mirrors
- Selectable bandwidth
1 eV – 30 eV ✓ Goulielmakis *et al.*
Science **320**, 1614 (2008)
→80 as pulses
- High temporal resolution
as – fs ✓ Schultze *et al.*
Science **328**, 1658 (2010)
→Delay in photoemission
- Selectable spectral phase
0 – ± 20000 as² ✓ Hofstetter *et al.*
Optics Express **19**,1767 (2011)
→Attosecond chirp control
- High efficiency ✓ for this energy range with MoSi
or LaMo system already given

Attosecond Spectroscopy Requirements



Attosecond XUV/soft x-ray multilayer optics



Support of attosecond beamlines
for current experiments

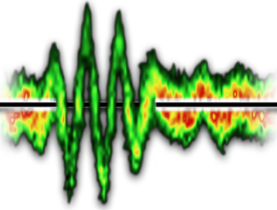
Development of optics for future
experiments at higher energies

≈ 400 eV (CrSc)

- Selectable central energy
20 eV – ~~120~~ 400 eV
- Selectable bandwidth
1 eV – 30 eV
- High temporal resolution
as – fs
- Selectable spectral phase
0 – ± 20000 as²
- High efficiency



Chromium/Scandium multilayer mirrors



Cr/Sc used for multilayer mirrors for quite a while

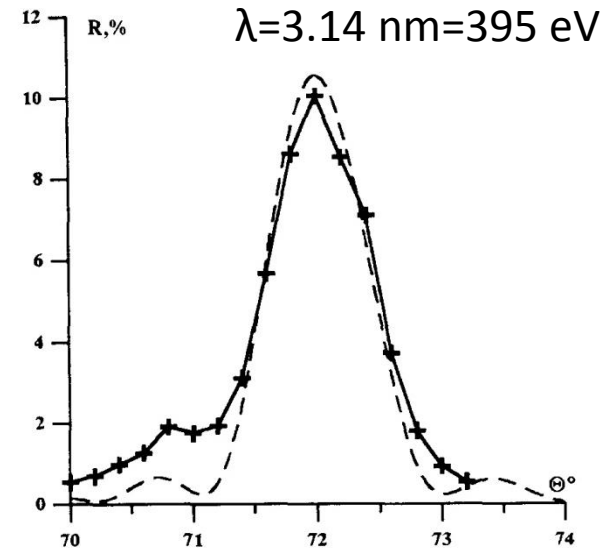
- Salashchenko *et al.* (1996)

Short-period X-ray multilayers based on Cr/Sc

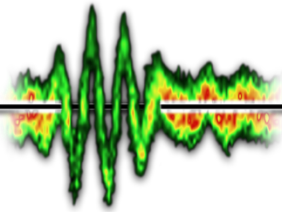
N.N. Salashchenko, E.A. Shamov

Institute for Physics of Microstructures RAS, Nizhni Novgorod, GSP-105, Russia

Received 6 June 1996; revised version received 15 August 1996; accepted 21 August 1996



Chromium/Scandium multilayer mirrors



Progress in short period multilayer coatings for water window applications

E.M. Gullikson, F. Salmassi, A.L. Aquila and F. Dollar

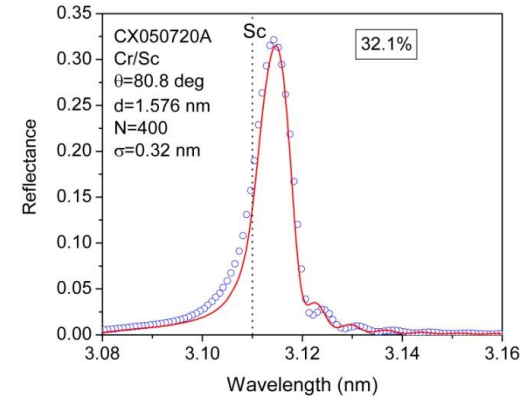
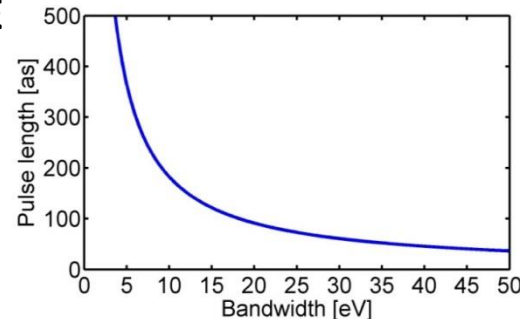
Center for X-Ray Optics, Lawrence Berkeley National Lab, Berkeley, CA 94720

Cr/Sc used for multilayer mirrors for quite a while

- Salashchenko *et al.* (1996)
- Schäfers *et al.* (2000)
- Eriksson *et al.* (2003)
- Gullikson *et al.* (2006)

Attosecond pulses: large bandwidth ΔE

$$E [\text{eV}] \times t [\text{as}] = 1824$$



High-periodic multilayer mirrors: intrinsically low bandwidth ΔE

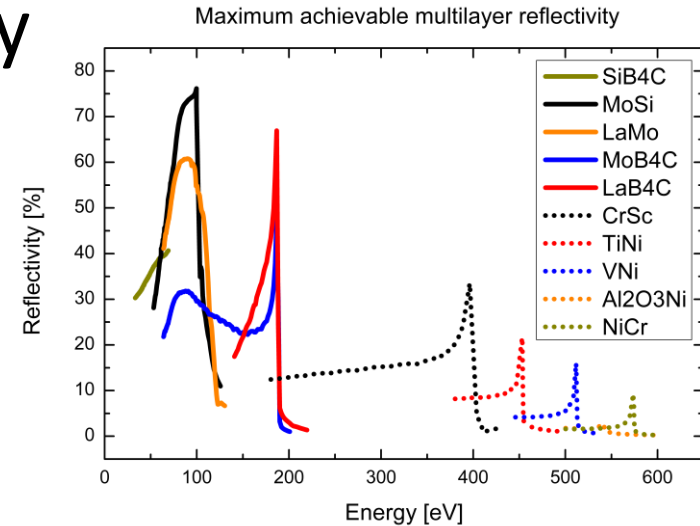
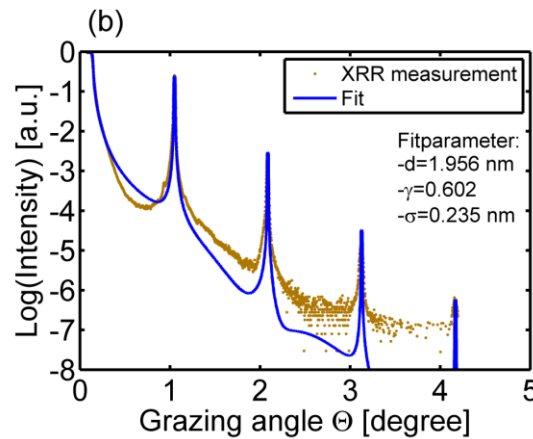
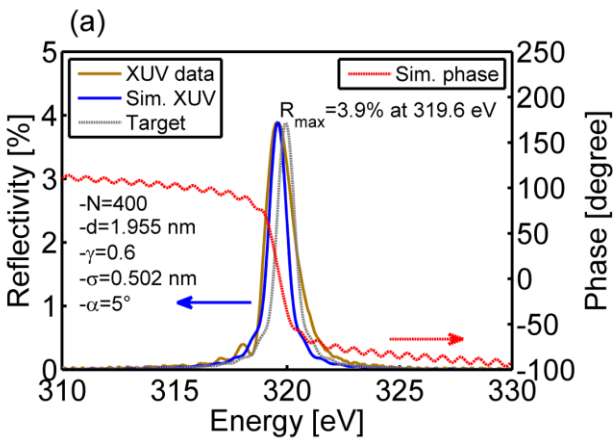
$$\frac{\Delta E}{E} = \frac{1}{N}$$

Threshold:
Ref. > 10% at 5°

System	E [eV]	N	ΔE [eV]
Si/B4C	50	6	6
Mo/Si	93	6	13
Cr/Sc	150	70	3
Cr/Sc	300	400	1

Layer accuracy

- Selectable central energy
20 eV – 400 eV



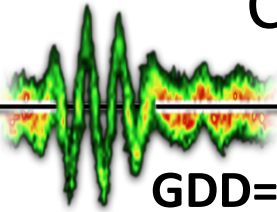
(up to 400 eV)

Deviation: <0.05 angstrom
 Guggenmos *et al.*

Optics Express **21**,21728 (2013)

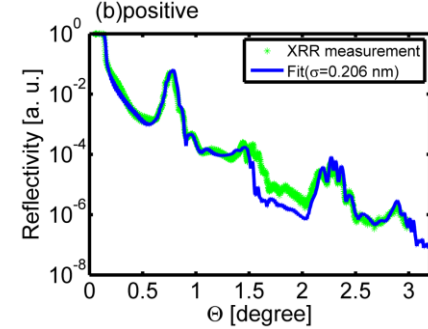
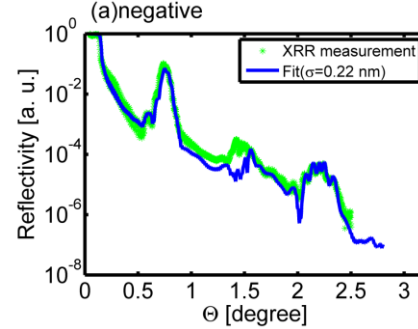
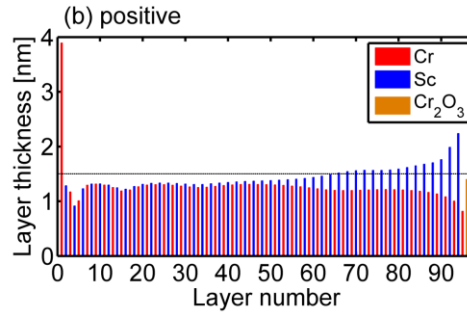
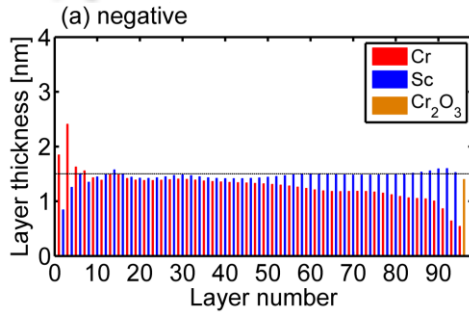
Method	Total [nm]	Period [nm]	σ [nm]	gamma	Deviation [%]
Target:	782.3	1.953		0.6	
Profilometry:	782.959	1.9546			0.08
XRR:	783.5	1.956	0.235	0.602	0.15
XUV:	783.1	1.955	0.502	0.6	0.10

Chirped CrSc multilayer mirrors above 300 eV



GDD=-8000as²

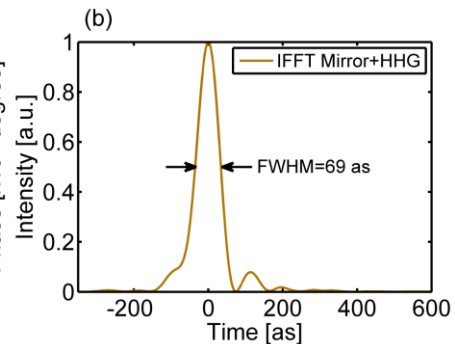
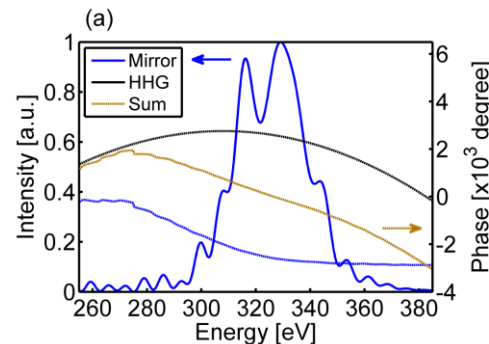
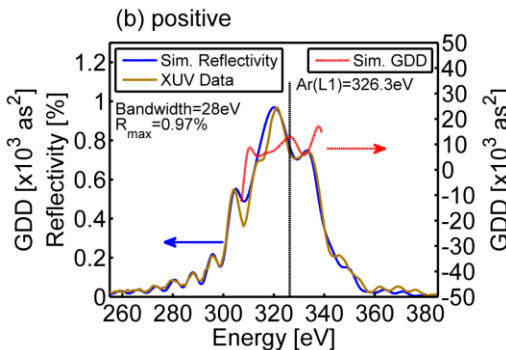
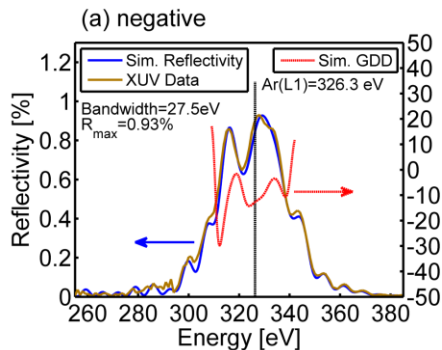
GDD=+8000as²



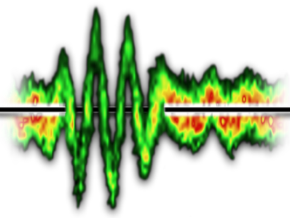
- Selectable bandwidth
1 eV – 30 eV ✓
- High temporal resolution
as – fs ✓
- Selectable spectral phase
0 – ± 20000 as² ✓

Aperiodic CrSc multilayer mirrors for attosecond water window pulses

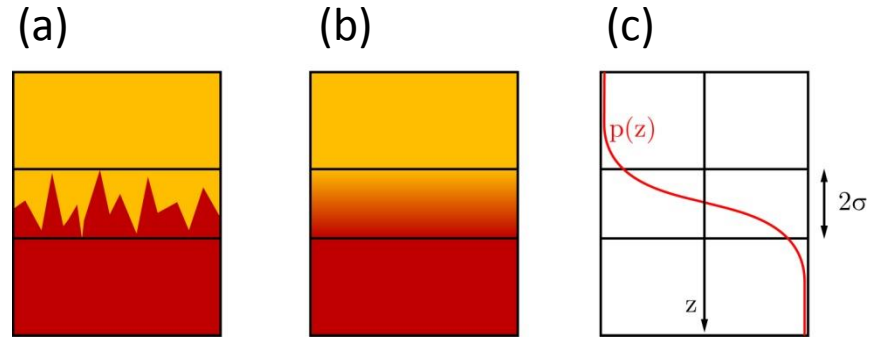
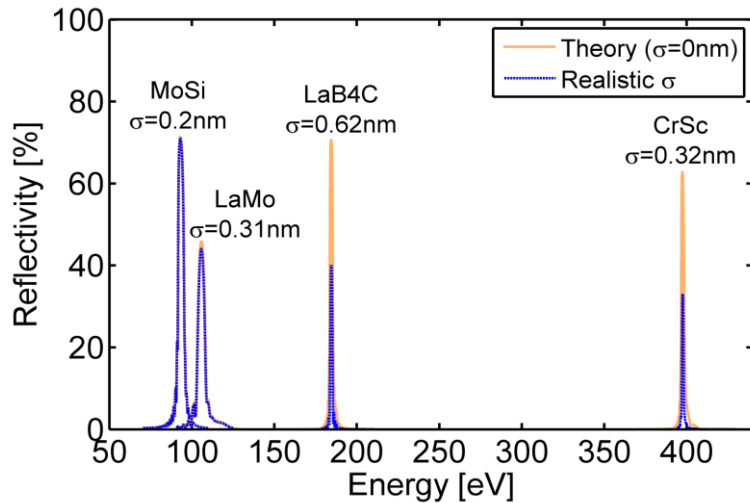
Guggenmos *et al.* Optics Express **21**,21728 (2013)



Attosecond physics towards water window



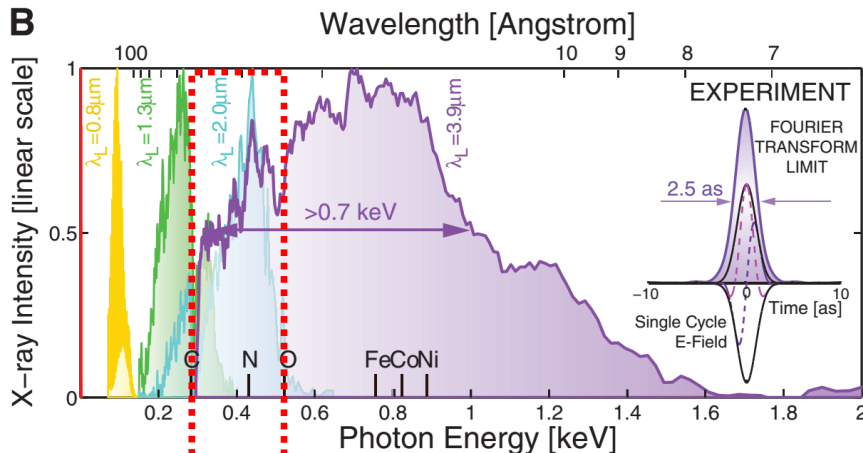
• High efficiency



- a) roughness
- b) interdiffusion/intermixing
- c) interface profile function

$$R = R_0 \exp\left(-2\pi m \frac{\sigma}{\lambda}\right)^2$$

HHG Supercontinua already measured:



Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers

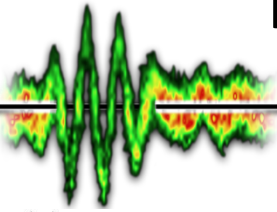
Popmintchev *et al.* Science **336**, pp. 1287-1291 (2012)

Potential applications:

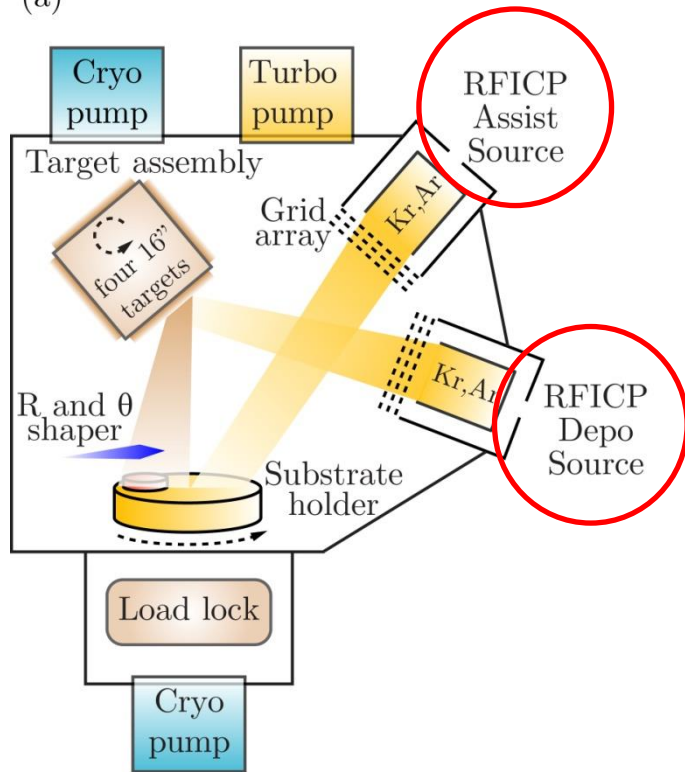
- access to deeper core states dynamics
- access to biological relevant edges (C, Ca, K, N, O)
- shorter pulses due to larger bandwidth access

water window: 1s C (284eV) – 1s O (543eV)

Ion Beam Deposition - Multilayer realization



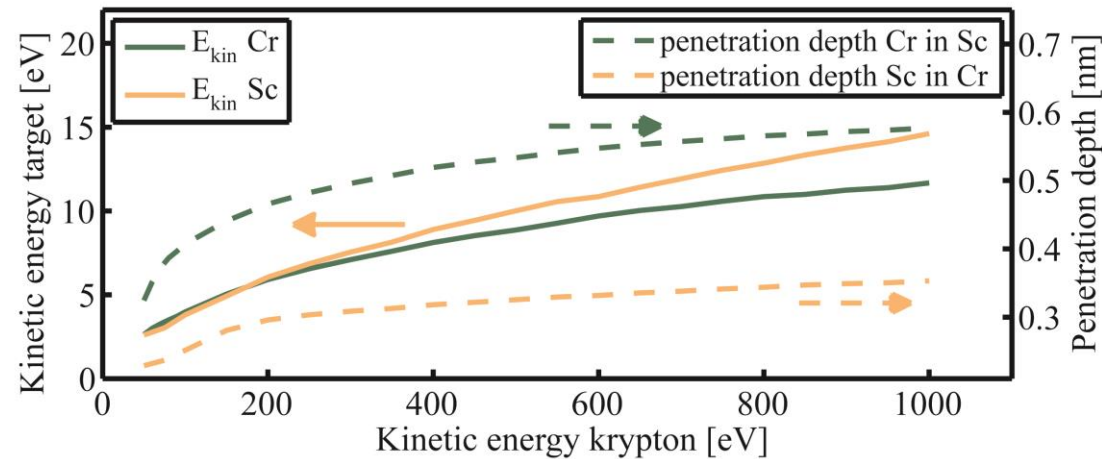
(a)



- 1) variation of ion energy
- 2) interface polishing



Using Monte-Carlo simulation method:

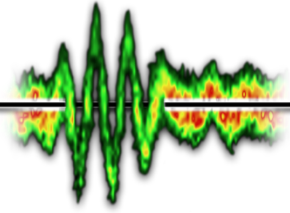


10eV for layer-by-layer growth

Hubler *et al.* Surf. Coat. Tech. **81**, pp. 29 (1996)

- four target materials possible
- in-situ ellipsometric control possible
- load-locked system
- shaper for homogenous layer
- high reproducibility (background pressure 10^{-9} mbar)

TEM - Cross Section Analysis



mirror specs for TEM analysis: $N = 80$, $d = 2.95 \text{ nm}$, $\alpha = 45^\circ$

- 1) variation of ion energy for better layer growth
- 2) every 10 periods assist-ions for polishing the Cr layer

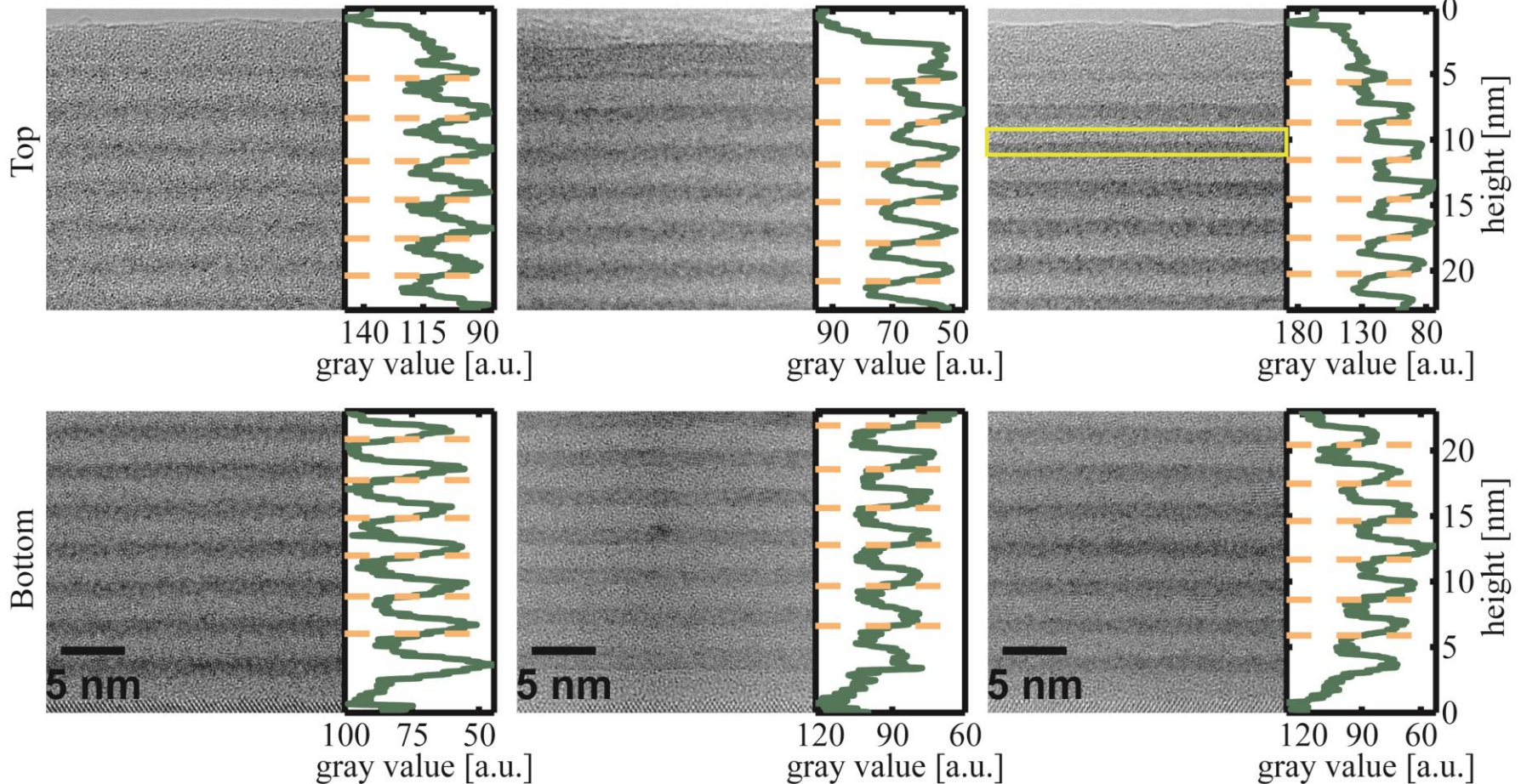
Sc

Cr

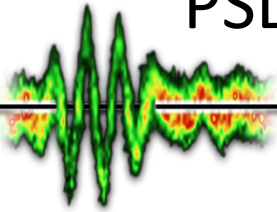
(a) default

(b) optimized

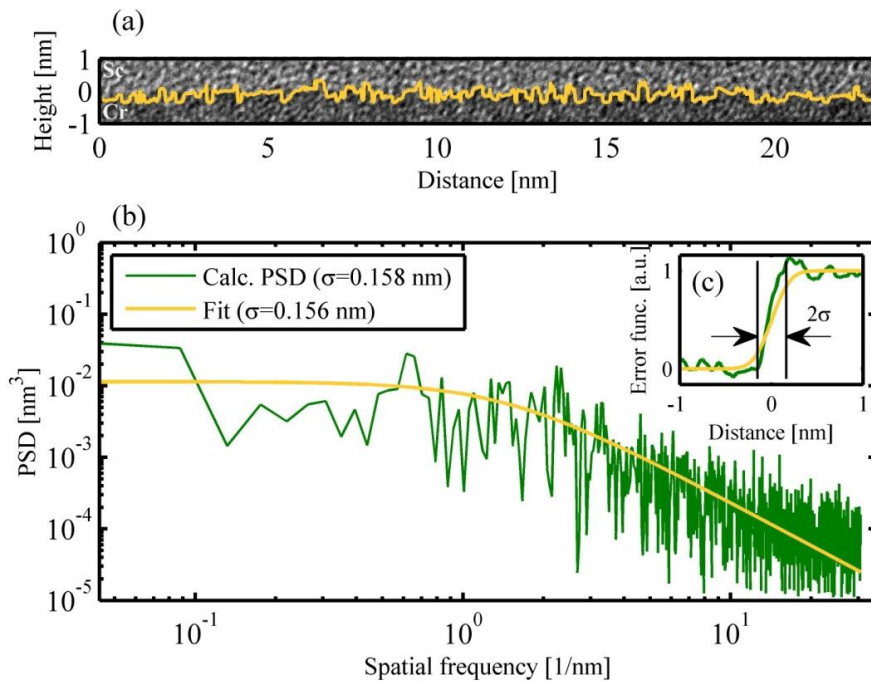
(c) optimized + assist



PSD – Power Spectral Density – TEM data analysis



Horizontal gray value analysis:



Roughness σ dependence on interface „fluctuations“:

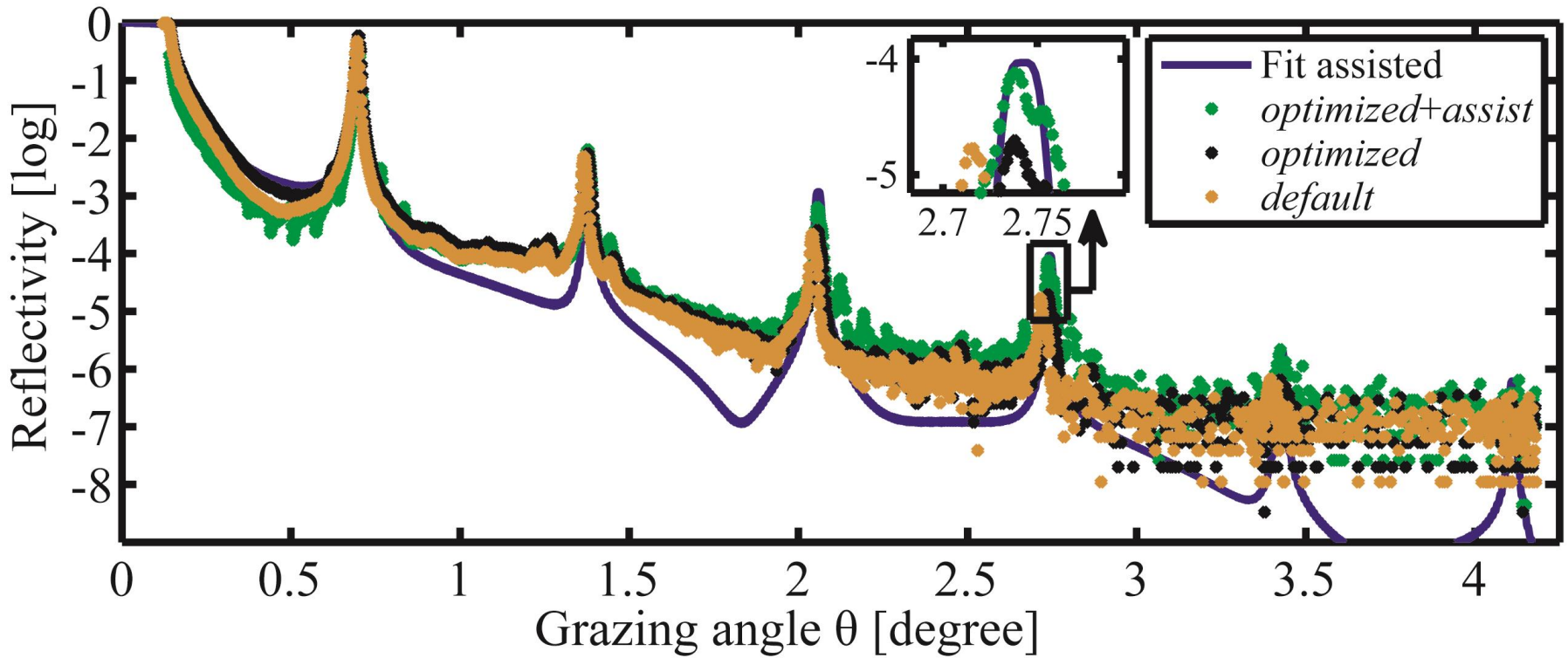
$$\sigma = \left(\int_{f_{\min}}^{f_{\max}} \text{PSD}(f_x) df_x \right)^{1/2}$$

Roughness evolution:

	default	optimized	optimized + assist
σ_{top} (nm)	too blurry	0.3	0.24
σ_{bottom} (nm)	0.25	0.18	0.18

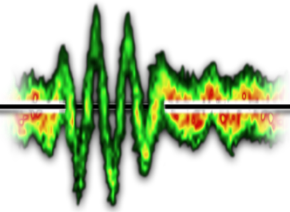
XRR analysis

XRR analysis (Mo K_{α} at $\lambda=0.07\text{nm}$) of TEM-designed mirrors

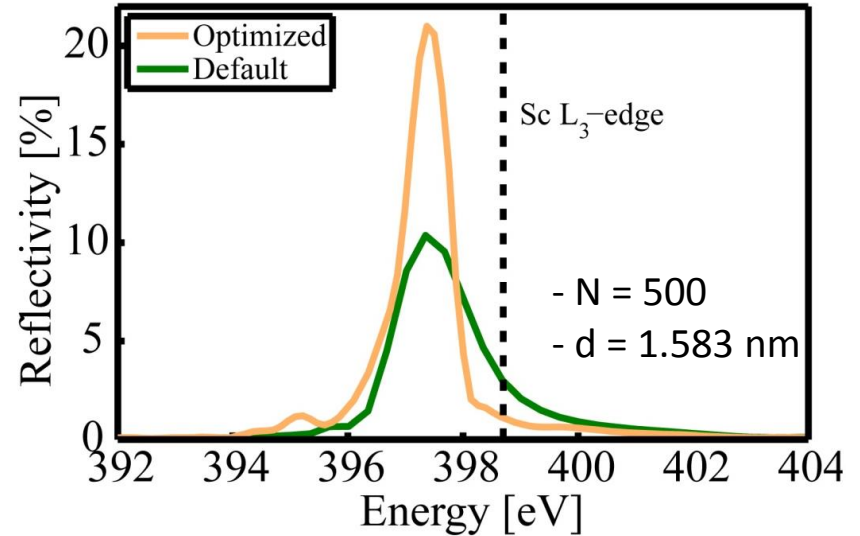


	default	optimized	optimized + assist
Roughness σ [nm]	0.26	0.24	0.21

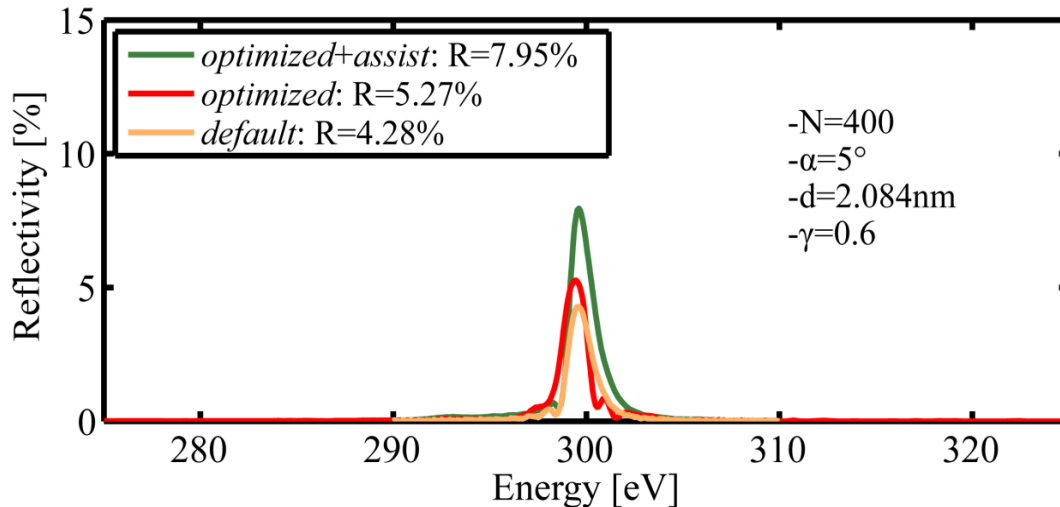
Soft X-ray measurement



doubled the reflectivity at Sc L_3 -Edge
with optimized parameters
(ion polish data still pending)



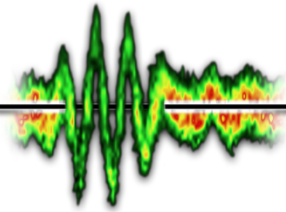
Measured at ALS in Berkeley



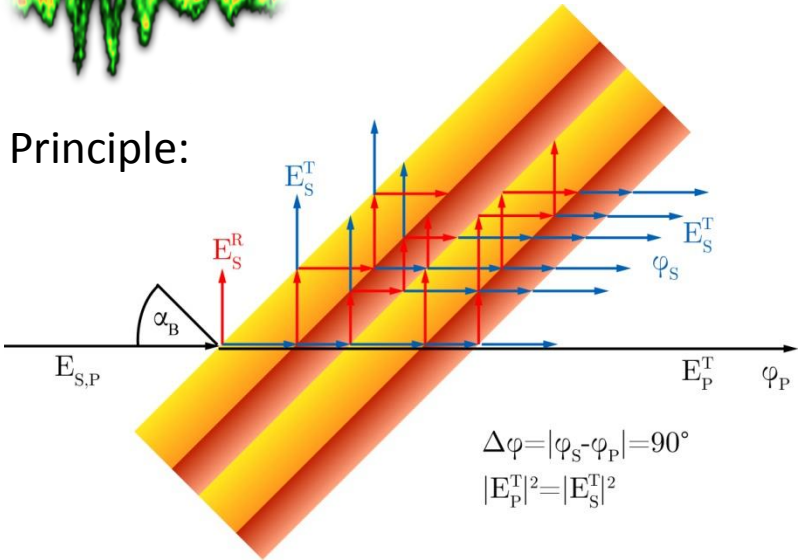
Measured at PTB (BESSY II in Berlin)

**Ion polished Cr/Sc attosecond
multilayer mirrors for high water
window reflectivity**
Guggenmos *et al.* Optics Express **22**,
26526 (2014)

Polarizer (for circularly polarized XUV)



Principle:



$$\Delta\varphi = |\varphi_S - \varphi_P| = 90^\circ$$

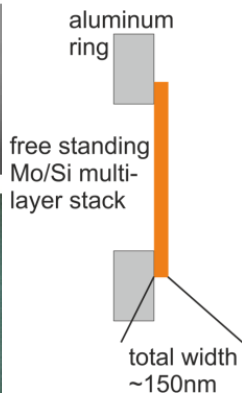
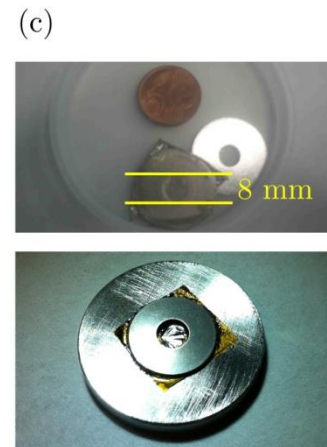
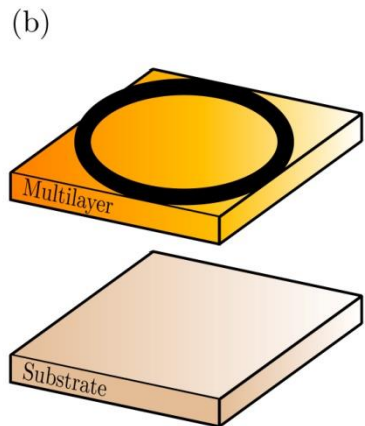
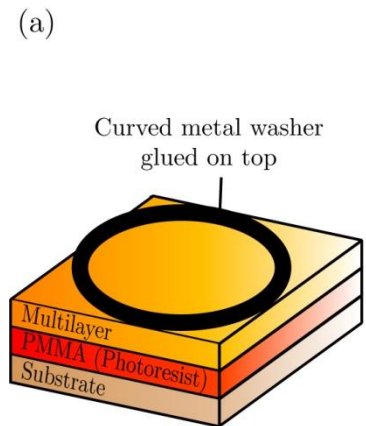
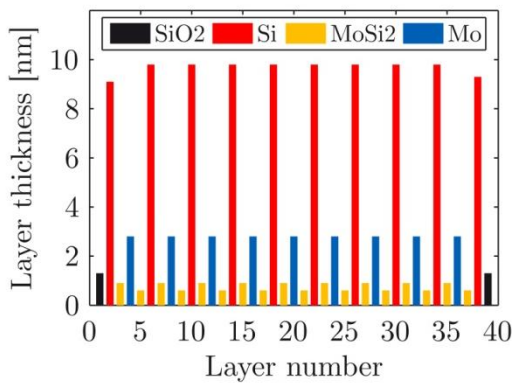
$$|E_P^T|^2 = |E_S^T|^2$$

Application:

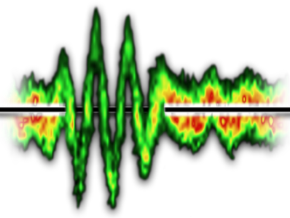
- access to M3 p-splitting of Ni
- spin dependent electron emission due to left or right handed circular XUV
- XUV magnetic circular dichroism (XMCD)
- time-resolved utilizing attosecond pulses

Distinguishing the ultrafast dynamics of spin and orbital moments in solids
 Boeglin *et al.* Nature **465**, 458 (2010)

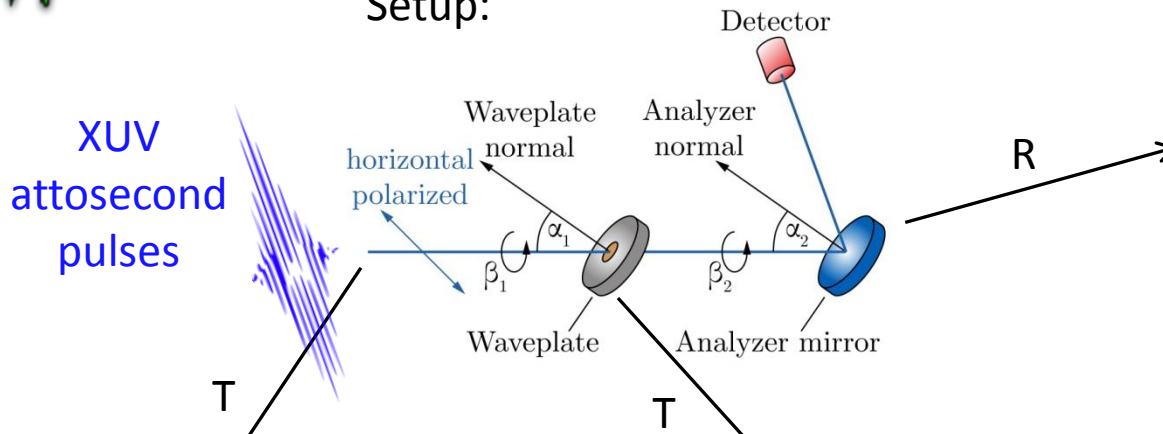
Realization:



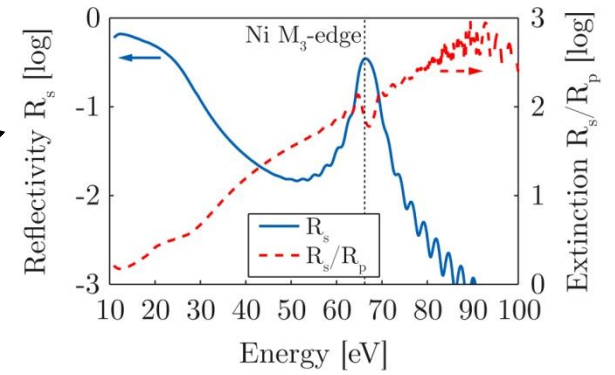
Polarizer (for circularly polarized XUV)



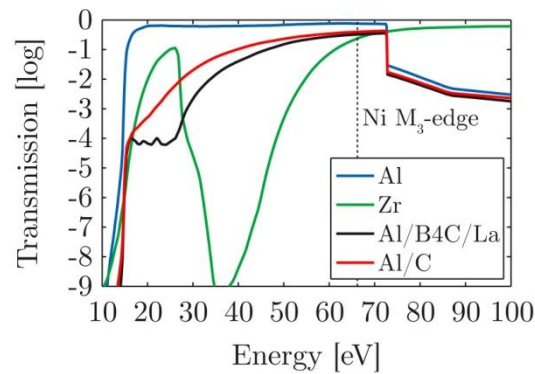
Setup:



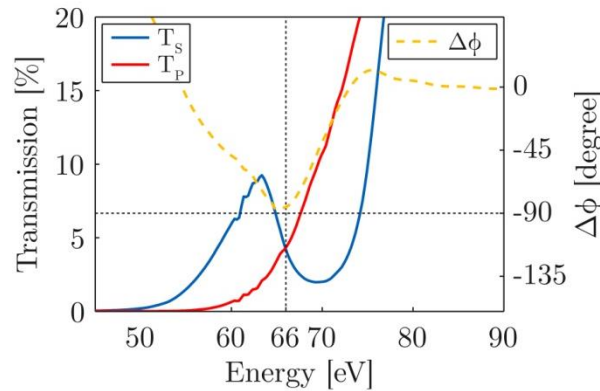
Analyzer:



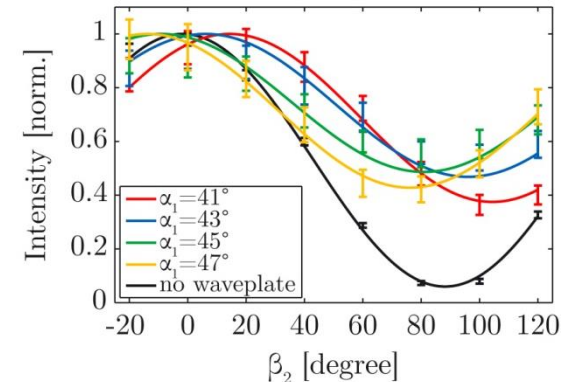
Filter:



Polarizer:



Results:

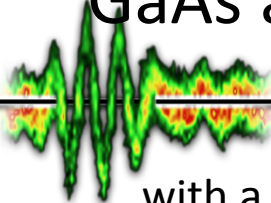


Generation of circularly polarized high harmonic radiation using a transmission multilayer quarter waveplate

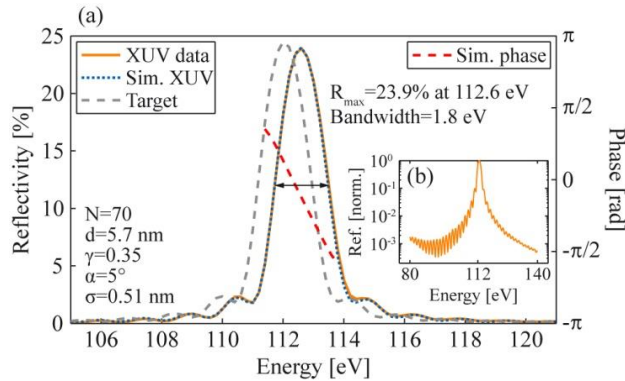
Schmidt, Guggenmos *et al.* Optics Express **23**, 33564 (2015).

Ellipticity (measured): $\epsilon=0.75$
 Ellipticity (simulated): $\epsilon=0.79$

GaAs at 112 eV - Limitations – Experimental bottlenecks

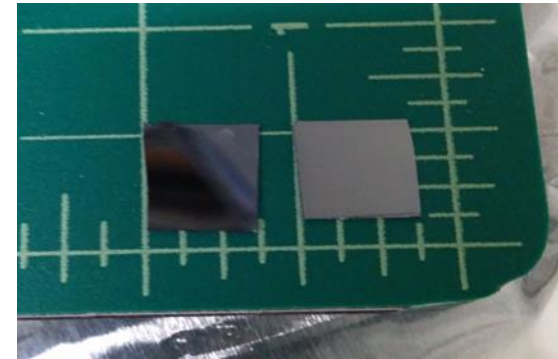


XUV multilayer mirror
with a spectral bandwidth of 1.8 eV



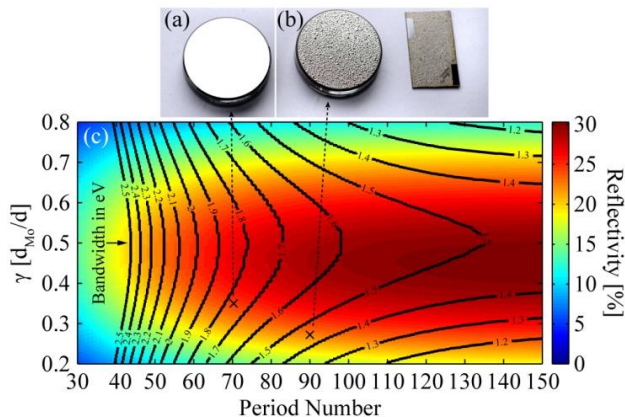
Characterized at PTB, BESSY II in Berlin

As-capped GaAs for pure GaAs after bake out



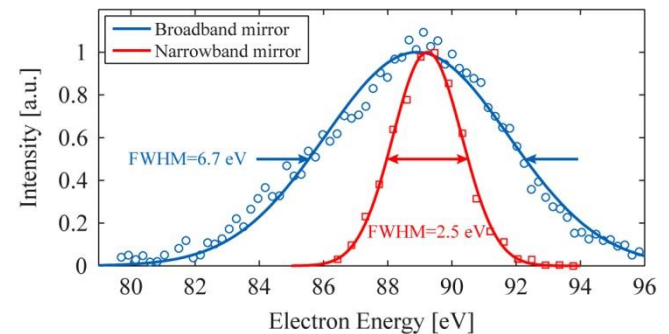
Collaboration with Prof. Amann from the WSI in Garching

Material dependent limitations



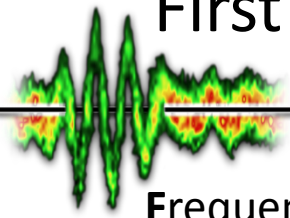
Delamination for detuned Mo/B4C material system
 Optimizing deposition process for finetuning the bandwidth

Spectral resolution for GaAs



Unstreaked photoelectron spectra of the gallium 3d peak

First clean electron streaking measurement on GaAs

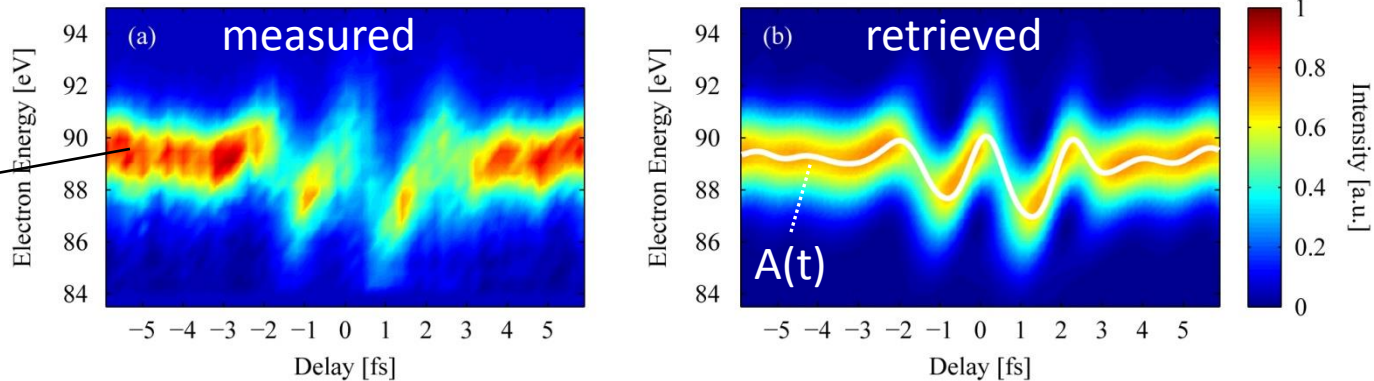


Frequency-resolved optical gating for complete reconstruction of attosecond bursts:

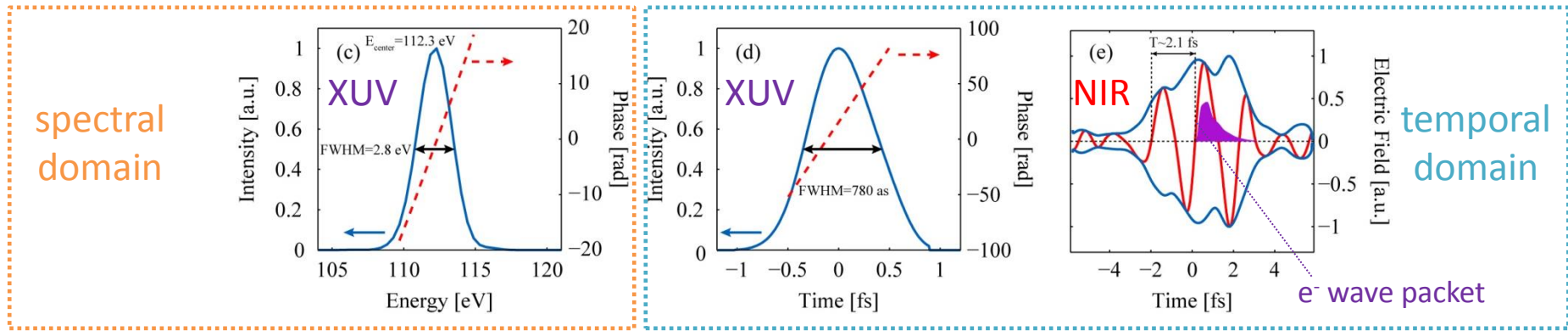
FROG/CRAB – analysis for attosecond pulse characterization

Trebino *et al.* Rev. Sci. Instrum. **68**, (1997)

J. Gagnon *et al.* Appl. Phys. B **92**, (2008)



shifted by the binding energy of Ga 3d electrons (18.7 eV) and the work function of GaAs (4.69 eV)

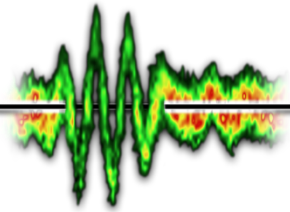


Simulated electron wave packet duration : 870 as ($*2.5 \sim T_{E\text{-Field NIR}}$)

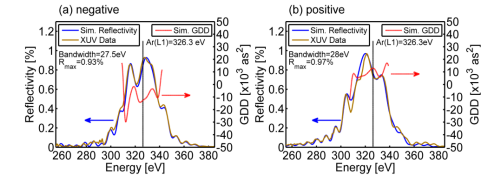
Attosecond photoelectron streaking with enhanced energy resolution for small-bandgap materials

Guggenmos *et al.* Optics Letters 41, 3714 (2016)

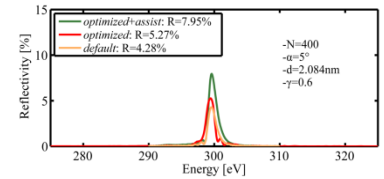
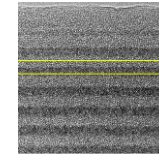
Conclusions



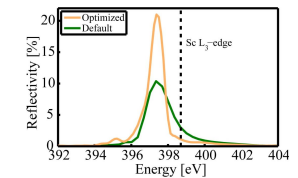
First aperiodic mirrors for chirped attosecond water window pulses



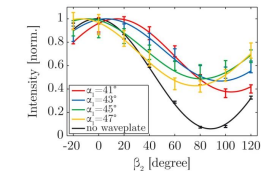
Interface optimization by optimized kinetic energy and ion polishing



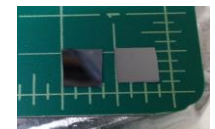
Up to now, doubled the reflectivity at Sc L₃-edge >20% for a pure Cr/Sc multilayer (without barrier layers) (Assisted one should show an even higher value.)



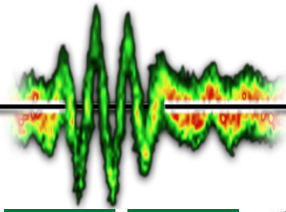
Generation of circularly polarized high harmonic radiation utilizing a transmission multilayer quarter waveplate



The first time-resolved attosecond measurement of GaAs, one of the most important direct bandgap semiconductor



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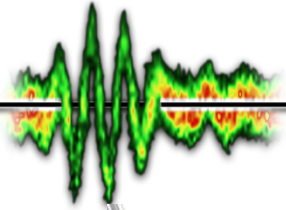
Deanship of Scientific
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Team Attoworld



Thank you for your attention – Questions?