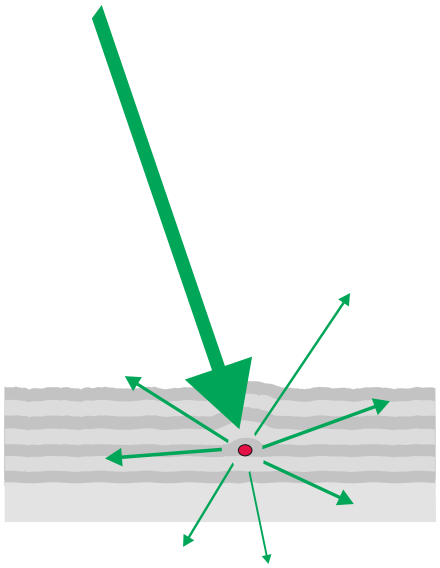
Assessing critical imperfections of high-end optical coatings



Sven Schröder

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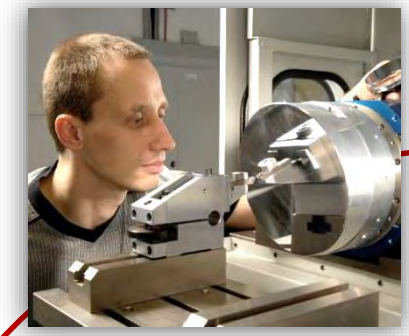
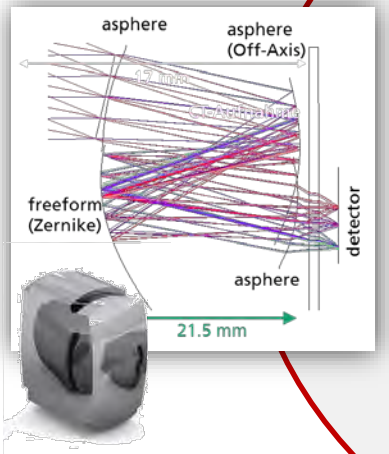
Fraunhofer
IOF

Fraunhofer Institute IOF: Development of optical systems

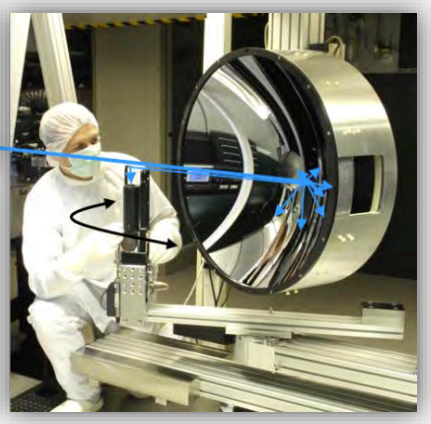
Ultra-precision machining



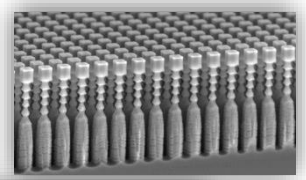
Optical and functional coatings

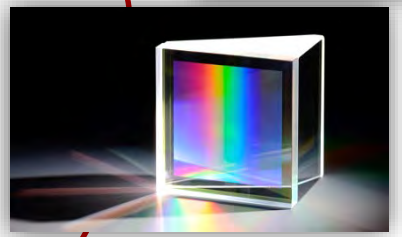


Optical systems design



Measurement and modeling: Roughness, scattering, functional properties



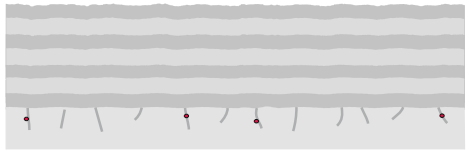


Micro- and nano structuring

Types of imperfections in coatings

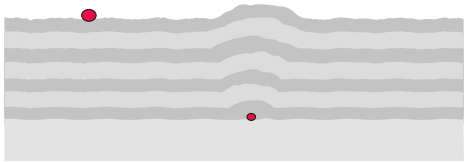
Interface roughness

Substrate roughness, material, polishing process, intrinsic film roughness, deposition process, adatom mobility / growth process, …



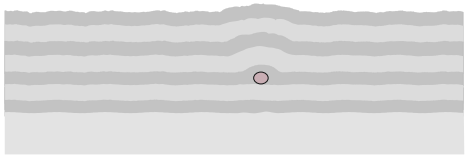
Subsurface defects

Polishing process, materials, Bilby layer, cleaning, …



Surface defects

Scratch, dig, particle contaminations, cleaning, handling, pre-treatment, …



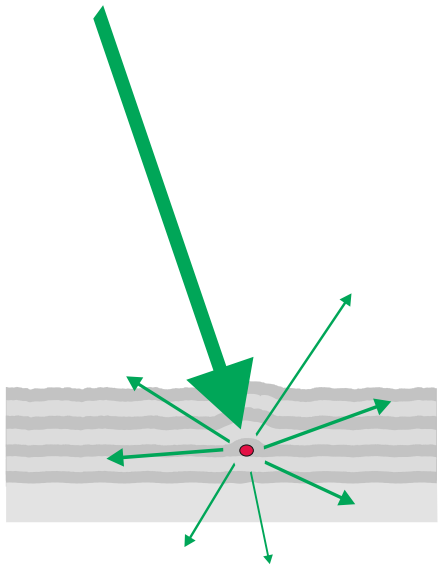
Coating defects

Deposition process, machine type, maintenance, cleanliness, …

+ molecular contaminations, stoichiometric defects, bulk inhomogeneities, …

Most imperfections can be assessed by light scattering methods

Impact of imperfections onto performance



Impact of imperfections onto optical and functional properties depends on:

- Density and size of imperfections
- Dielectric properties of imperfections
- Position in film system
- Local field strength / interference conditions

Light scattering can be made sensitive to all these factors

Yet, detecting imperfections does not necessarily allow conclusions about their impact onto certain performance parameters

→ Comprehensive pool of characterization methods and expertise

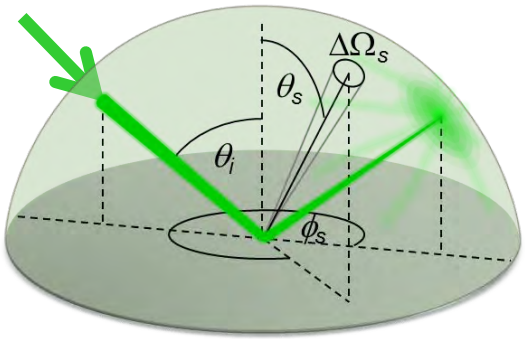
Characterization methods

- **Optical properties**
  - Spectral reflectance, transmittance
  - Optical losses: CRD, absorption, light scattering
  - LIDT: ns, fs, spectral
- **Structural properties**
  - Roughness: AFM, WLI, light scattering
  - Morphology: SEM, TEM, …
  - Molecular / atomar: XPS, TOF-SIMS, …
- **Functional properties**
  - Scratch resistance / tribology
  - Environmental stability
  - Thermal stability
  - Contact angle analysis

+ Understanding to link optical, structural, and functional properties

Strong expertise in some fields and cooperation with partners for other methods.

Light scattering ARS vs TS



θ_s … polar scatter angle
P_s … scattered power
P_i … incident power
$\Delta\Omega_s$ … detector solid angle

BSDF… bidirectional scattering distribution function

Angle Resolved Scattering (ASTM E2387, ISO/WD 19986)

$$\mathrm{ARS}(\theta_s) = \frac{\Delta P_s(\theta_s)}{\Delta\Omega_s P_i} = \mathrm{BSDF}\cos\theta_s$$

Total Scattering (ISO 13696)

$$TS = \frac{P_s}{P_i} = \int ARS \, \mathrm{d}\Omega_s$$

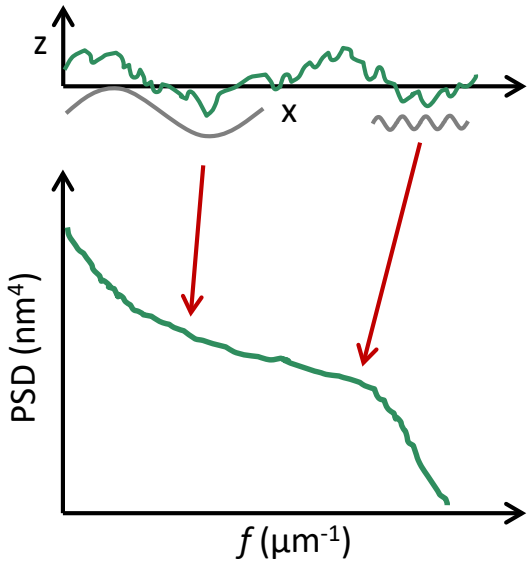
(ISO: 2° - 85°)

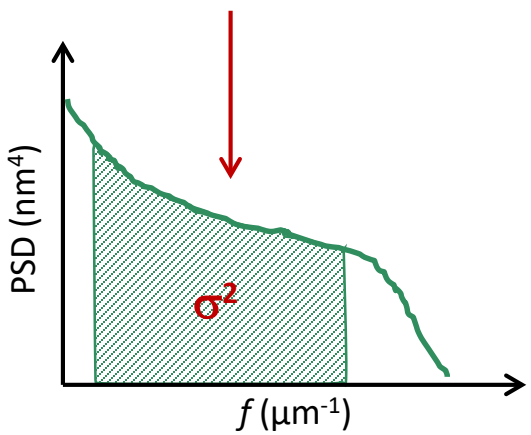
→ Goniometric scatterometers
+ more flexibility
+ more information
- more challenging

→ Coblentz-/ integrating spheres
+ fast
+ single number
- limited information
- only small and plane samples

A. Duparré, J. M. Bennett et al., Applied Optics 41 (2002)
J. M. Bennett, L. Mattsson, Introduction to Surface Roughness and Scattering, (OSA, Wash. D.C., 1999)
J. C. Stover, Optical Scattering: Measurement and Analysis, 2nd ed., (SPIE, Bellingham, Wash., 1995)
S. Schröder, A. v. Finck, A. Duparré: "Standardization of light scattering measurements", Adv. Opt. Technol. (2015)

Surface roughness





Surface Power Spectral Density function

$$PSD(f_x, f_y) = \lim_{L\to\infty} \frac{1}{L^2} |FT\{z(x,y)\}|^2$$

surface spatial frequencies

surface topography

- Power of different roughness components
- Fourier Transform of Autocovariance Function
- Isotropic roughness: PSD(f_x,f_y) → PSD(f)

Rms roughness

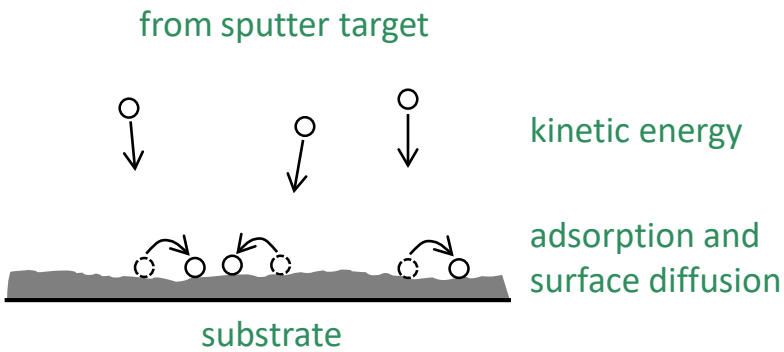
$$\sigma = \sqrt{2\pi \int_{f_{min}}^{f_{max}} PSD(f)\, f \, \mathrm{d}f}$$

- Standard deviation of surface topography
- Band-limited / relevant roughness

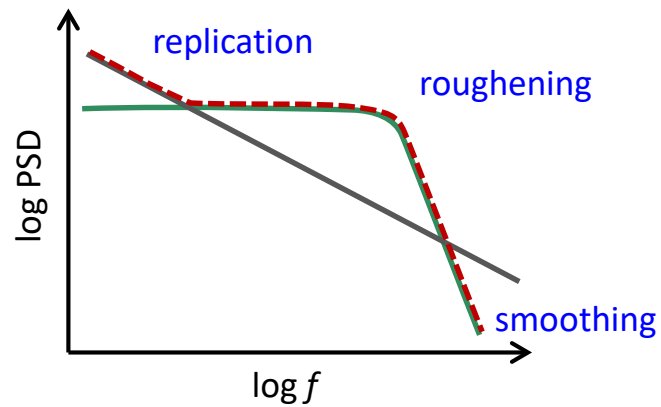
A. Duparré, J. M. Bennett et al., Applied Optics 41 (2002)
J. M. Bennett, L. Mattsson, Introduction to Surface Roughness and Scattering, (OSA, Wash. D.C., 1999)
J. C. Stover, Optical Scattering: Measurement and Analysis, 2nd ed., (SPIE, Bellingham, Wash., 1995)

Roughness evolution models: Qualitative model for thin film growth

Physical vapor deposition process



Thin film roughness spectrum

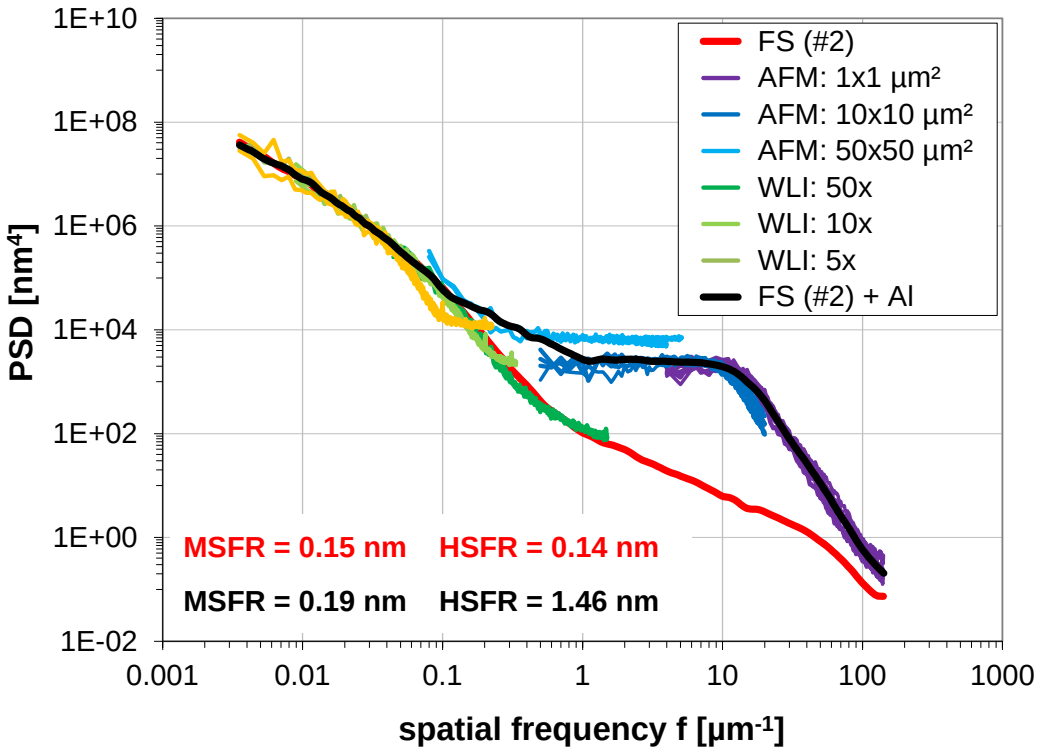


- Replication of low-frequency roughness (substrate)
- Roughening at higer spatial frequencies (shot noise)
- Smoothing at cut-off (surface diffusion)
- Analytic models involving particle size, energy, diffusion length

→ Coating roughness = substrate roughness + intrinsic thin film roughness

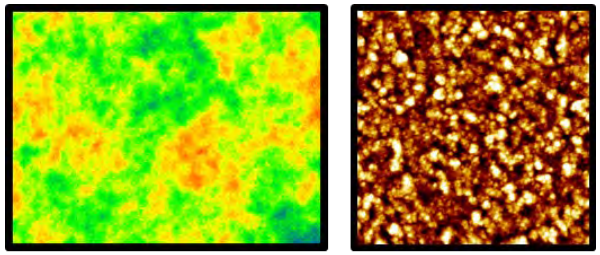
M. Kardar, G. Parisi, and Y.-C. Zhang, Physical Review Letters 56 (1986)
D. G. Stearns, Appl. Phys. Lett. 62 (1993)
D. G. Stearns, D. P. Gaines, D. W. Sweeney, E. M. Gullikson, J. Appl. Phys. 84 (1998)

Example: Aluminum coating on superpolished fused silica



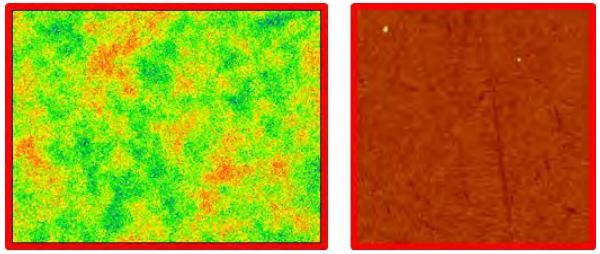
WLI 50x AFM 1x1 µm²

Aluminum coating



σ = 0.14 nm σ = 1.52 nm

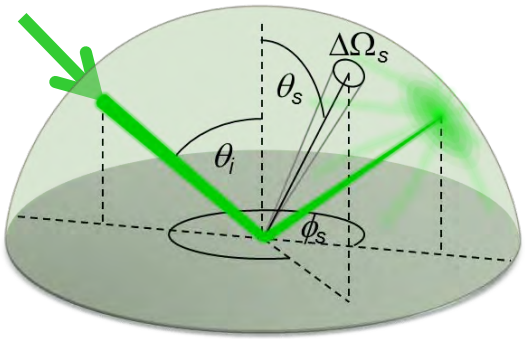
Uncoated substrate



σ = 0.20 nm σ = 0.17 nm

→ **Which part of the roughness spectrum is relevant for the application?**

Relationship light scattering – roughness (single surfaces)



$$\mathrm{ARS}(\theta_s) = \underbrace{\frac{16\pi^2}{\lambda^4}\cos\theta_i \cos^2\theta_s \, Q}_{\text{Optical factor}} \; \underbrace{\mathrm{PSD}(f)}_{\text{Roughness factor}}$$

Optical factor

- optical constants
- geometry
- polarizaton

Roughness factor
= surface PSD

Relevant spatial frequencies

$$f = \frac{\sin\theta_s - \sin\theta_i}{\lambda}$$

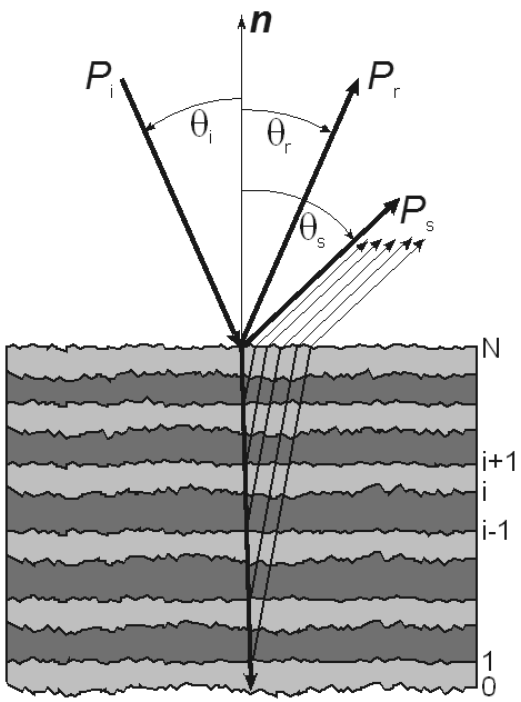
→ **For a given wavelength and geometry, scattering into a certain direction corresponds to the power of a corresponding roughness component with spatial frequency *f*.**

Lord Rayleigh, Proc. Roy. Soc. (1907), S.O. Rice, Commun. Pure Appl. Math. (1951)

E. L. Church et al., Appl. Opt. (1975), E. L. Church, P. Z. Takacs, Appl. Opt. (1993)

J. C. Stover, Optical Scattering: Measurement and Analysis (SPIE, 1995); A. Duparré, In Encyclopedia of Modern Optics (Elsevier, Oxford, 2004); J. M. Bennett, L. Mattsson, Introduction to Surface Roughness and Scattering, 2nd ed. (OSA, Washington, D.C., 1999)

Scattering from thin film coatings



Vector perturbation theory ($\sigma << \lambda$)

$$\mathrm{ARS}(\theta_s) \sim \frac{1}{\lambda^4} \sum_{i=0}^{N} \sum_{j=0}^{N} \underbrace{F_i F_j^*}_{\text{Optical factors}} \underbrace{\mathrm{PSD}_{ij}(f)}_{\text{Roughness factors}}$$

Optical factors

- Multilayer design
- Optical constants
- Polarization

Roughness factors

- PSDs of individual surfaces
- Cross-correlation properties ($i \neq j$)

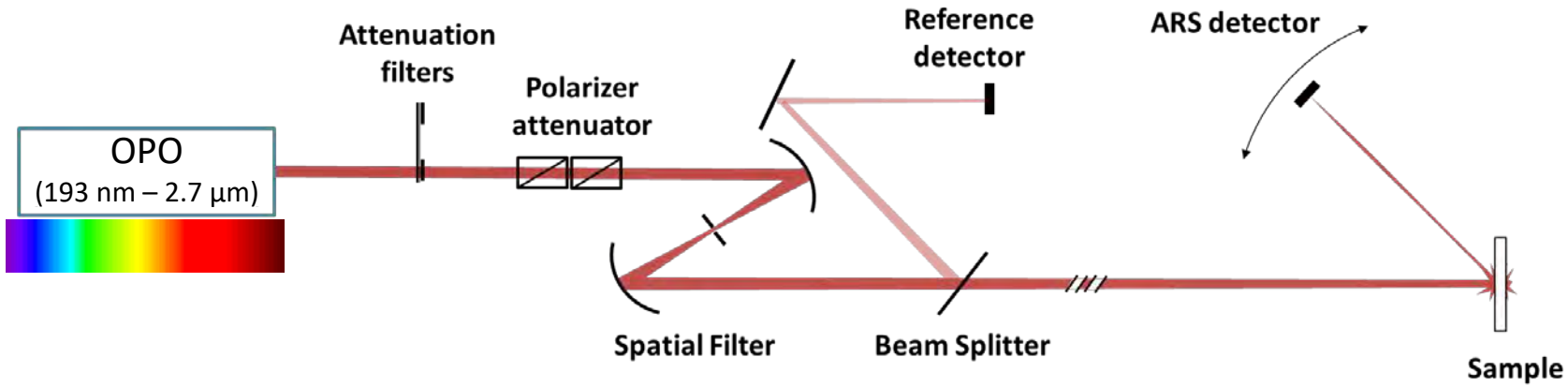
- **Multilayer scatter influenced by roughness and interference effects**
- **N^2 parameters → roughness and correlation models required**
- **Verification through scatter measurements**

J. M. Elson et al., Appl. Opt. 19, 669-679 (1980)

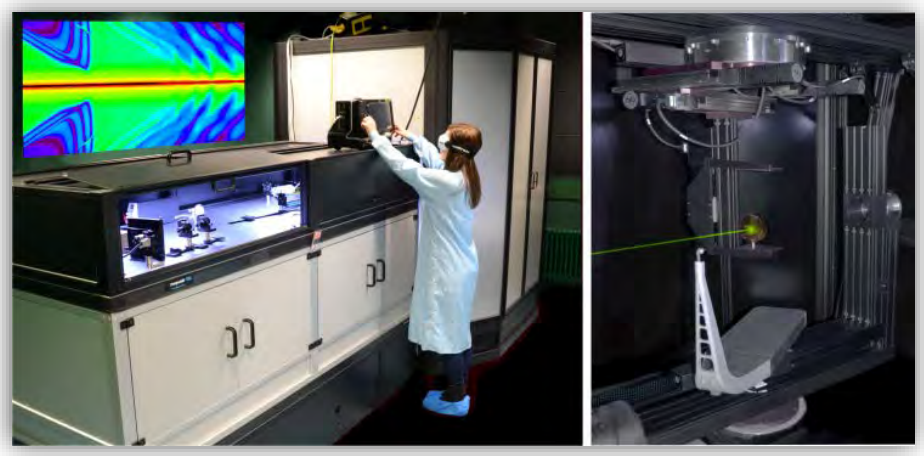
P. Bousquet et al., J. Opt. Soc. Am. 71, 1115-1123 (1981)

Fraunhofer IOF

System for spectral angle resolved scatter measurements
MLS 1600



- 3D Angle resolved light scattering (BSDF), transmittance, reflectance
- UV-VIS-NIR (193 nm – 2700 nm, bandwidth < 0.1 nm)
- High sensitivity (<10^{-6} sr^{-1}), high angular resolution (<0.1°) large samples (up to 700 mm)
- Applications: surfaces, coatings, filters, gratings, materials, …



S. Schröder, D. Unglaub, M. Trost, X. Cheng, J. Zhang, and A. Duparré, "Spectral angle resolved scattering of thin film coatings," Appl. Opt. 53, A35-A41 (2014).

Instruments for angle resolved scatter measurements developed at Fraunhofer IOF

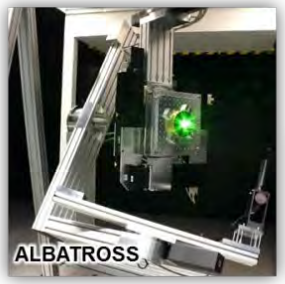
Laboratory scatterometers (EUV-DUV-VIS-IR) → Compact tools → Customized scatterometers developed for partners

Instruments for the DUV and EUV

13.5 nm

193 nm

Big UV-VIS-IR system (325 nm – 10.6 µm)

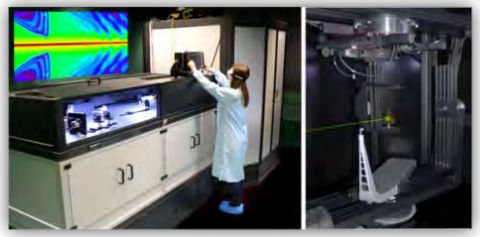


TS (193 nm – 10.6 µm)

Table-top 3D ARS system (VIS-NIR)

Portable scatter sensor (VIS)

New instrument for spectral scatter measurement and damage testing (250 nm – 1750 nm)



Basis: ALBATROSS TT
532 nm, 1064 nm

Basis: ALBATROSS TT
325 nm, 633 nm, 805 nm, 1064 nm,
fiber port, motorized sample stage

New IR scatterometer
(in-plane)
532 nm, 4.6 µm, 10.6 µm,
motorized sample stage

Basis: ALBATROSS TT
405 nm, 532 nm, 640 nm

Basis: ALBATROSS TT
405 nm, 532 nm, 640 nm,
fiber port, motorized sample stage

Basis: horos
roughness sensor

Basis: horos
Premium
BRDF Sensor

→ Scatterometer development based on R&D on high-end optics

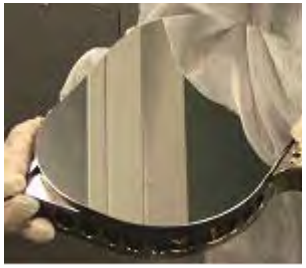
S. Schröder, A. Duparré, A. Tünnermann, Appl. Opt. 47 (2008)
S. Schröder, T. Herffurth, H. Blaschke, A. Duparré, Appl. Opt. 49 (2010)
M. Trost, S. Schröder, T. Feigl, A. Duparré, A. Tünnermann, Appl. Opt. 50 (2011)
A. von Finck, M. Hauptvogel, A. Duparré, Appl. Opt. 50 (2011)
T. Herffurth, S. Schröder, M. Trost, A. Duparré, A.Tünnermann, Appl. Opt. 52 (2013)

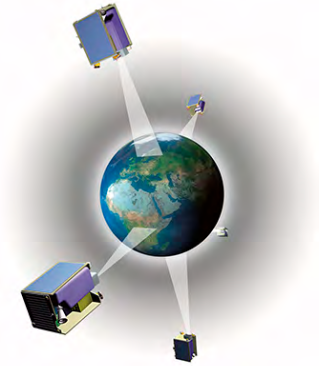
Fraunhofer IOF

Surface roughness and defects

Telescope mirror

Diamond turned and polished Al

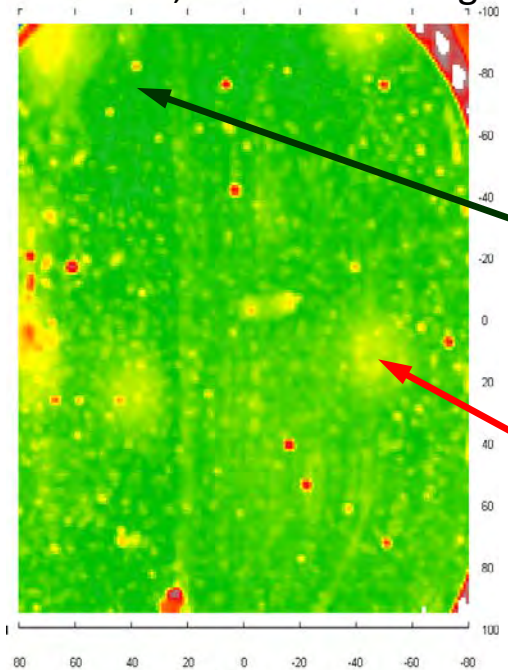




http://www.satimagingcorp.com

Scatter map 532 nm

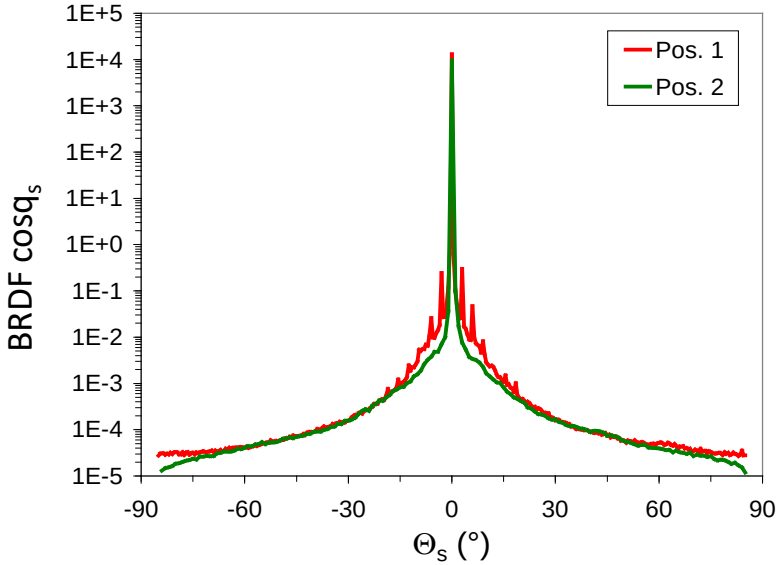
(200x160 mm², fixed scatter angle of 25°)



Pos. 2
(σ = 1.9 nm)

Pos. 1
(σ = 3.0 nm)

Local BRDF



→ **Full area characterization of BRDF, homogeneity, roughness, defects**

→ **Further development to characterize EUV mirrors before coating…**

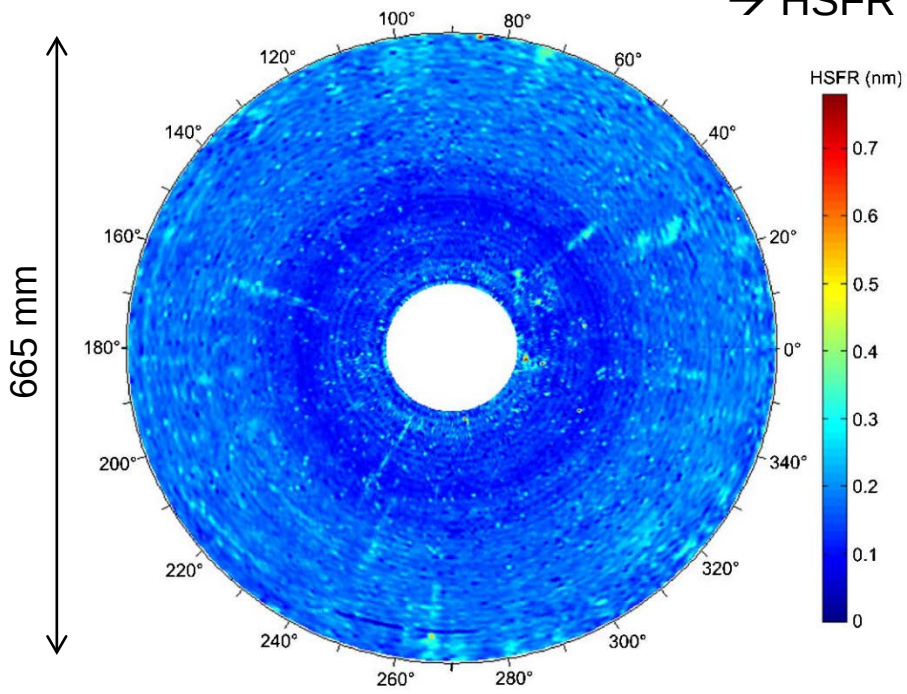
S. Schröder, A. Duparré, Proc. SPIE 7102 (2008)

Fraunhofer IOF

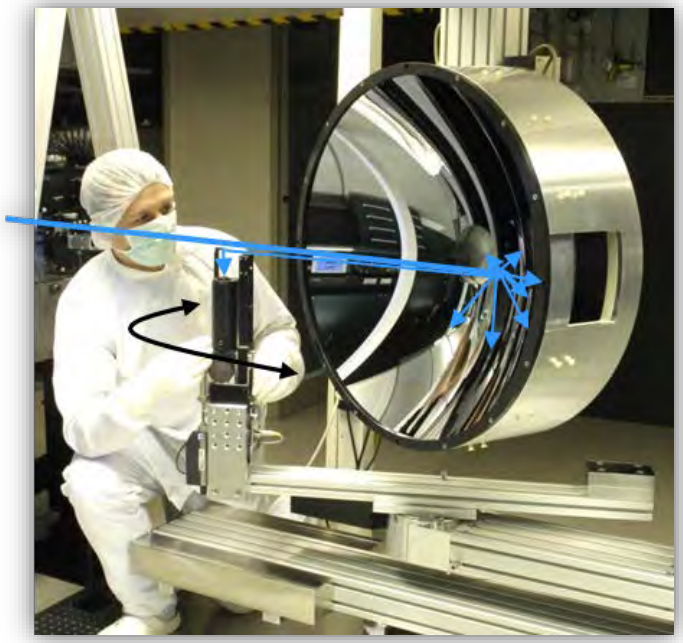
Characterization of EUV mirrors

Scatter map 405 nm

BRDF ~ PSD
→ HSFR (nm)



Instrument ALBATROSS
with EUV collector mirror



→ **Unique / superior technique for roughness characterization of high-end optical surfaces**

M. Trost, S. Schröder, T. Feigl, A. Duparré, Appl. Opt. (2011)

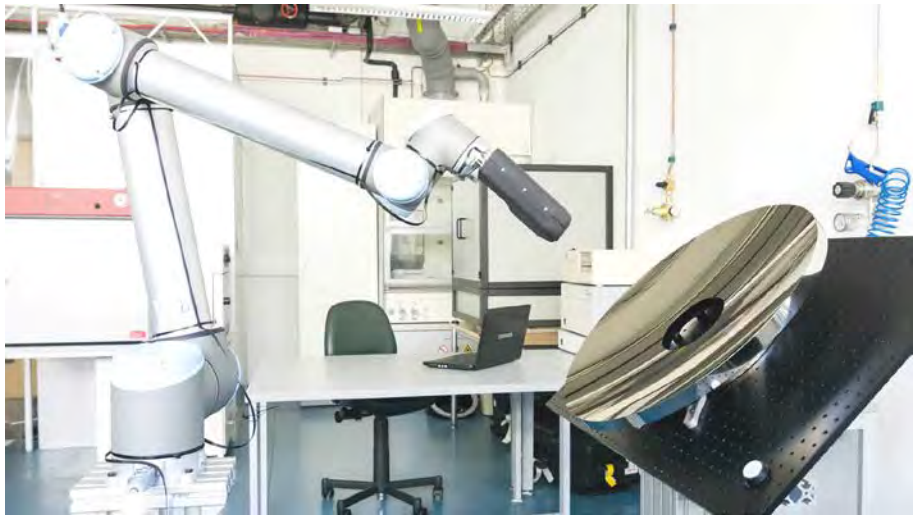
CYMER

Fraunhofer IOF

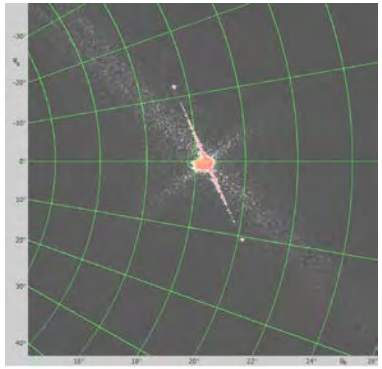
Robotic scatter sensor

- Automatic mapping of freeform surfaces based on CAD data
- Fast and robust CMOS-based scatter sensor for rapid BRDF imaging
- Roughness measurement and defect classification from scatter data

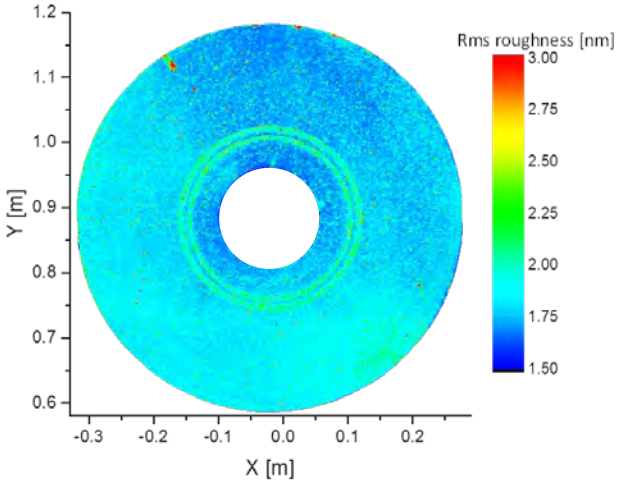
→ **Full area inspection of optical surfaces**



Scatter image



Roughness and defect map



T. Herffurth, S. Schröder, M. Trost, A. Duparré, A. Tünnermann, Applied Optics (2013)

Patent: Patent: DE102009036383B3 (2009)

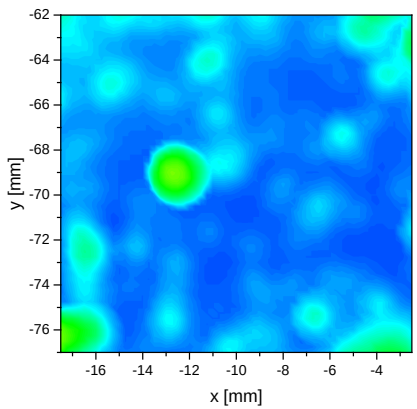
Roughness vs. defects

Defect-induced scatter does depend on beam size

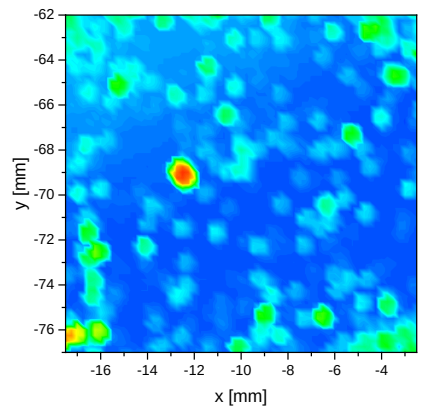
Example: Scatter map of Al mirror at 650 nm

Total scatter
(rms roughness [nm])

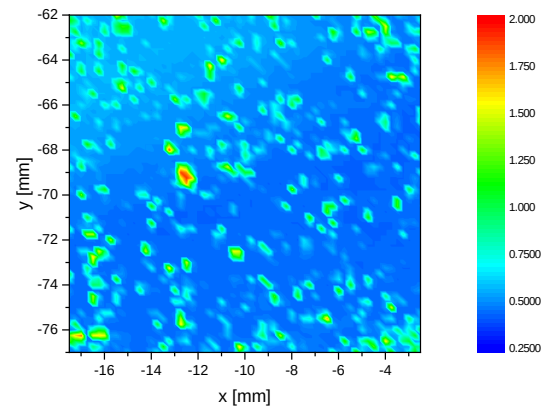
beam diameter d
3 mm



1 mm



200 µm



Closer to application

Higher resolution / sensitivity

Prediction of scattering at application beam diameter *D* from scattering measured at smaller beam diameters *d* (single defect):

$$ARS(D) = ARS_r(d) + ARS_d(d)\left(\frac{d}{D}\right)^2$$

Thin film coatings

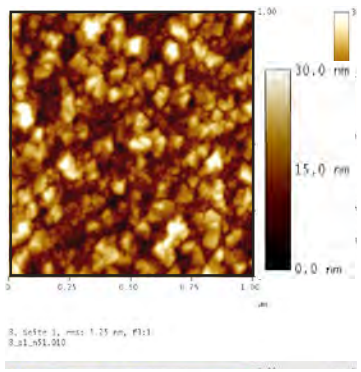
HR coating at 193 nm

HR 193 nm, $(AlF_3/LaF_3)^{20}$ on fused silica (thermal boat evaporation)

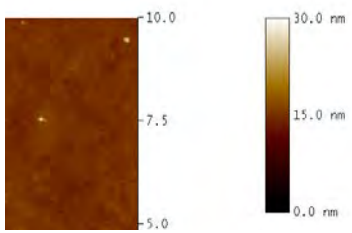
Surface roughness

(AFM 1x1 µm²)

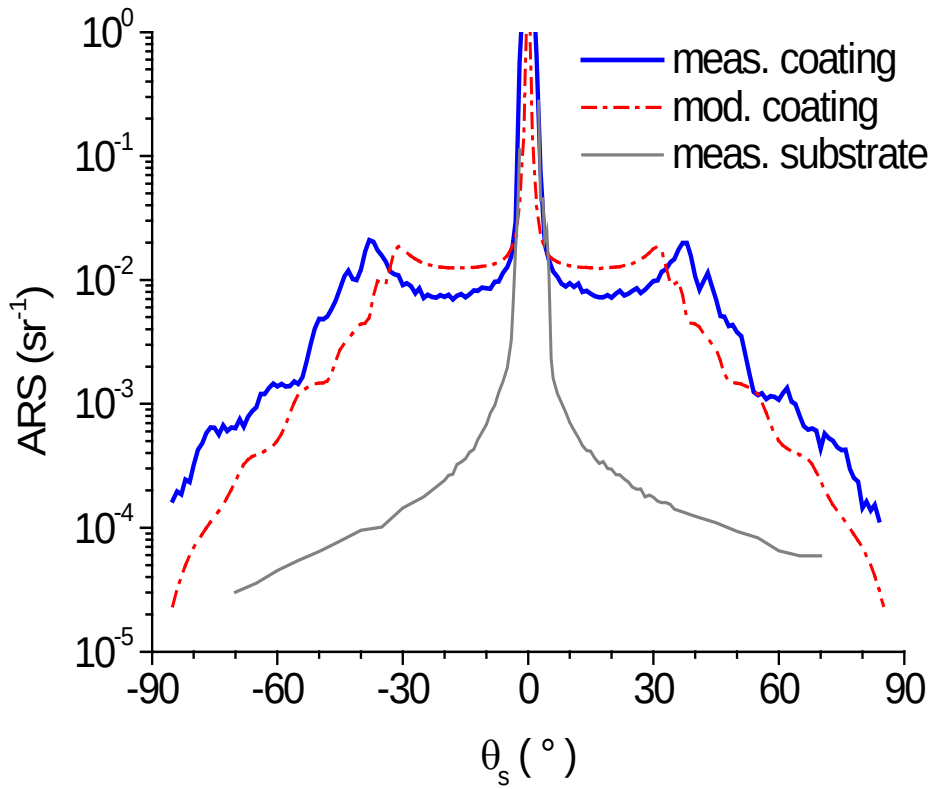
coating
σ = 2.2 nm



uncoated
substrate
σ = 0.2 nm



ARS measurement and initial modeling



TS increases from 0.11% (substrate) to 2.8% (coating)

Reasons: a) Increased σ , b) enhanced R , c) ???

S. Schröder, A. Duparré, A. Tünnermann, Appl. Opt. 47 (2008)

Fraunhofer IOF

HR coating at 193 nm - modeling

Simplified scatter model

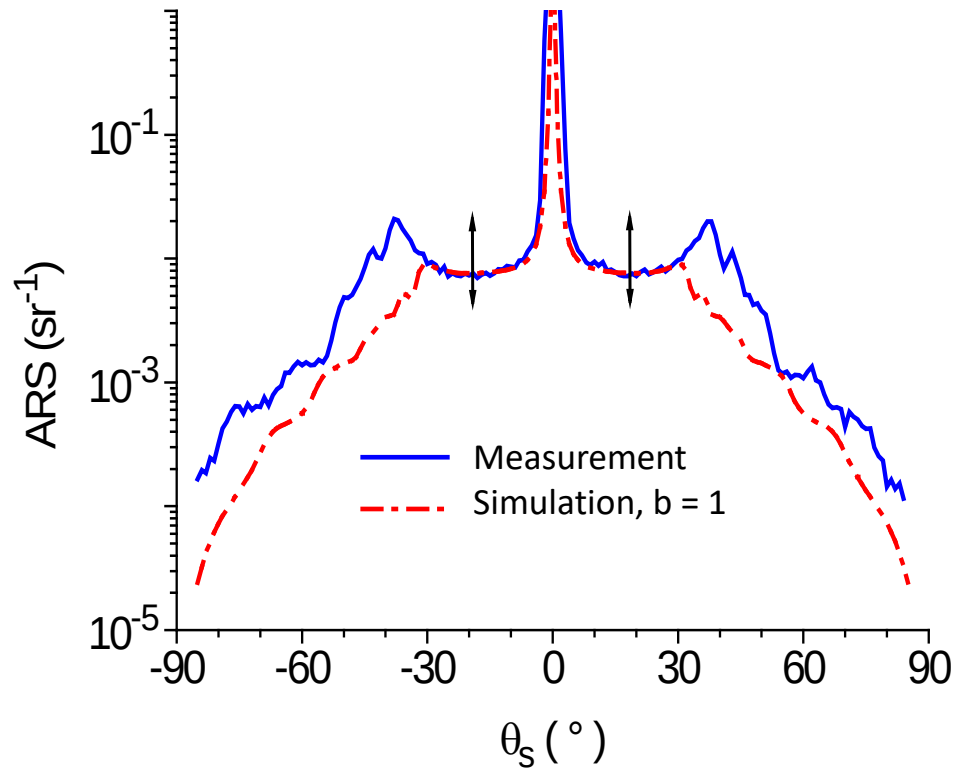
$$\mathrm{ARS}(\theta_s) \sim \sum_{i=0}^{N}\sum_{j=0}^{N} F_i F_j^{*}\, \underbrace{\mathrm{PSD}_{ij}(f)}_{\beta}$$

β

roughness evolution

$$\sigma_i \sim i^{\beta}$$

Variation of β



→ Rapid roughening

Underlying interfaces are much smoother than top-surface

S. Schröder, T. Herffurth, H. Blaschke, and A. Duparré, Appl. Opt. 50 (2011)

HR coating at 193 nm - modeling

Simplified scatter model

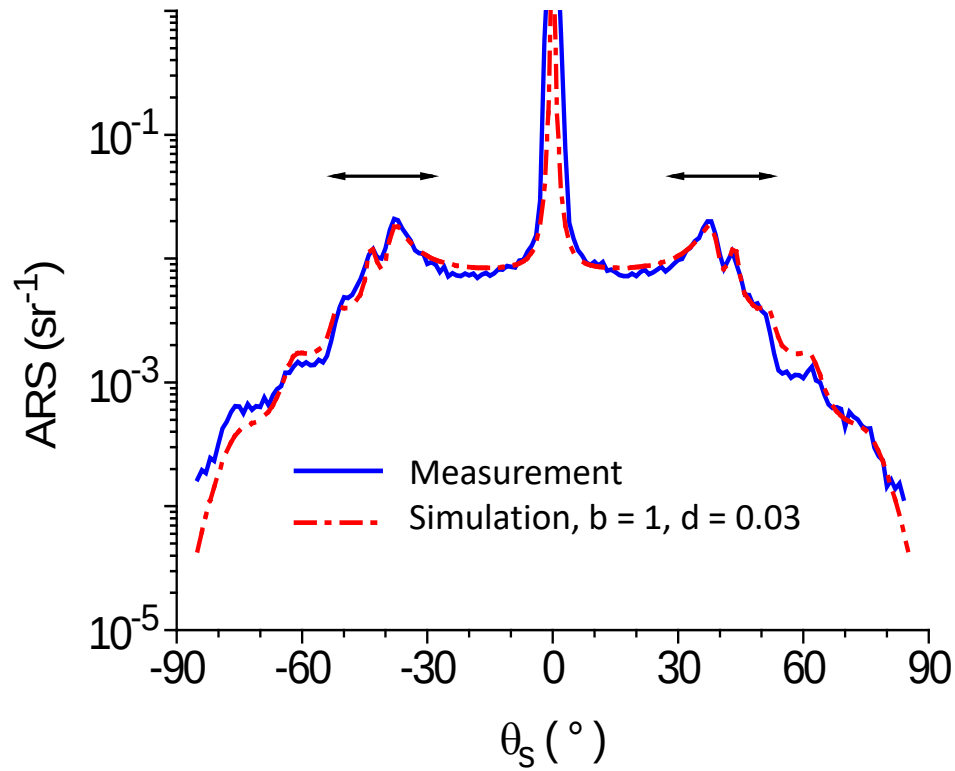
$$\mathrm{ARS}(\theta_s) \sim \sum_{i=0}^{N}\sum_{j=0}^{N} \underbrace{F_i F_j^{*}}_{} \,\mathrm{PSD}_{ij}(f)$$

δ

thickness deviations

$$OT' = (1+\delta)OT$$

Variation of δ



→ **Enhanced scatter caused by film thickness deviations**

→ **Indicates enhanced fields, linked to laser stability**

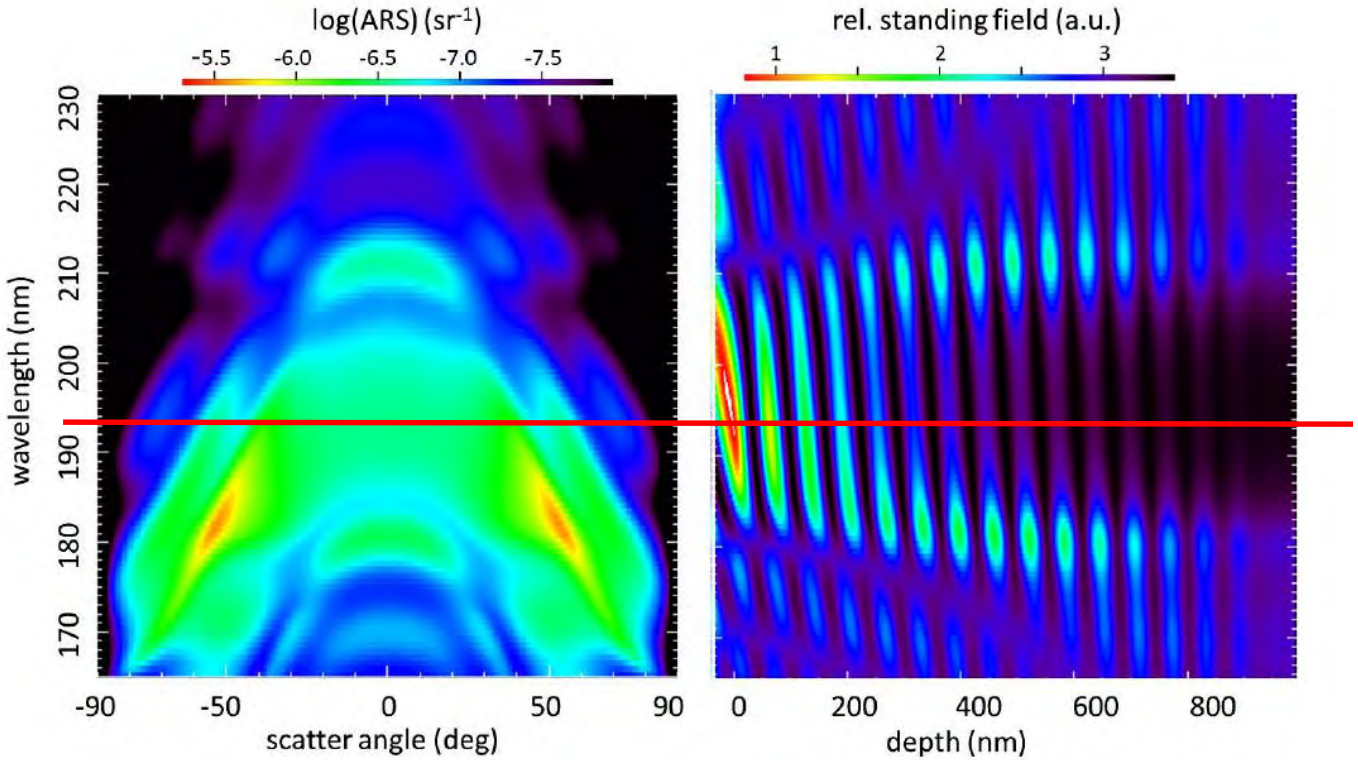
S. Schröder, T. Herffurth, H. Blaschke, and A. Duparré, Appl. Opt. 50 (2011)

H. A. Macleod, J. Vac. Sci. Technol. A 4, 418–422 (1986)

Light scattering depends on roughness and field distribution

Spectral angle resolved scattering (modeling)

Spectral field distribution (modeling)



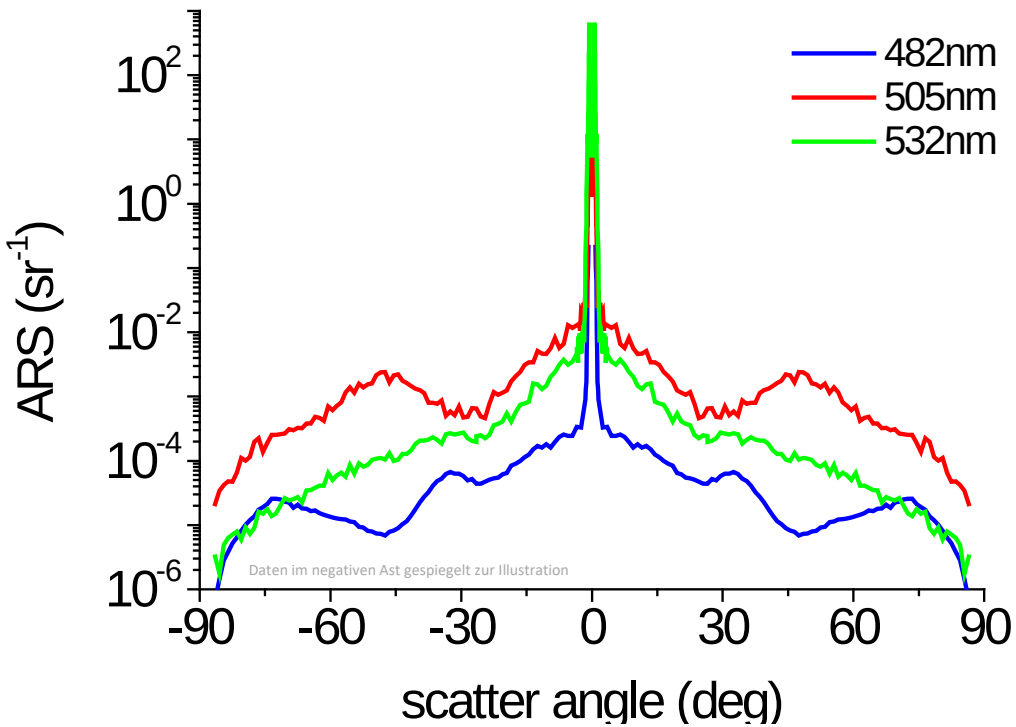
→ **Resonant scatter indicates enhanced fields**

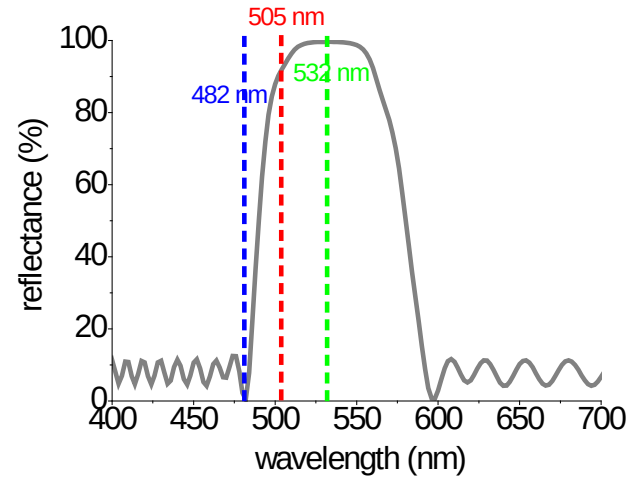
→ **Also correlated with laser stability**

Spectro-Scat: Rugate filter (continous index profile)

Sample: HR 532 nm Rugate, Fraunhofer FEP

ARS at different wavelengths (VIS)



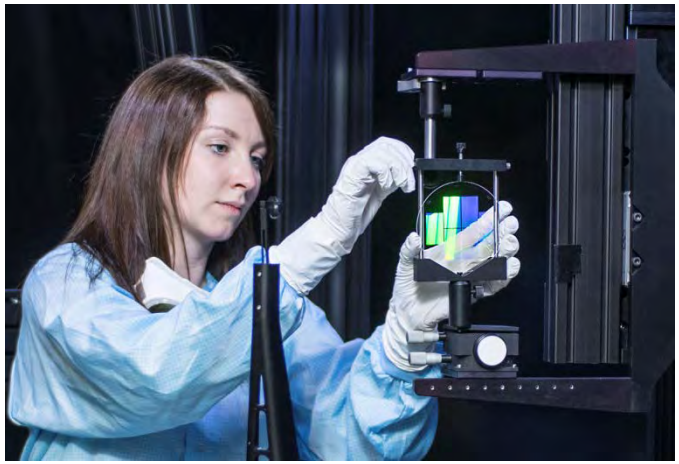


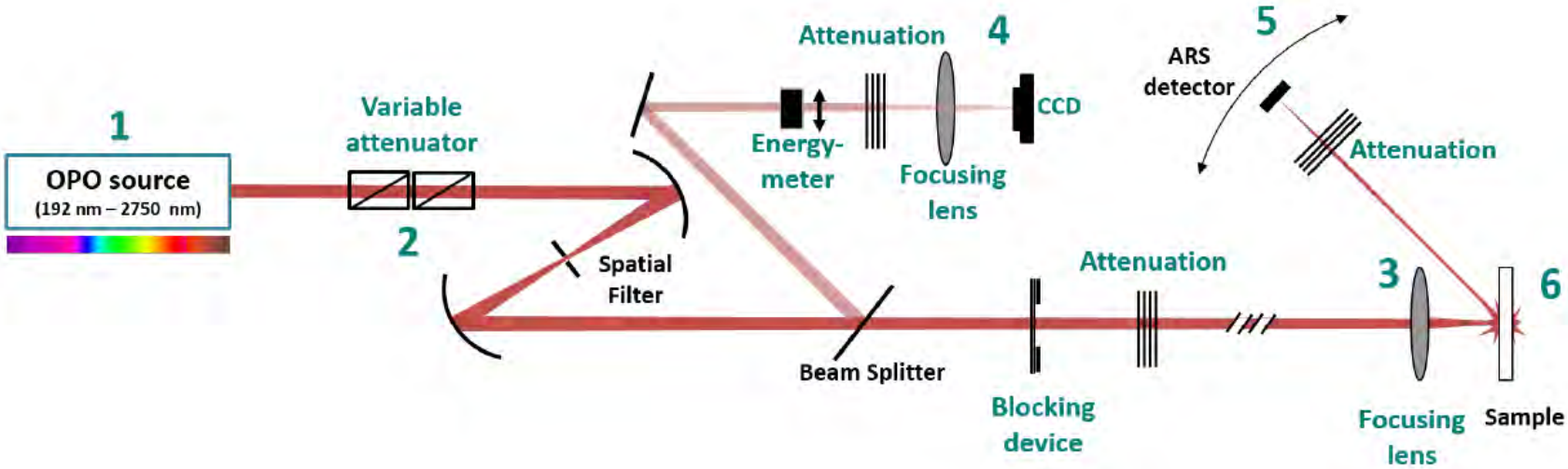
→ **Scatter loss increases from 0.1% at 532 nm to 2.2% at 505 nm!**

→ **Critical effect for spectral filters, spectral shifts, …**

New: Spectral laser-induced damage testing

- Implementation of procedure for laser-induced damage testing into OPO-based scatterometer
- Spectral LIDT testing (ISO 21254), 5 ns pulses,
  Spectral range: 192 nm - 2600 nm,
  Max. fluence: ~10^3 J/cm²
- On- and offline monitoring of structural and material properties using angle-resolved scattering (ARS)
- Combination of (pre-damage) scatter/defect mapping with damage testing





S. Schröder, D. Unglaub, M. Trost, X. Cheng, J. Zhang, and A. Duparré, "Spectral angle resolved scattering of thin film coatings," Appl. Opt. 53, A35-A41 (2014).

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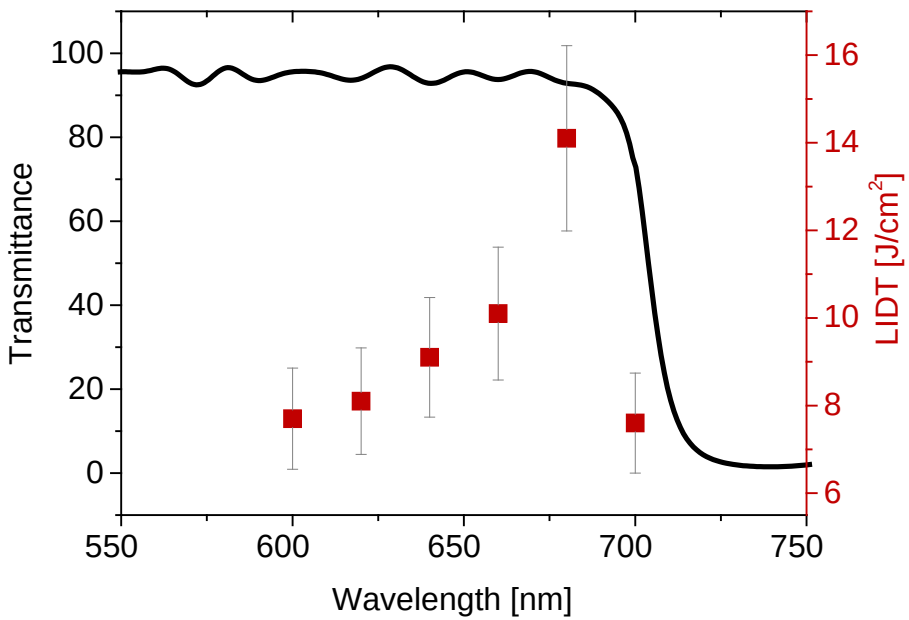
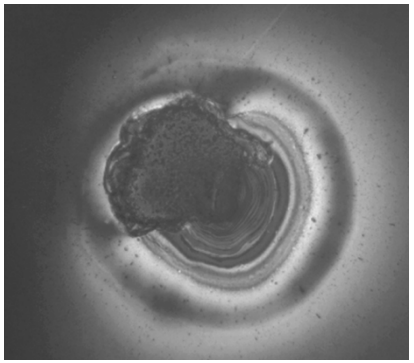
Examples of spectral LIDT measurements

Interference coating



WLI 10x

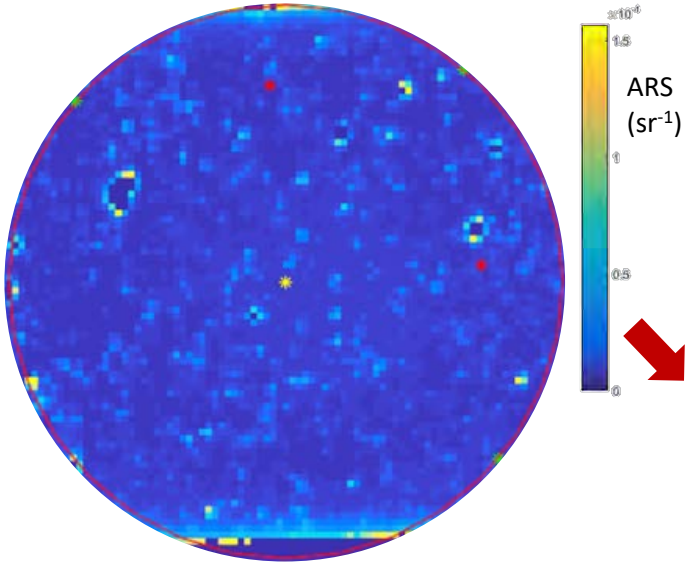


- NIR edge filter (Tongji University, Shanghai)
- Strong variation of LIDT, not necessarily according to simple scaling laws
- LIDT closely related to field distribution and defect locations

S. Schröder, M. Garrick, S. Wilbrandt, O. Stenzel, and A. Duparré, "Spectral Damage Testing of Thin Film Coatings," in Optical Interference Coatings 2016, OSA Technical Digest (online) (OSA, 2016), paper WD.7.

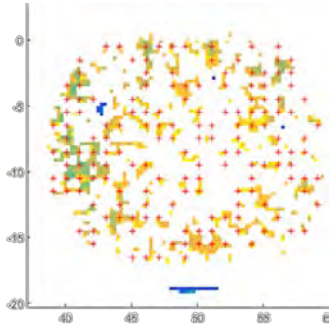
Selective laser damage test

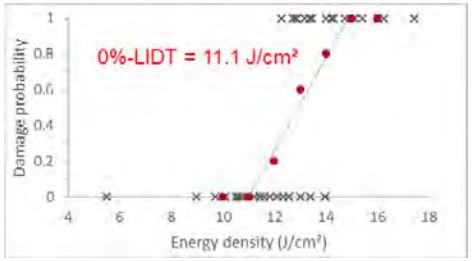
Scatter mapping
(532 nm, AOI 0°,
scatter angle 10°)
before damage test



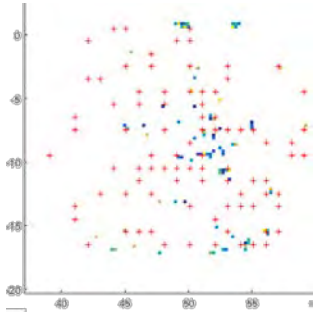
Sample:
HR 1064 nm +
532 nm (Optics
Balzers Jena)

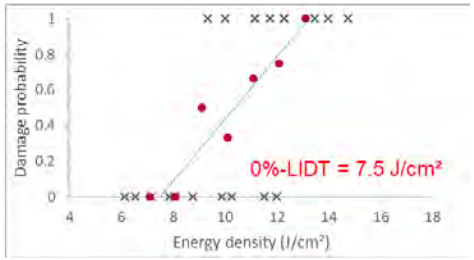
161 defect-free positions





78 defect positons





S-on-1 damage test at 532 nm
(S = 3000, 20 Hz, 5ns, 170 µm)

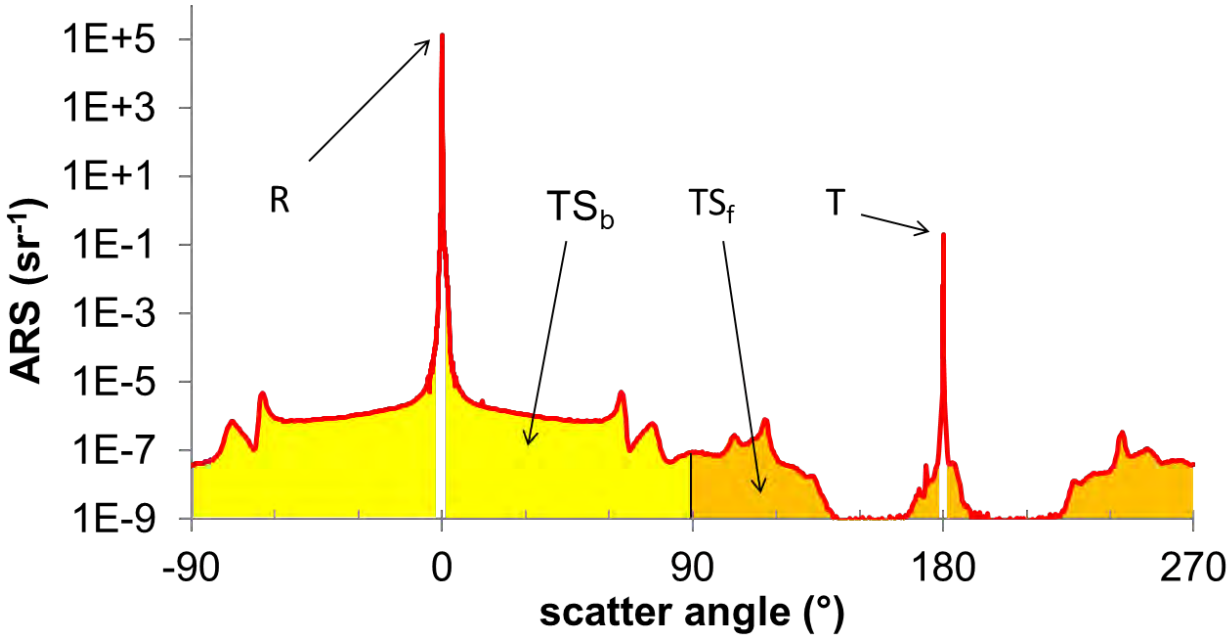
→ 33% lower LIDT on defect locations

→ Additional influence caused by nanodefects not detected by light scattering

Loss analysis using scatterometry

Angle Resolved Scatter measured at 640 nm of HR coating designed for 630 nm - 640 nm

$$1 = R + T + TS_b + TS_f + A$$



R = 0.99… ±0.006

T = $(1.5\pm0.1)\cdot10^{-6}$

TS_b = $(5.9\pm0.6)\cdot10^{-6}$

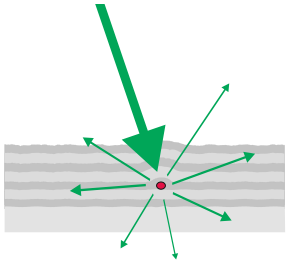
TS_f = $(5.6\pm0.6)\cdot10^{-7}$

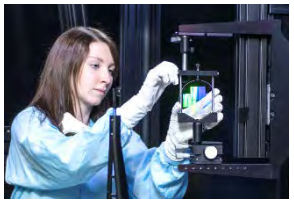
→ Less layers for higher R!

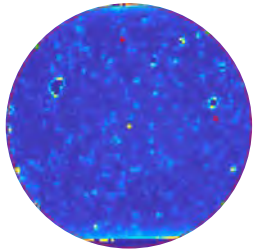
A known → R with ppm precision

R known → A with ppm precision (at low fluence)

Summary







- Manufacturing of high-end optical coatings requires controlling imperfections (roughness, substrate and coating defects, …)
- Collaboration with partners and own expertise
- Angle resolved light scattering to characterize imperfections:
  - Of substrates before coating (roughness, defects, contaminations, …)
  - Of coatings (roughness evolution, thickness errors, …)
  - Of Materials (inhomogeneities, defects, …)
- Significant impact of defects onto LIDT shown by selective laser damage testing using combination of scattering and damage test
- Highly sensitive loss analysis using angle resolved scatterometry

Thank you !

Merci – Grazie – Danke ;)

Thanks to colleagues at IOF: Marcus Trost, Matthias Hauptvogel, Tobias Herffurth, Alexander von Finck, Méabh Garrick, Luisa Coriand, Nadja Felde, Angela Duparré, Andy Tänzer, Ronald Schmidt and mechanical workshop, Thomas Ganz, Beate Wendt, …

As well as partners: Optixfab, Opcrown, Lenstec, Optics Balzers Jena, Lasos, IPHT, LZH, …

Financial support: EU / TAB project SpectroScat, BMBF ZIM project LOSASS