Overview of ALD-Activities for Optical Applications: Materials, Refractive and Diffractive Optics

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Workshop Optical Coatings for Laser Applications 2016 - Buchs

Outline

- Introduction
- Coating equipment
- Results
  - Materials
  - Interference Coatings
  - Diffractive Optics
- Outlook

Dr. Adriana Szeghalmi

- **Professional career since final degree**
- 10/2015 –to date — Group Manager ALD for Optics FhG-IOF Jena
- 05/2010–to date — Emmy Noether Group Leader FSU Jena
- 05/2007–04/2010 — Postdoc, Max Planck Institute of Microstructure Physics (Prof. U. Gösele, Dr. M. Knez) Halle (Saale)
- 04/2005–04/2007 — Postdoc, University of Manitoba, Canada (Prof. K. M. Gough) Winnipeg

- **University education**
- 07/2001-02/2005 — PhD studies, University of Würzburg (Prof. W. Kiefer)
- 04/2000-03/2001 — Exchange student at the University of Würzburg
- 10/1997-06/2001 — Study of Chemistry and Physics, Babes-Bolyai University, Romania

ALD Team

Lilit Ghazaryan
Kristin Pfeiffer
Vivek Beladiya
Svetlana Shestaeva
Astrid Bingel
David Kästner

Alumni: M. Sc. Haiyue Yang
Dr. Pascal Genevée
M. Sc. Ernest Ahiavi
Dr. Stephan Ratzsch

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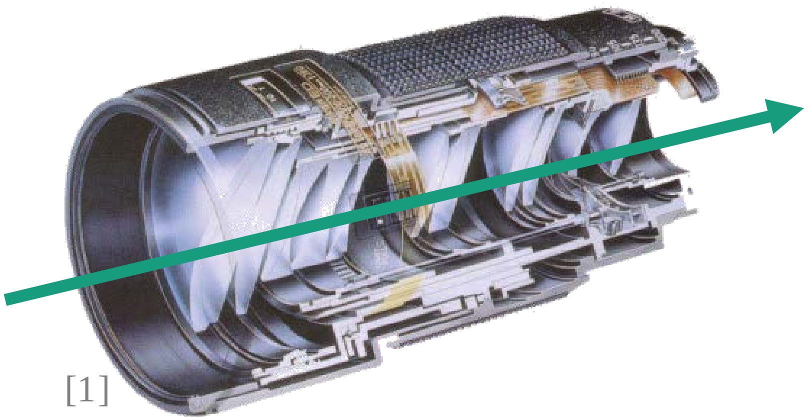
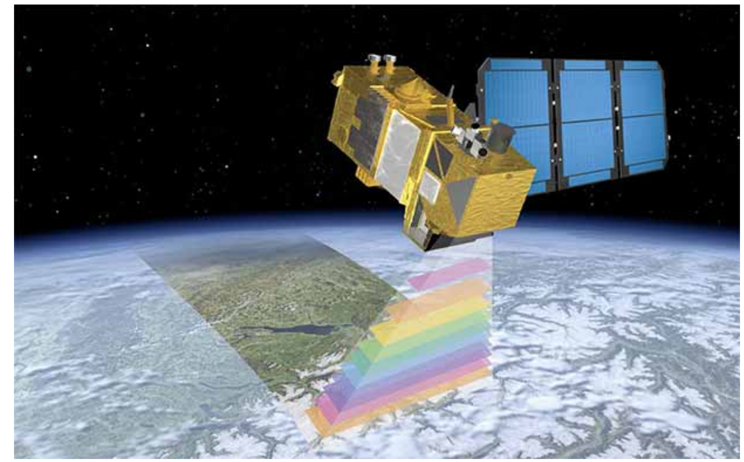
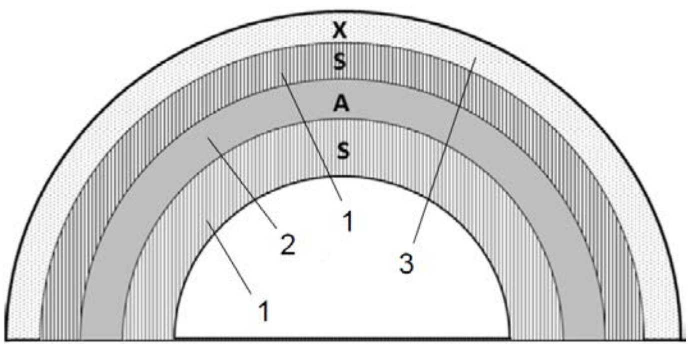
The ALD Solution to Enhance Optical Performance

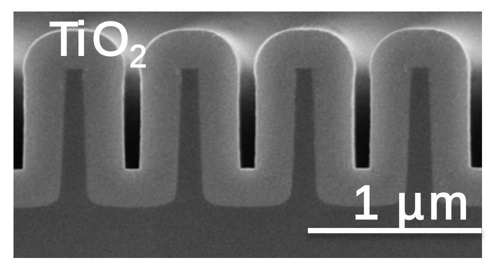


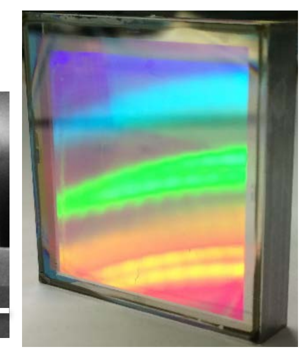
[1]

ESA, Sentinel-2



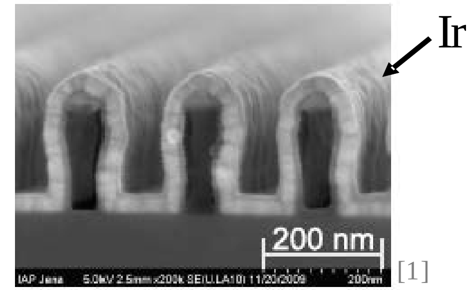






[1] www.marquardt-kempen.de/s/cc_images/cache_2435481376.jpg?t=1362993658
[2] L. Ghazaryan, A. Szeghalmi, E. B. Kley, U. Schulz, DPMA Anmeldung. 2016

Why ALD?



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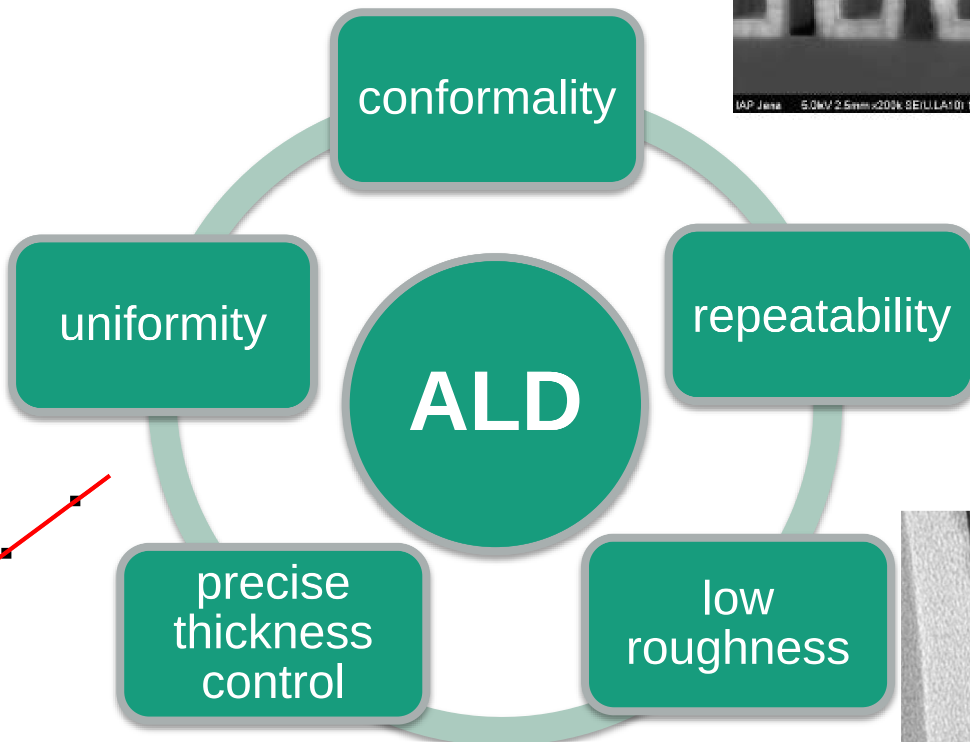

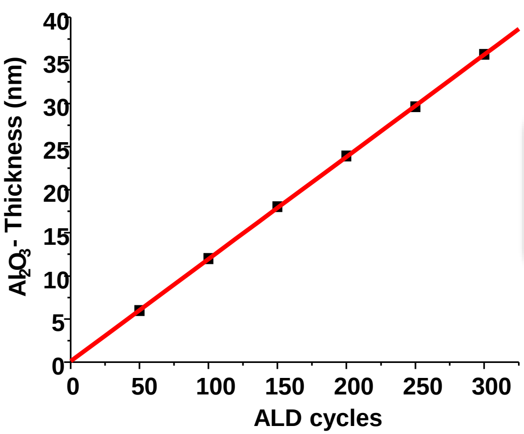

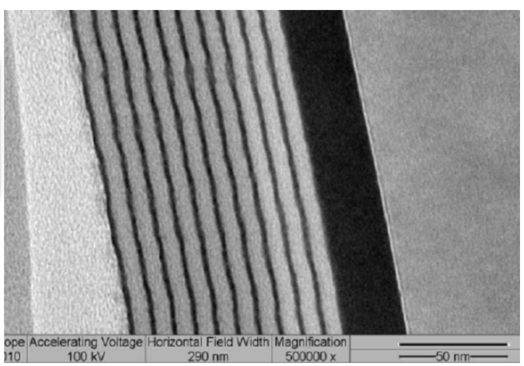

Ta_2O_5 / Al_2O_3 Nanolaminate (2.3 nm / 6.7 nm) x N [2]

[1] T. Weber, T. Käsebier, A. Szeghalmi, M. Knez, E. B. Kley, A. Tünnermann. Nanoscale Research Lett. 6 (2011) 558
[2] A. Szeghalmi et al., Applied Physics Letters, 2009, 94, 133111-1/3.

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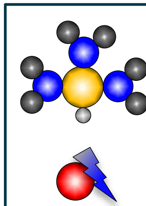
ALD atomic layer-by-layer coating technology

SiO_2 ALD cycle



$HSi(N(CH_3)_2)_3$

O_2 Plasma

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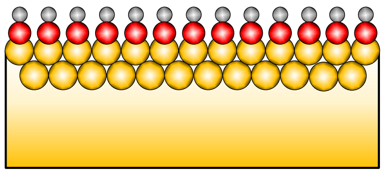
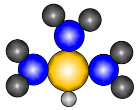
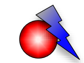
ALD atomic layer-by-layer coating technology

SiO_2 ALD cycle





$HSi(N(CH_3)_2)_3$



O_2 Plasma

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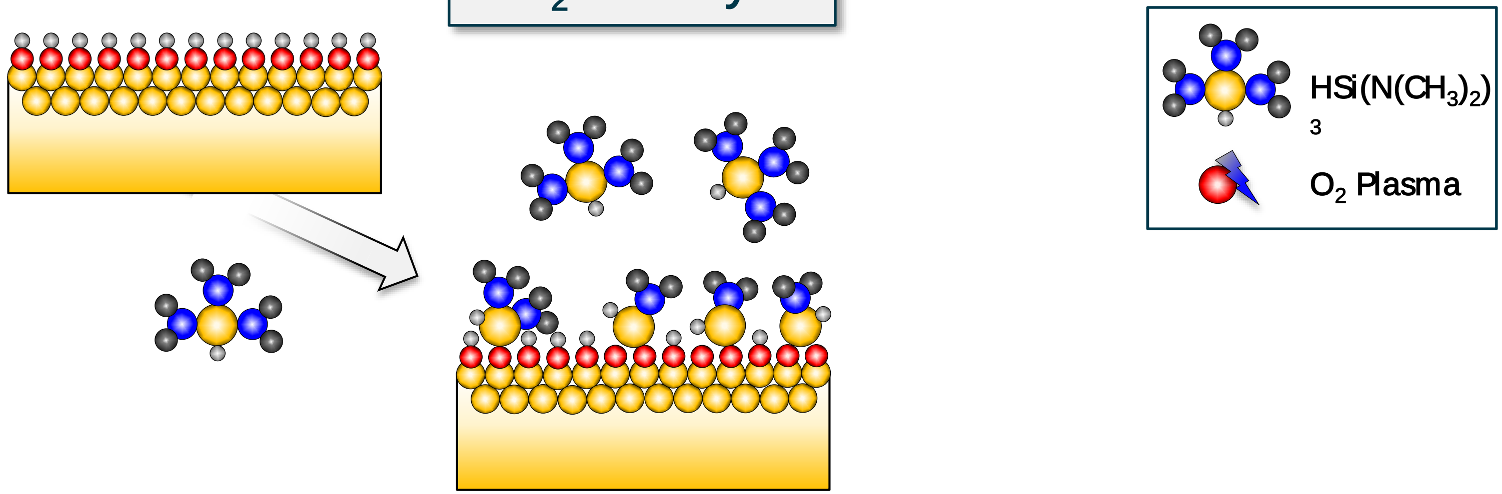
ALD atomic layer-by-layer coating technology

SiO_2 ALD cycle



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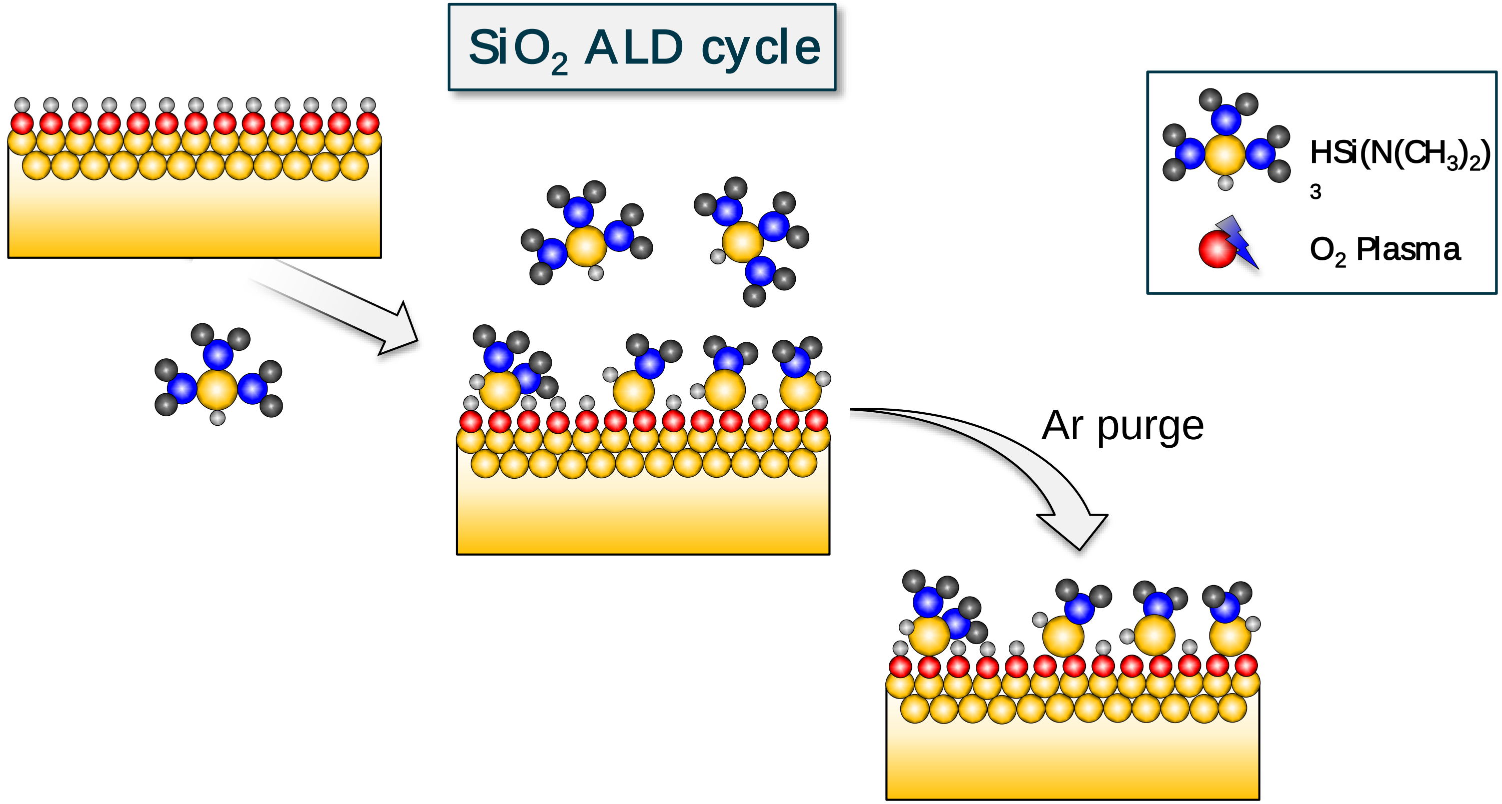
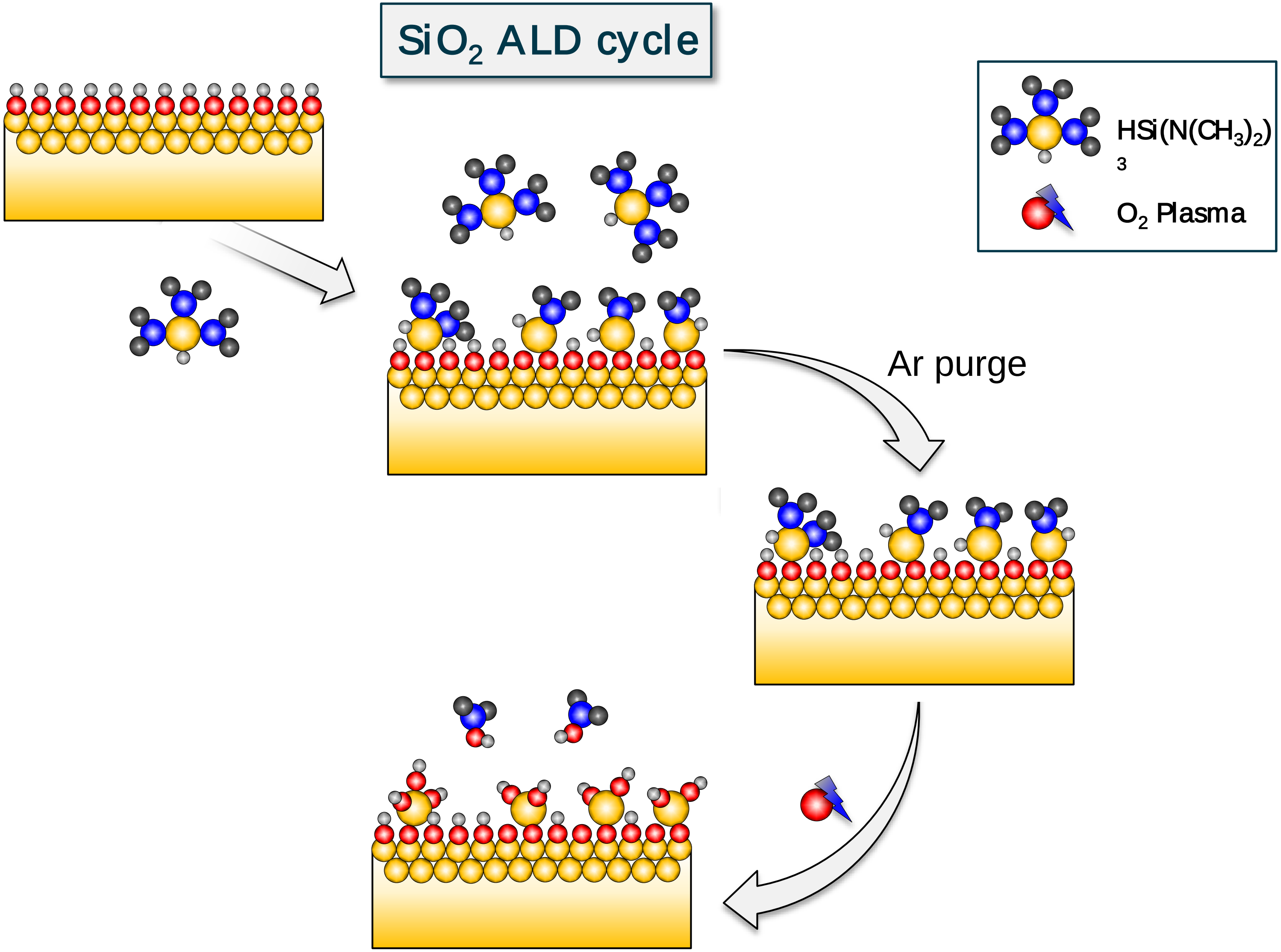
ALD atomic layer-by-layer coating technology



ALD atomic layer-by-layer coating technology



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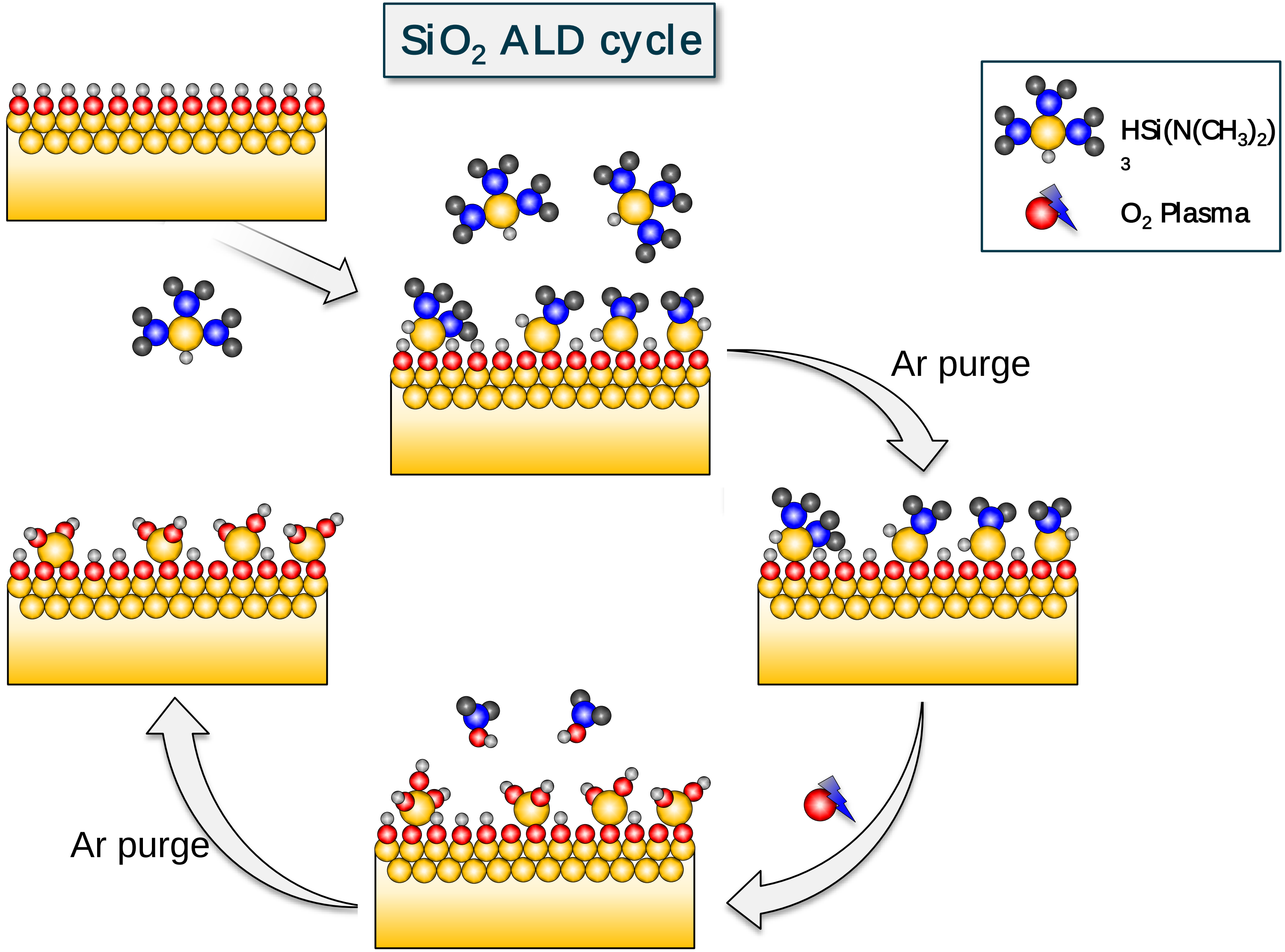
ALD atomic layer-by-layer coating technology



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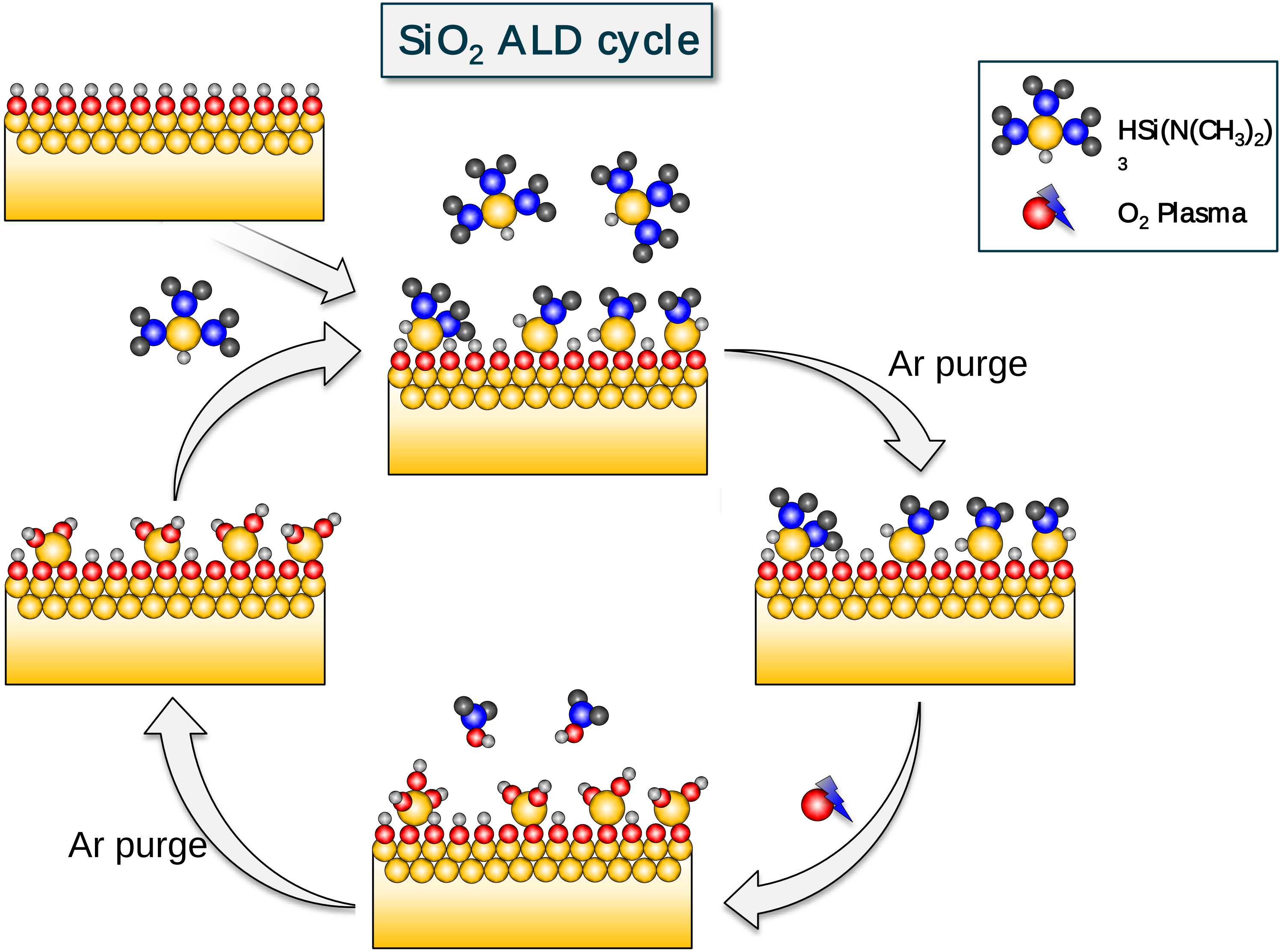
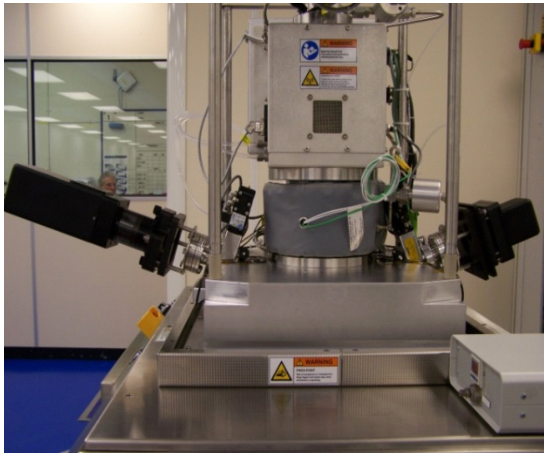
ALD atomic layer-by-layer coating technology



Coating Equipment

1. OpAL PEALD tool

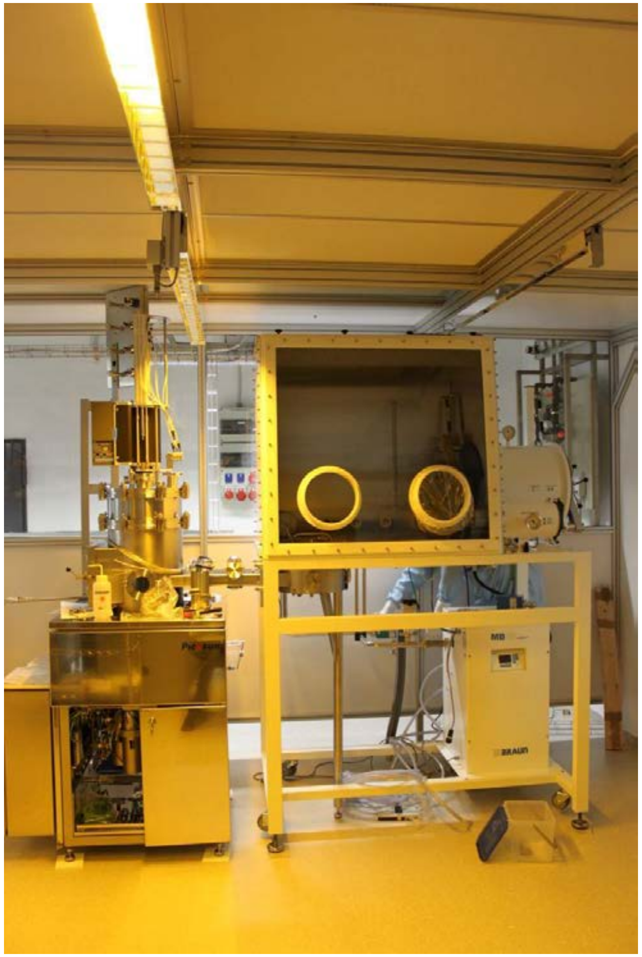
- Oxford Plasma Technologies (UK)
- Open loaded ALD tool
- Top-Flow reactor
- ICP plasma
- Up to 200 mm diameter wafers (150 mm square)
- Height up to 20 mm
- Up to 4 precursors (H_2O + 3 Metal Precursors)
- Temperature range 25°C...200°C
- Ellipsometry ports, Woollam ellipsometer
- ca. 400k€
- Installed 2011
- Oxides



Coating Equipment

2. Sunale R200 PEALD tool

- Picosun Oy (F)
- Load-lock and open loaded ALD
- Hot-wall top-flow dual chamber design
- RF Plasma
- Up to 200 mm diameter wafers (150 mm square)
- Height up to 100 mm
- Up to 6 precursors (H_2O + 5 Metal Precursors)
- Temperature range 25°C...500°C
- Load-lock integrated in GloveBox
- ca. 400k€
- Installed 2013
- Metals and Oxides



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Larger Reactor Configuration

In commissioning

- Diameter 330 mm
- Height up to 100 mm
- Thermal and Plasma Enhanced ALD
- To be installed March 2017 @ IOF Jena

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Outline

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Materials

- development of the thermal and/or PEALD processes
  - SiO_2, Al_2O_3, TiO_2, HfO_2, Ta_2O_5
  - MgF_2, CaF_2, LaF_3
  - iridium
  - alucones & nanoporous Al_2O_3
  - composite materials & nanoporous SiO_2

- literature: Ru, Ag, Au, W, graphene, nitrides, $SrTiO_3$, Nb_2O_5, ZrO_2...

- optical properties
- morphology
- mechanical properties

$$h_c = \frac{\Gamma E_f}{Z\sigma^2}$$

h_c critical coating thickness
E_f Elastic modulus film
Γ fracture resistance
Z geometrical factor (1.976)
σ mechanical stress

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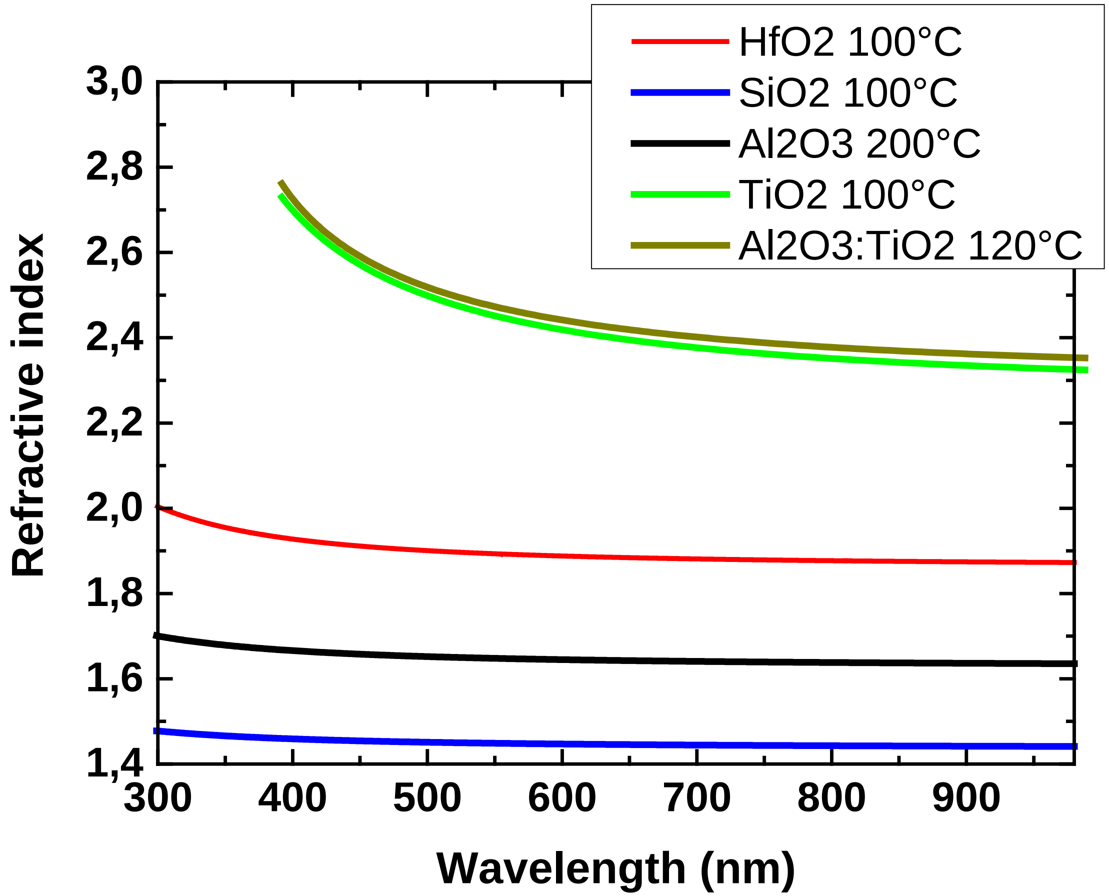
Oxides: Summary of Refractive Index



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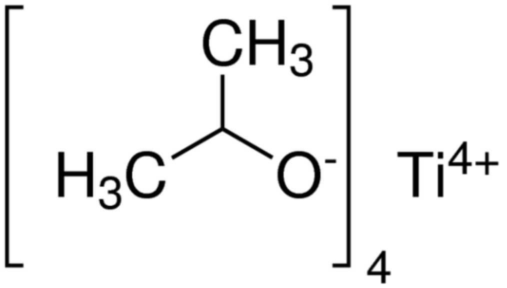
TiO_2

Titanium(IV) isopropoxide + H_2O

$Ti[OCH(CH_3)_2]_4$ H_2O_2

heated and bubbled at 45-60°C O_2 Plasma

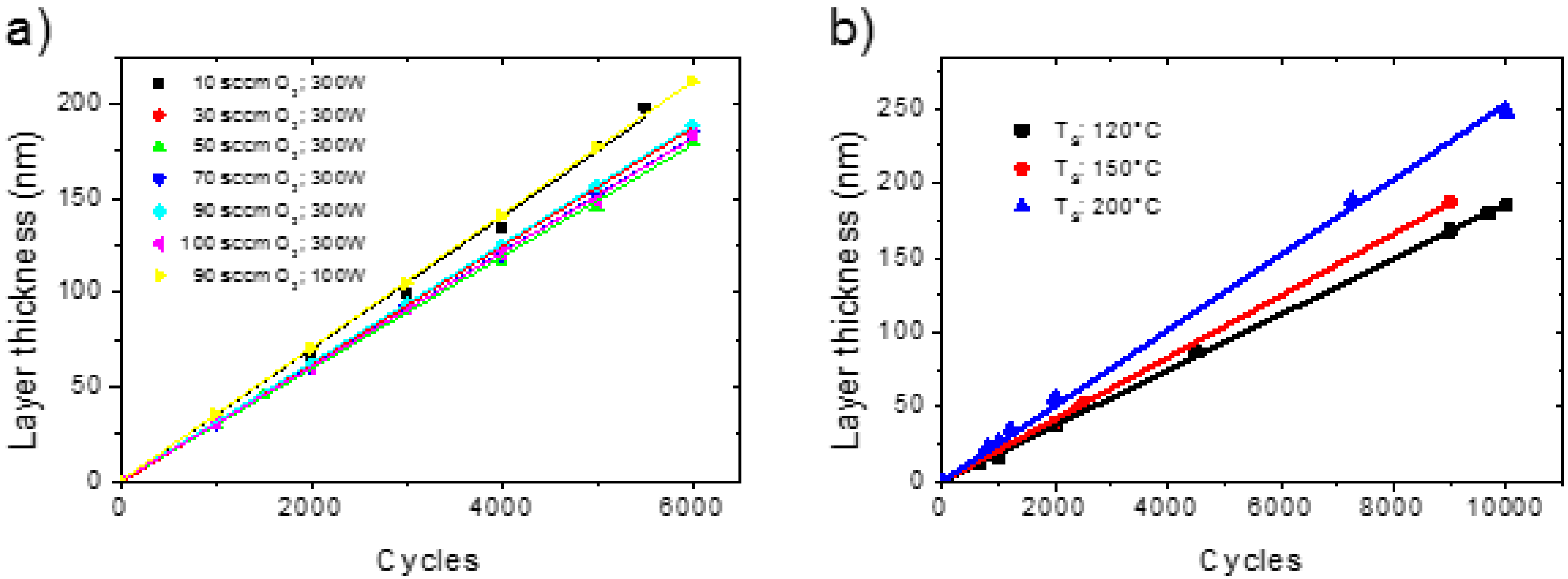


Titanium(IV) chloride + H_2O

$TiCl_4$

TiO_2 Film Growth

- Linear ALD growth with number of cycles
- Influence of Deposition Temperature and O_2-Plasma



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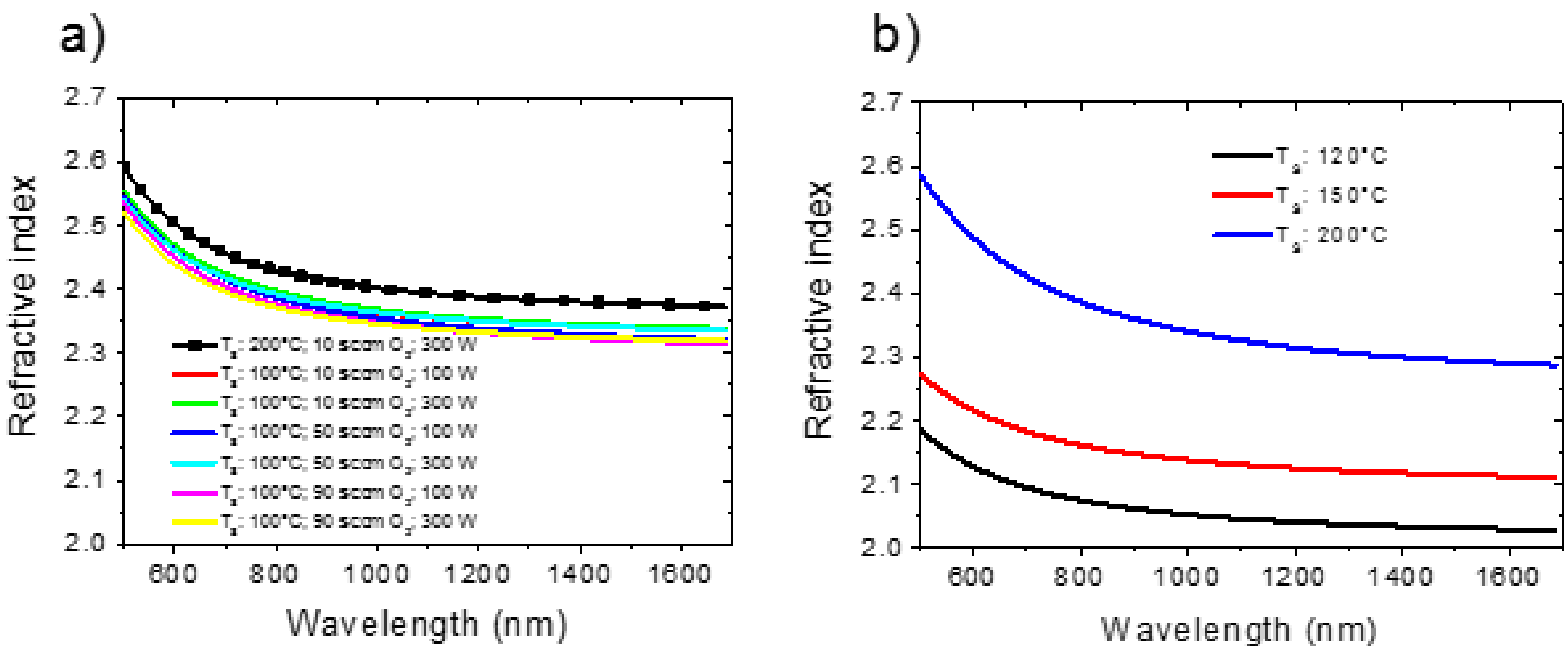
TiO_2 Refractive Index

- PEALD processes provide higher refractive index at lower temperature than the thermal ALD of titania
- Temperature has little influence in PEALD of titania



Δn = ~0.03

Δn = ~ 0.4

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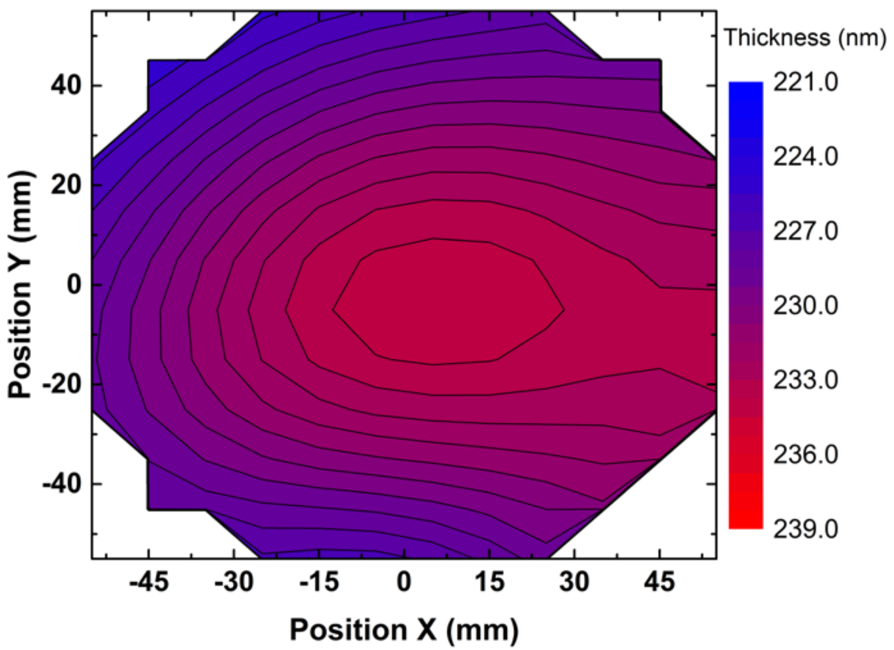
TiO_2 Thickness Uniformity

- TiO_2/Al_2O_3 PEALD 120°C
- Ø 150 mm Si wafer: SD 1 σ = 1.07%
- Homogeneity (Max-Min/2$d_{average}$) = 2.1%

n @633 nm = 2.4232 ± 0.004



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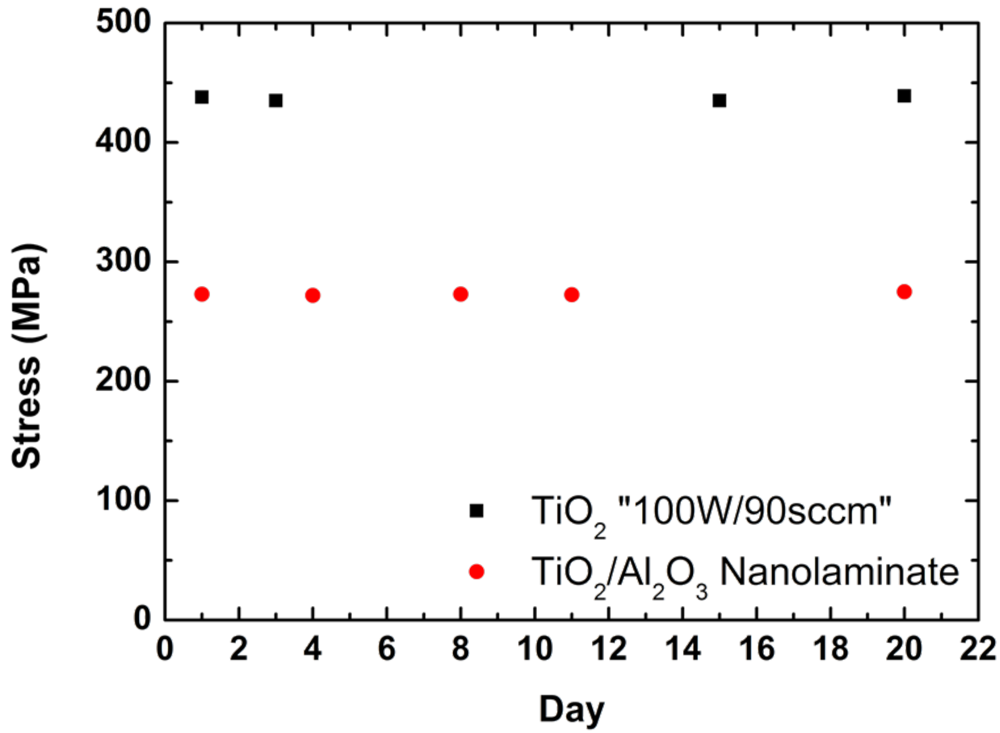
TiO_2 Tensile Mechanical Stress

- σ decreases in nanolaminates



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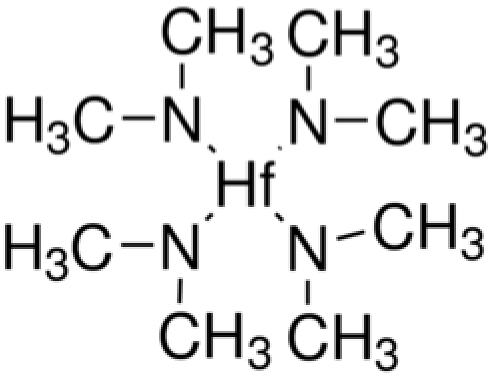
HfO_2

Tetrakis(dimethylamino)hafnium(IV) + O_2 Plasma

$[(CH_3)_2N]_4Hf$

heated and bubbled at 50°C



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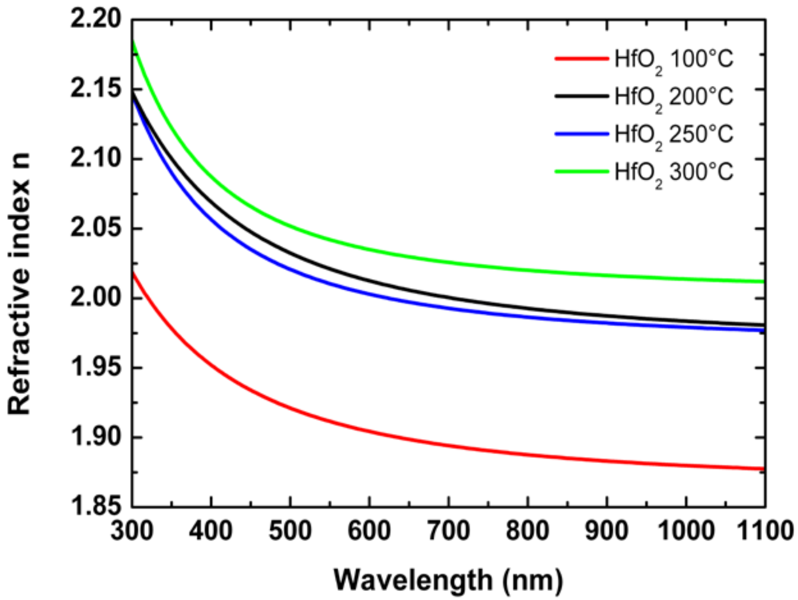
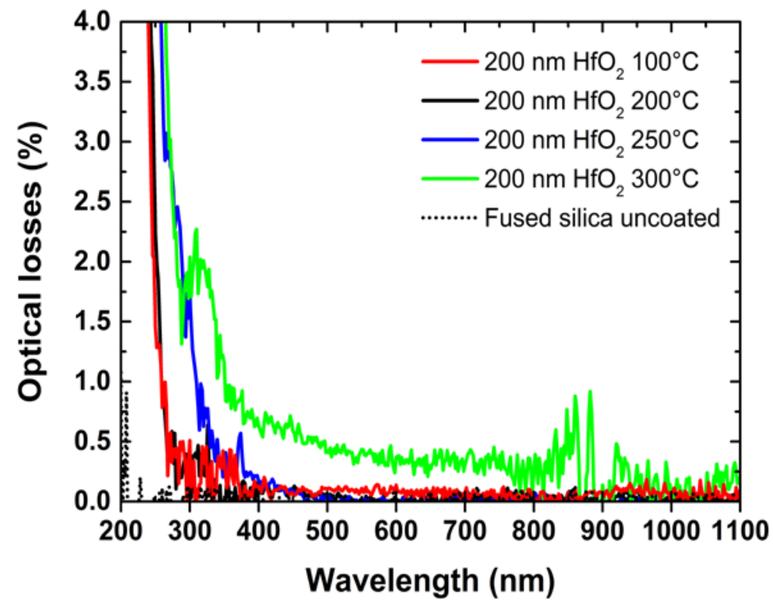
Optical Properties – Influence of Temperature

- high scattering losses above 200°C deposition temperature





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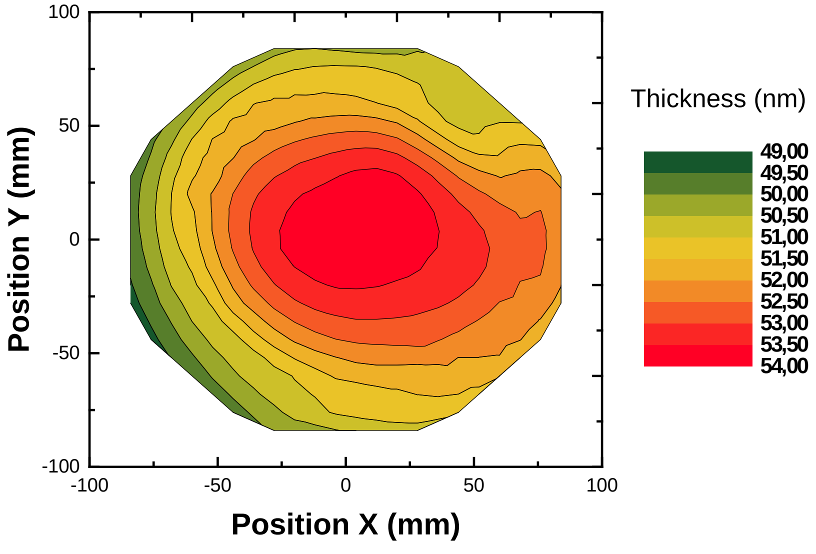
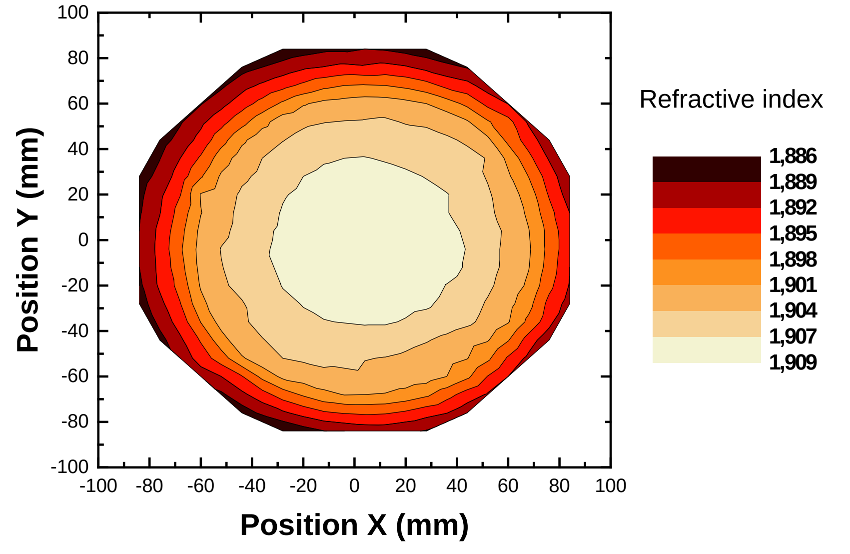
Thickness and Refractive Index Uniformity

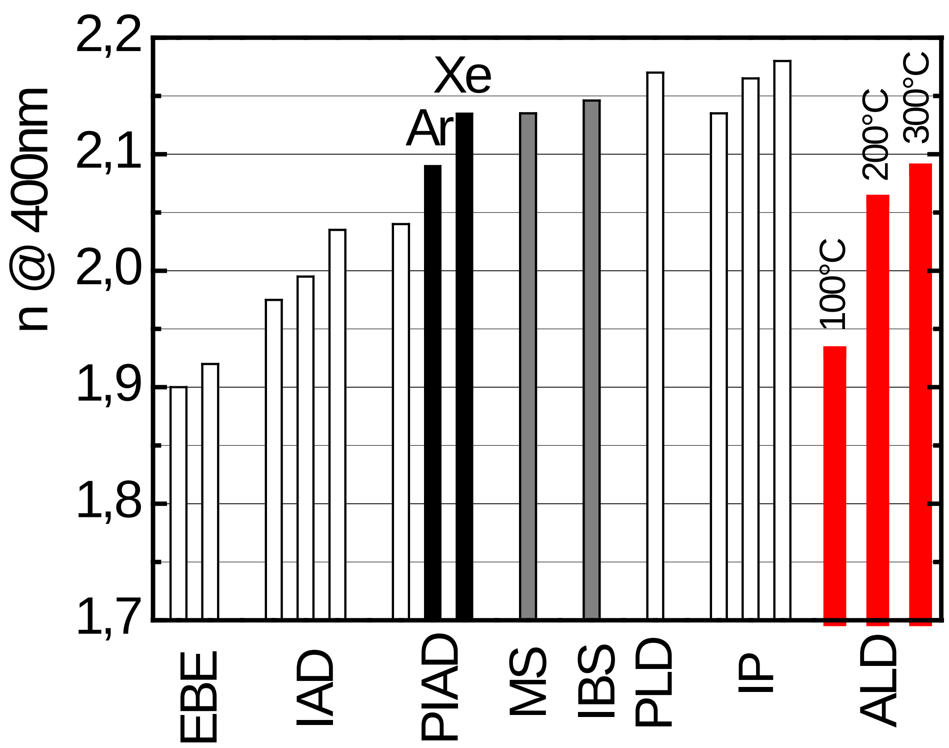
- HfO_2 PEALD 100°C
- Ø 200 mm Si wafer: SD 1 σ = 2.24%
- Homogeneity (Max-Min/2$d_{average}$) = 4.5%
- Plasma flow from the top



n = 1.8999 ± 0.0065



HfO_2 Refractive Index – PVD & ALD



O. Stenzel, S. Wilbrandt, S. Yulin, N. Kaiser, M. Held, A. Tünnermann, J. Biskupek, U. Kaiser, Opt. Mater. Express 2011, 1, 278.

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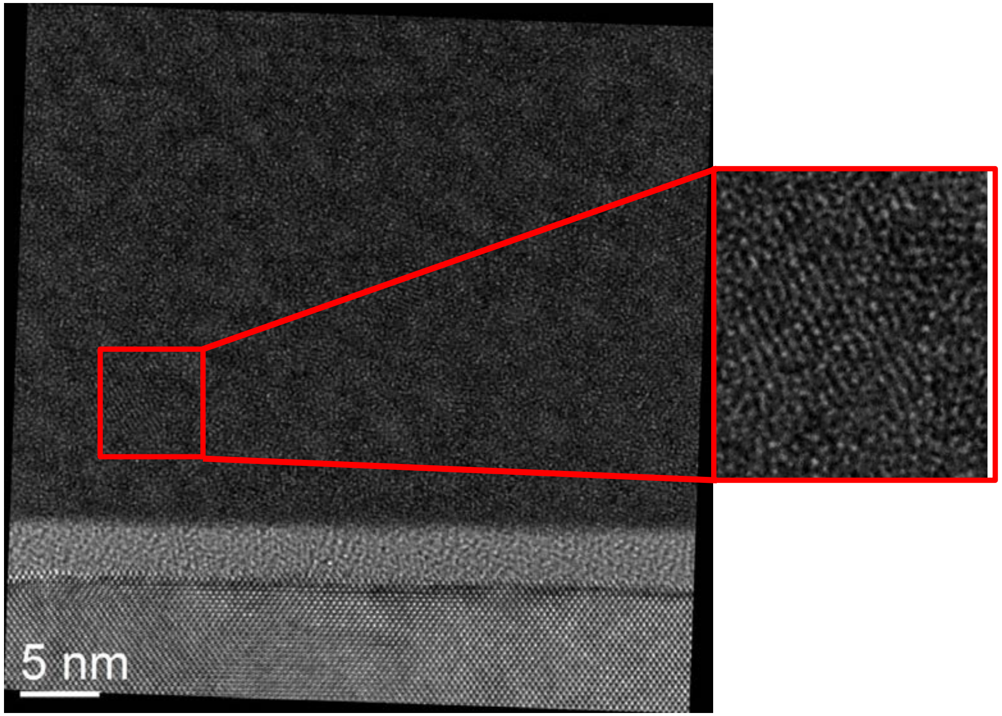
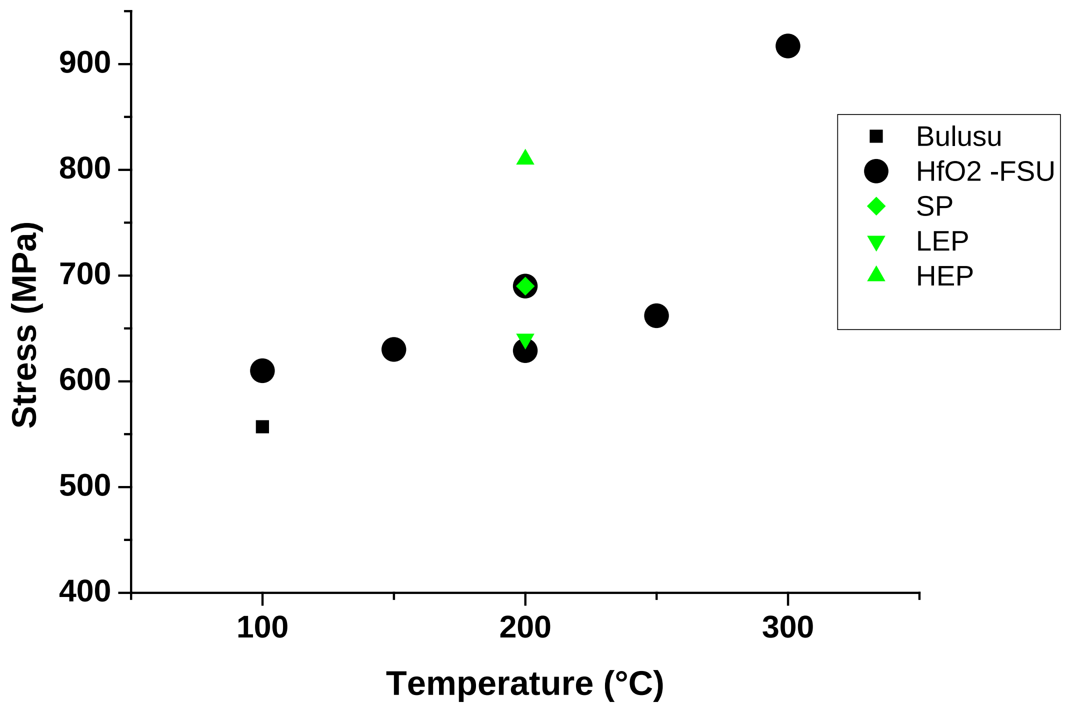
HfO_2 TEM image

- deposited at 100°C
- ca. 5 nm Nanocrystallites
- ca. 3.5 nm HfO_2/SiO_2 composite layer at the Si-wafer interface



HfO_2 Tensile Mechanical Stress

- σ increases with increasing deposition temperature due to crystallization
- σ slightly increases with increasing ion energy



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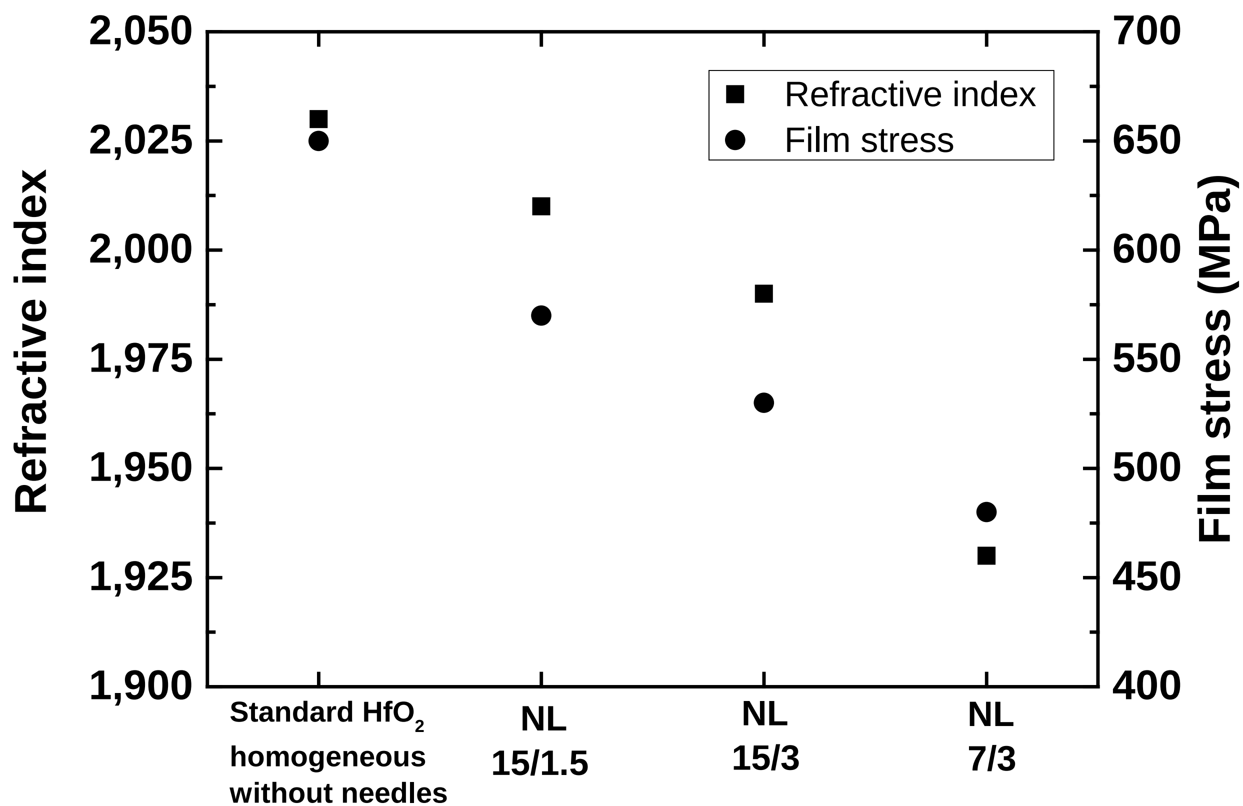
HfO_2 Tensile Mechanical Stress

- σ decreases in nanolaminates (HfO_2/SiO_2 and HfO_2/Al_2O_3)

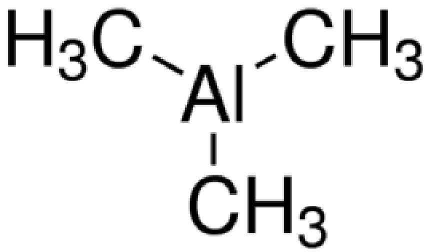


Al_2O_3

Trimethylaluminum + H_2O

$(CH_3)_3Al$ O_2 Plasma

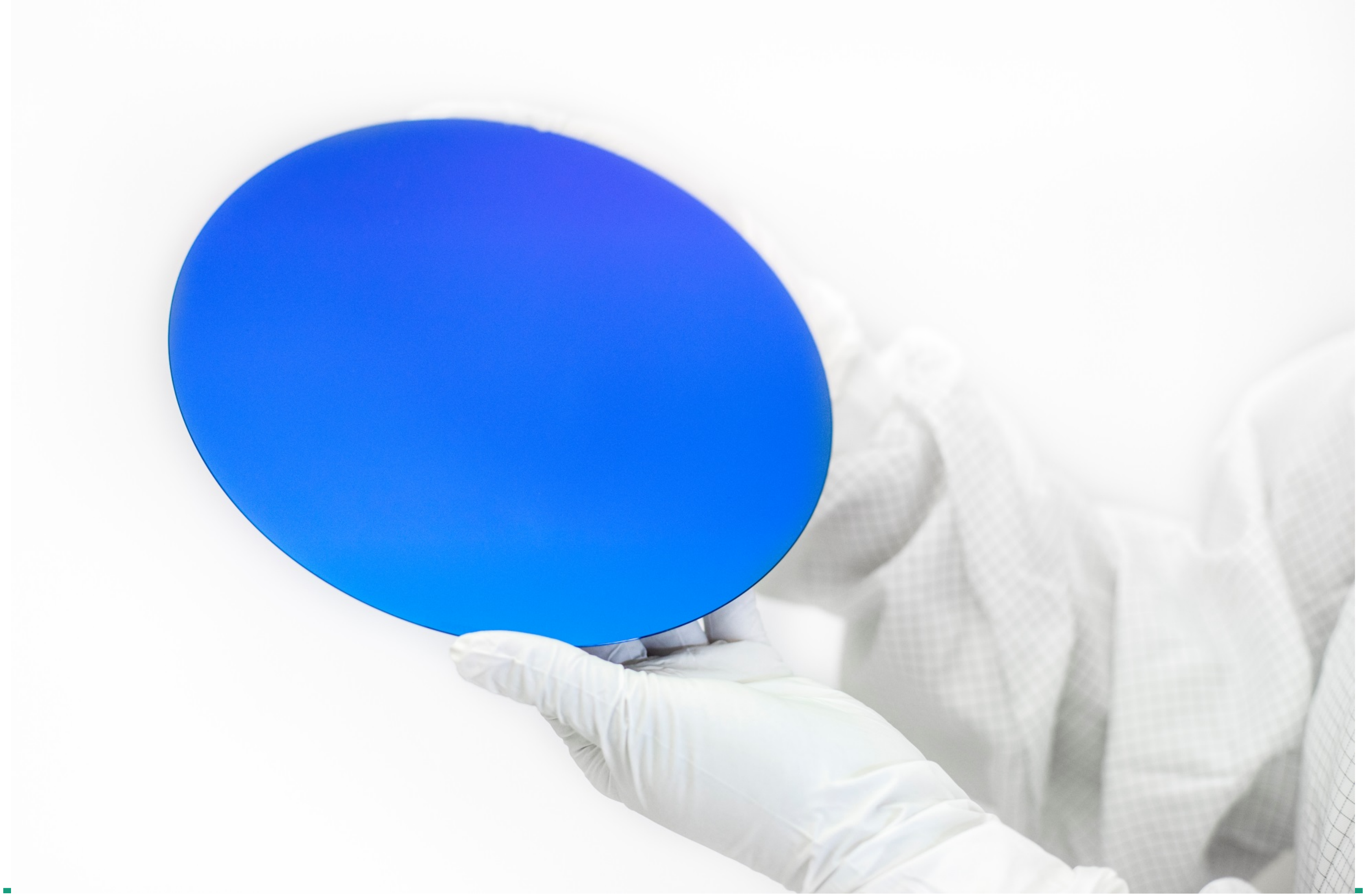
own vapour pressure



Aluminum chloride + H_2O

$AlCl_3$

100 nm Al_2O_3 on 200 mm Si wafer



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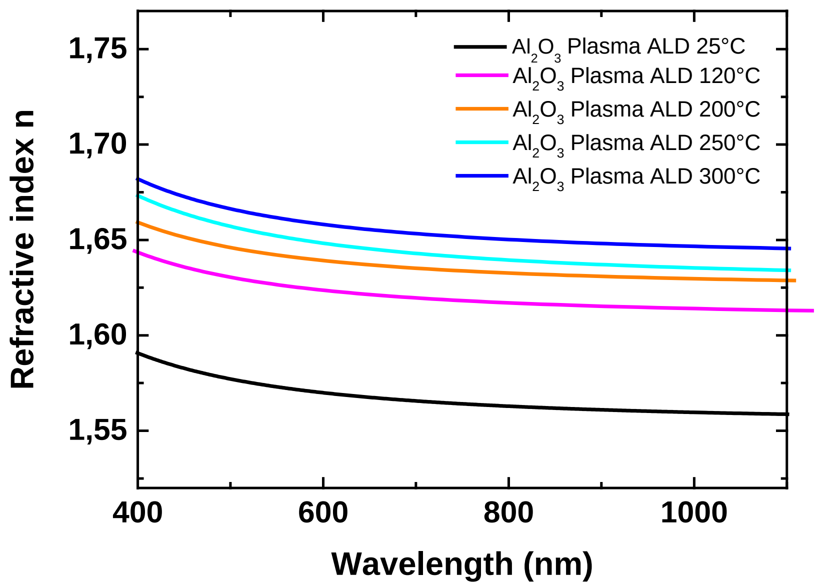
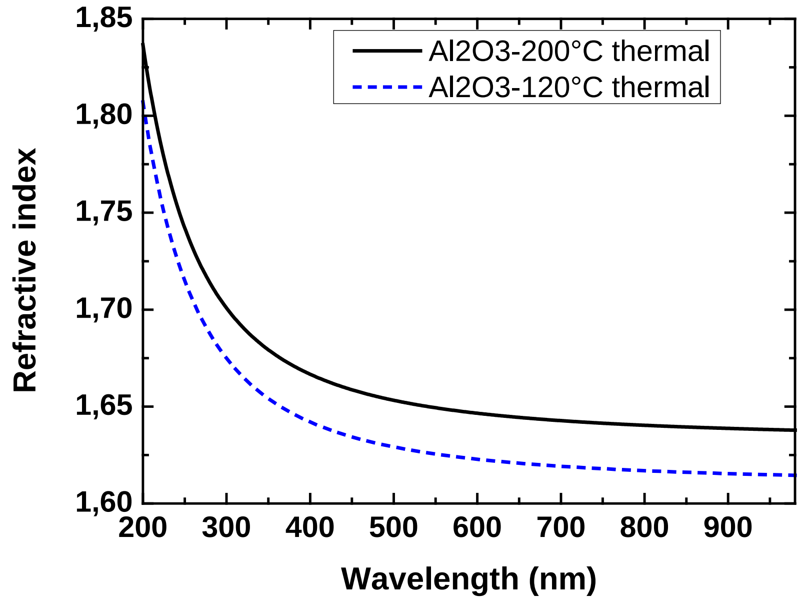
Influence of Deposition Temperature

- impact on reproducibility of refractive index
- temperature has less influence in PEALD processes than in thermal ALD





$\Delta n = 0.015$
within measurement accuracy

$\Delta n = 0.025$

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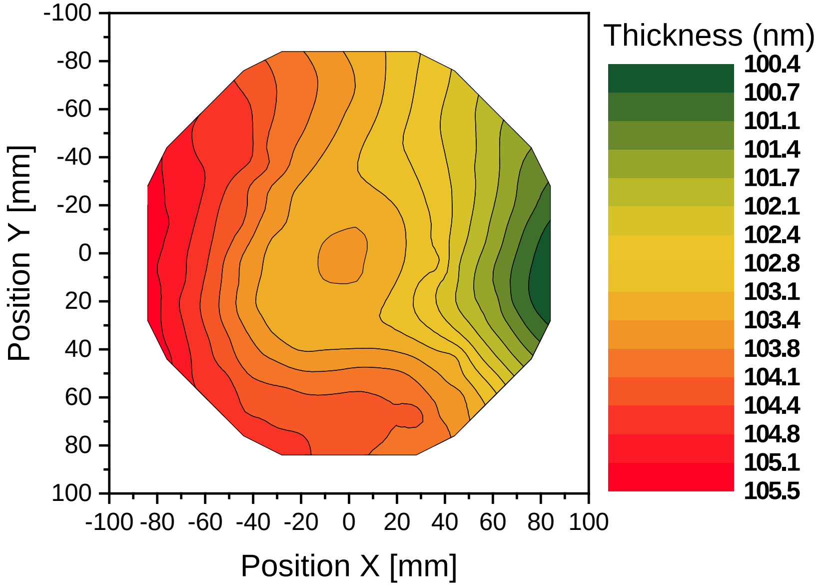
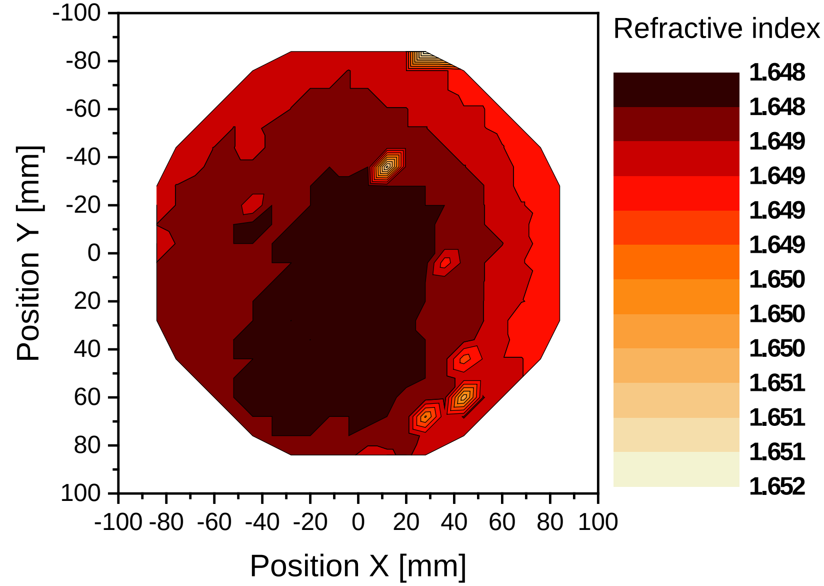
Thickness Uniformity

- Al_2O_3 thermal 300°C
- Ø 200 mm Si wafer: SD 1 σ = 1.1%
- Homogeneity (Max-Min/2$d_{average}$) = 2.44%

n = 1.6484 ± 0.0004





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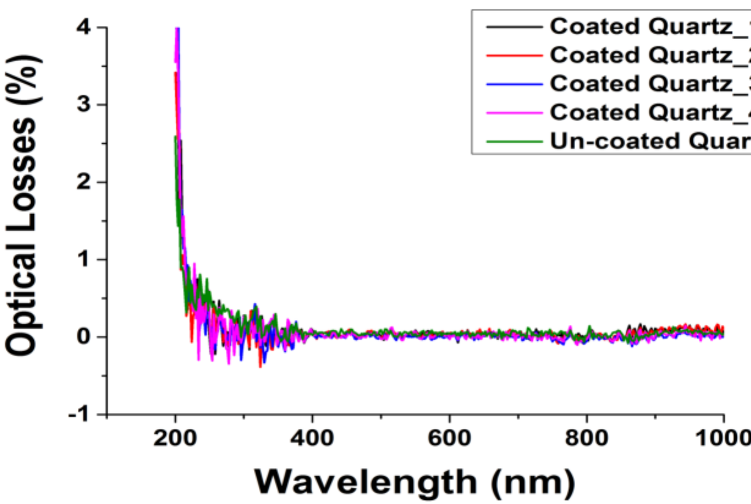
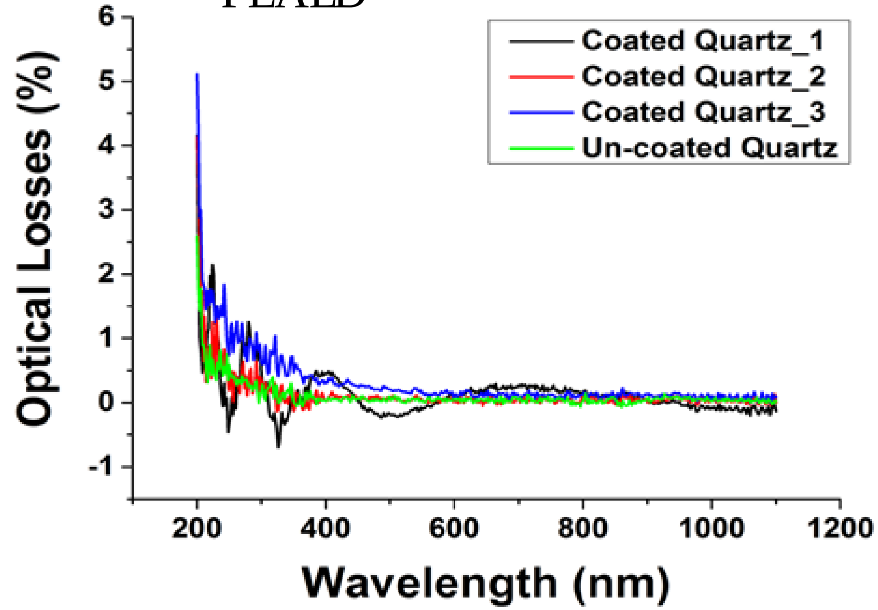
Optical Losses

- Al_2O_3 with low optical losses have been achieved
- confirmed by laser calorimetry measurements

Thermal



PEALD



@1064 nm on 300 nm thick Al_2O_3, ca. 2,5 ppm

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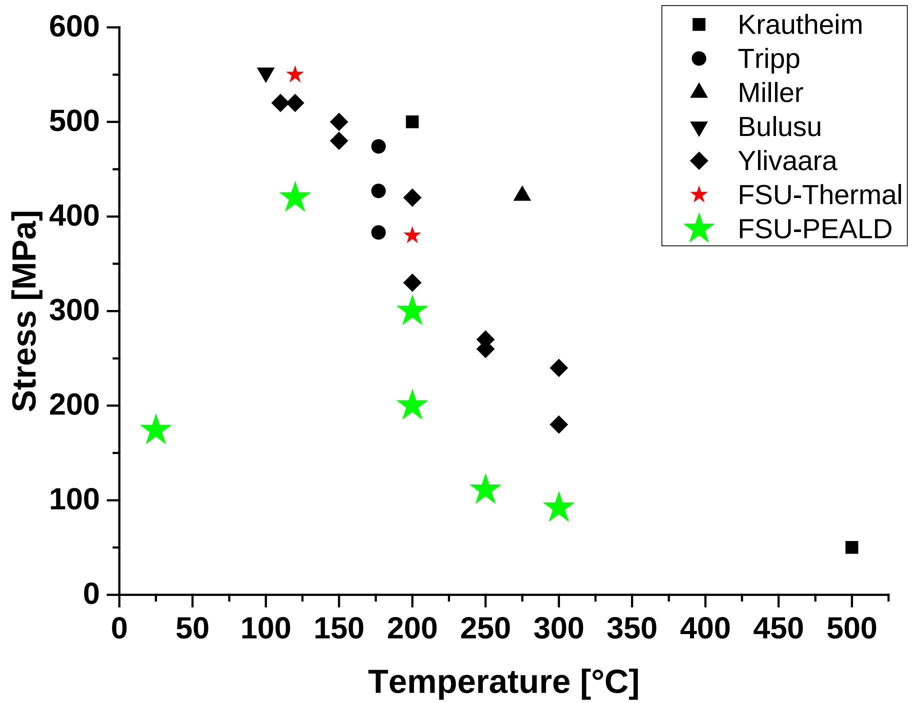
Al_2O_3 Tensile Mechanical Stress

- σ decreases with increasing deposition temperature
- PEALD coatings show lower mechanical stress than thermal ALD alumina
- residual stress = ∑(thermal stress + intrinsic stress)



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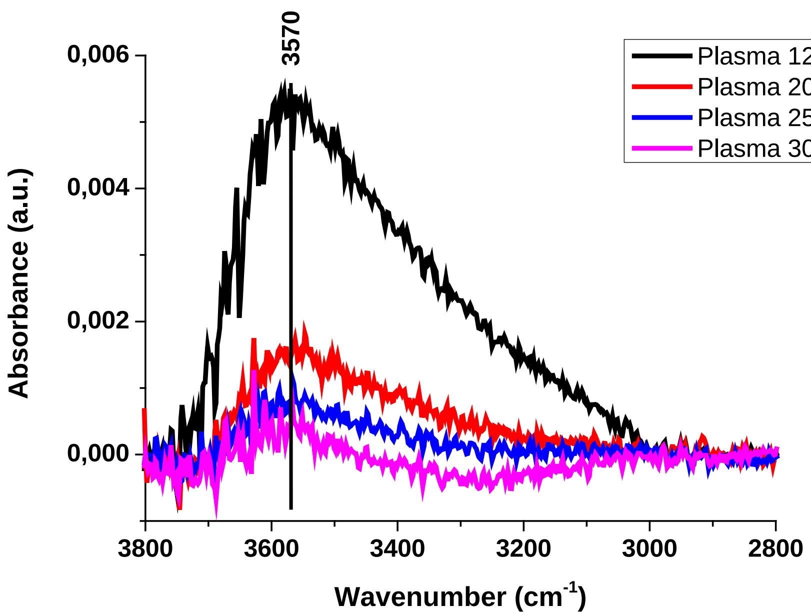
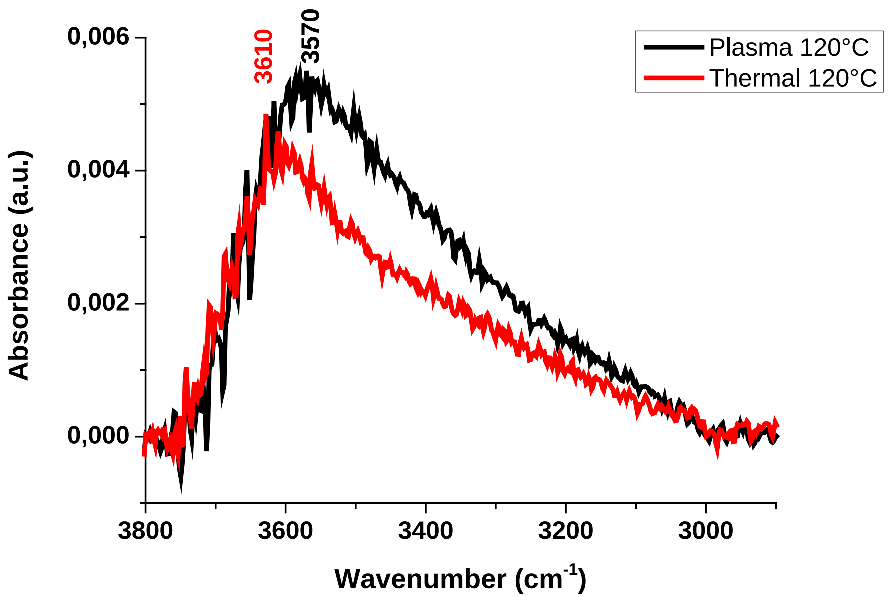
Al_2O_3 FTIR Spectra

- -OH content decreases with increasing temperature
- different hydrogen-bonding in thermal vs. PEALD processes





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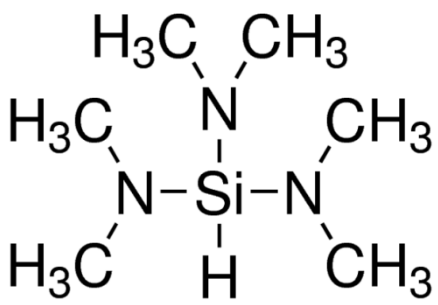
SiO_2

Tris(dimethylamino)silane + O_2 Plasma

$[(CH_3)_2N]_3SiH$

own vapour pressure



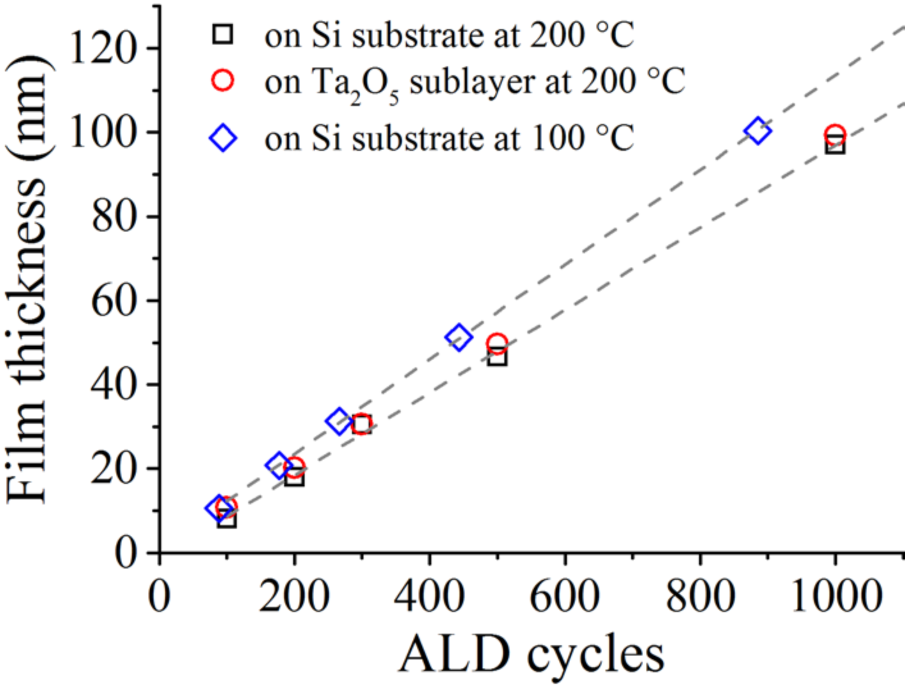
Bis(diethylamino)silane

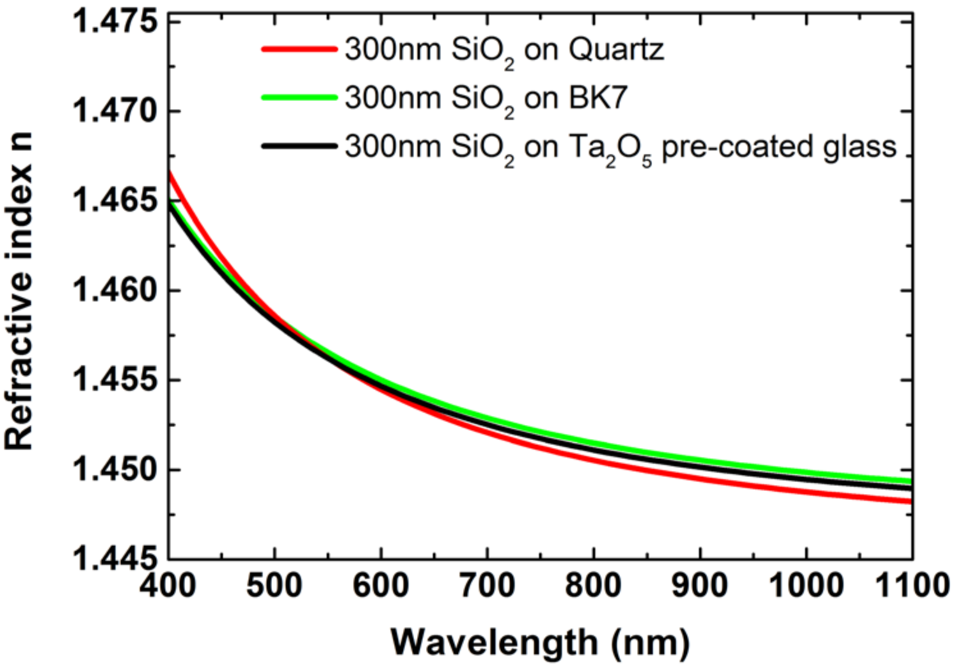
$[(C_2H_5)_2N]_2SiH_2$

heated and bubbled at 70°C

Influence of Substrate Material

- calibration curves are required





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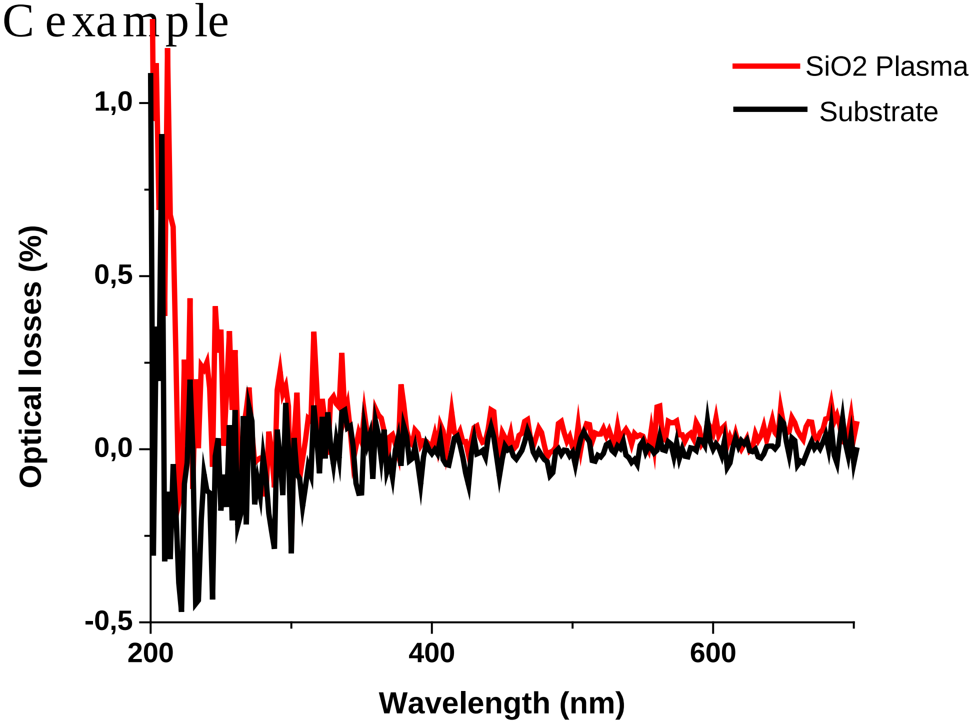
Optical Losses

- materials with low optical losses have been achieved
- confirmed by laser calorimetry measurements
- SiO_2 PEALD 200°C example



@1064 nm on 300 nm thick SiO_2, ca. 1,5 ppm (Laser Zentrum Hannover)

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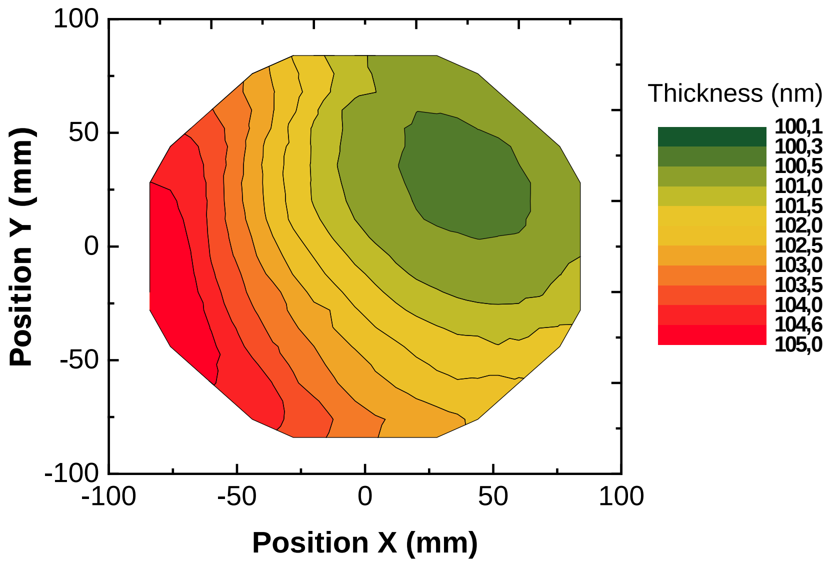
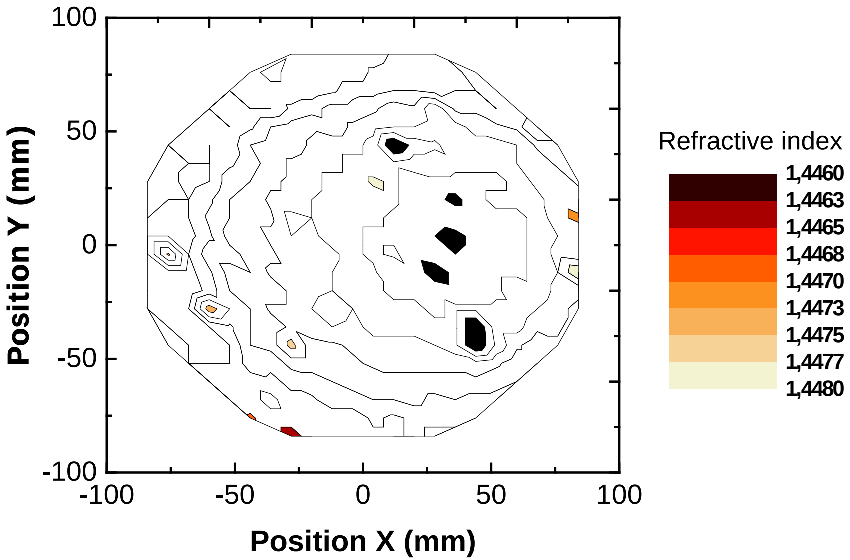
Thickness Uniformity

- SiO_2 PEALD 100°C
- Ø 200 mm Si wafer: SD 1 σ = 1.36%
- Homogeneity (Max-Min/2$d_{average}$) = 2.31%



n = 1.4472 ± 0.0005



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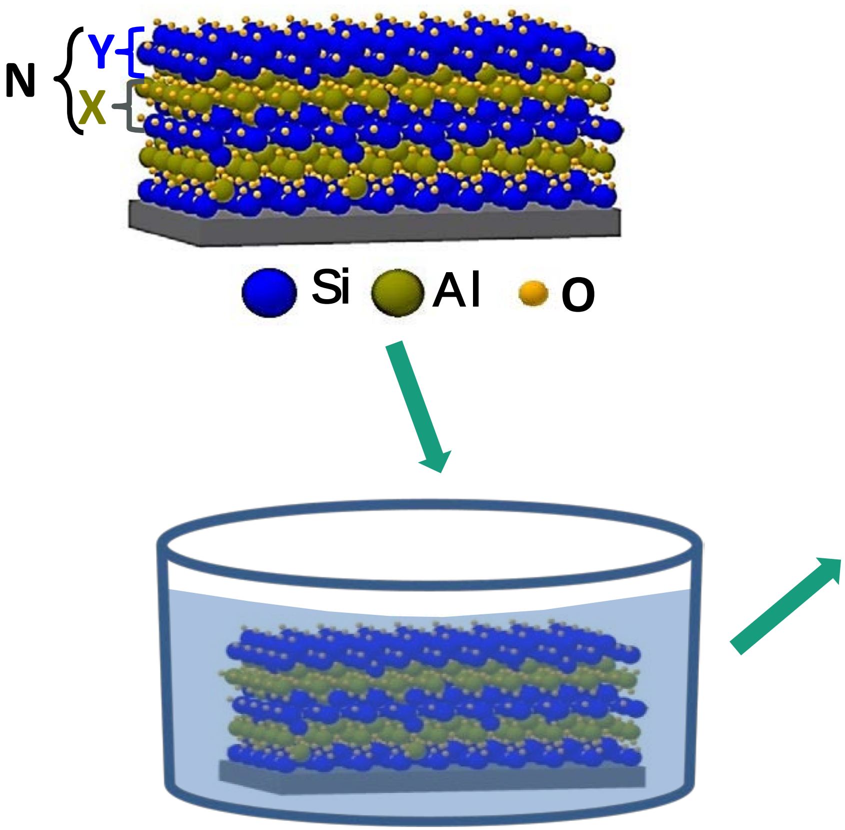
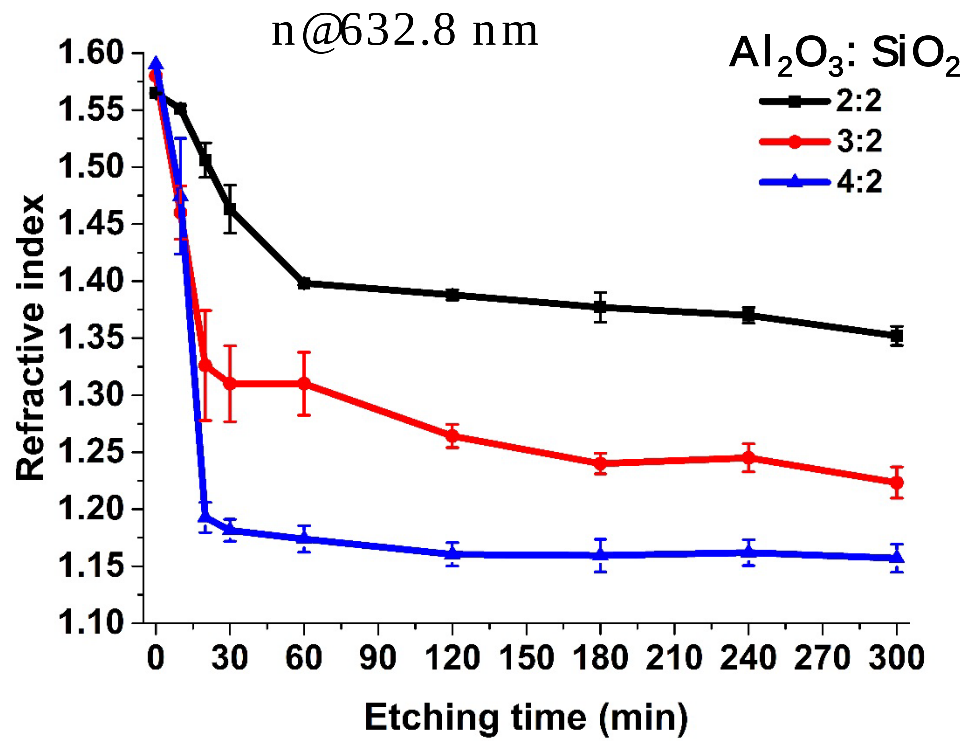
Composites and Nanoporous SiO_2

- atomically mixed Al_2O_3/SiO_2 composites and selective removal of Al_2O_3

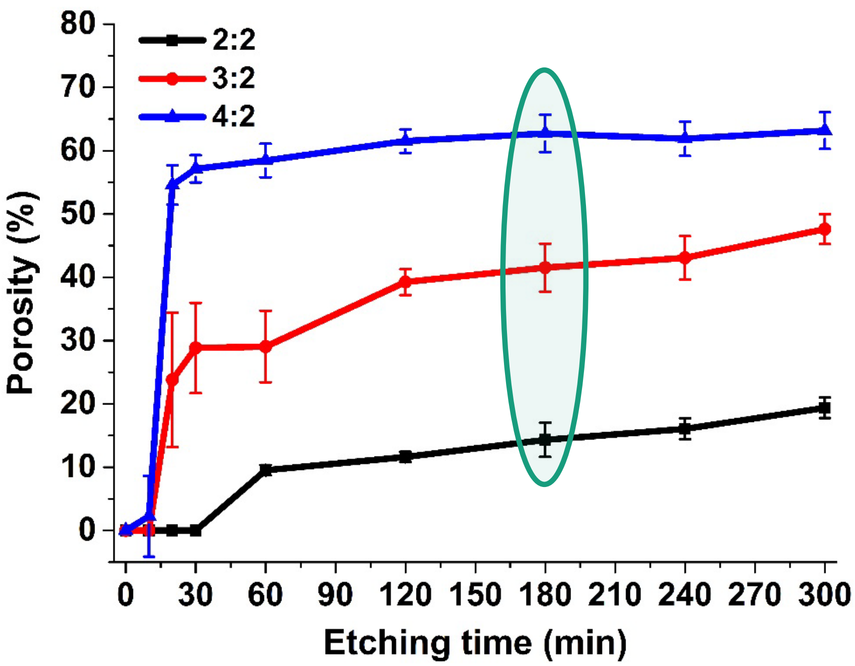


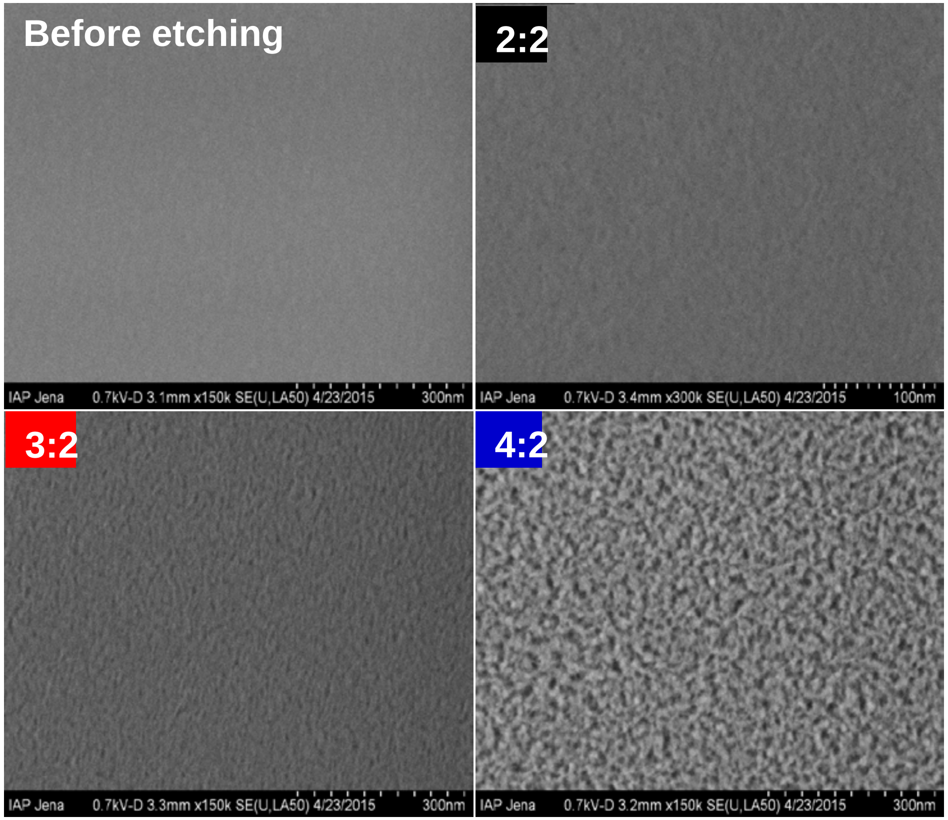


L. Ghazaryan, A. Szeghalmi, E. B. Kley, U. Schulz, DPMA Anmeldung. 2016
L. Ghazaryan, A. Szeghalmi, E. B. Kley, DPMA Anmeldung. Anmeldetag 24. Februar 2015. 10 2015 203 307.4

Nanoporous SiO_2

- atomically mixed Al_2O_3/SiO_2 composites and selective removal of Al_2O_3
- precisely control porosity and refractive index through atomic composition





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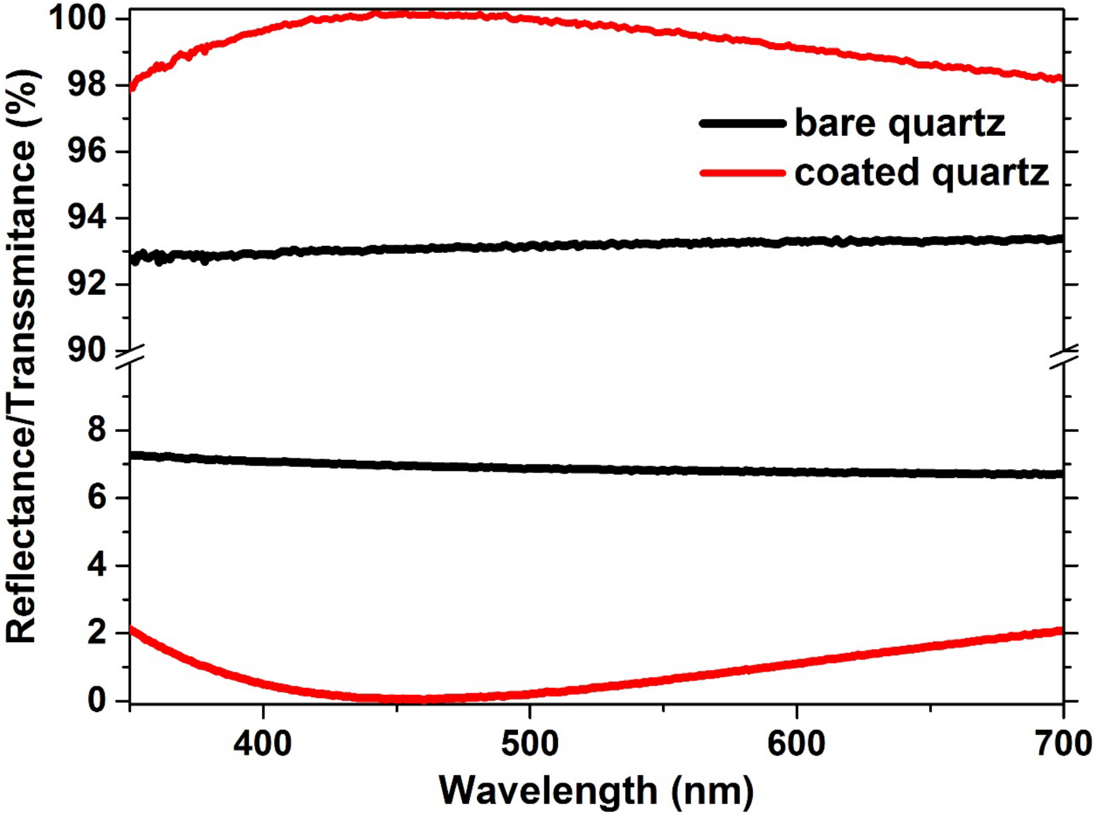
Applications of Nanoporous SiO_2

- antireflection coatings



- ✓ one layer
- ✓ double side coating
- ✓ 92 nm each side
- ✓ n = 1.22
- ✓ Over 98 % Transmittance in VIS
- ✓ Low optical losses

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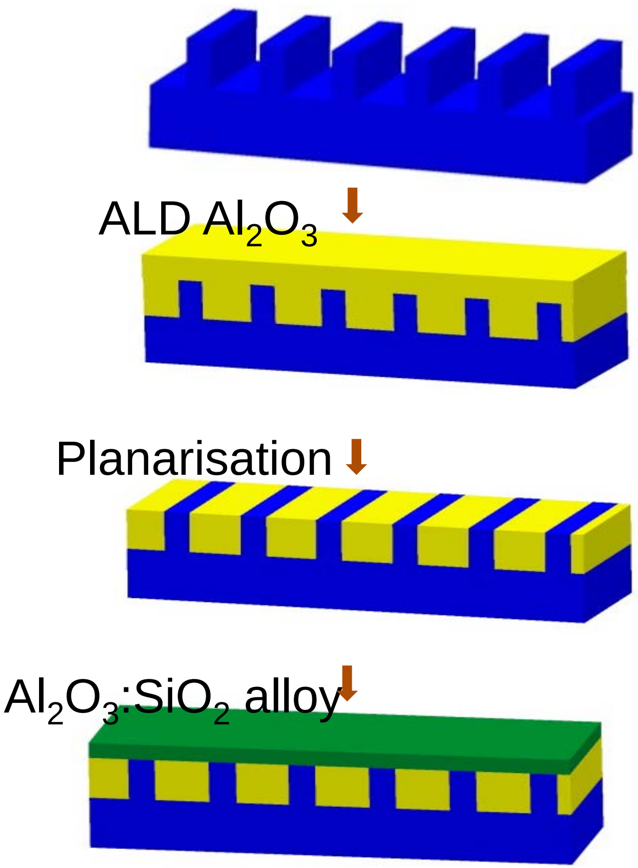
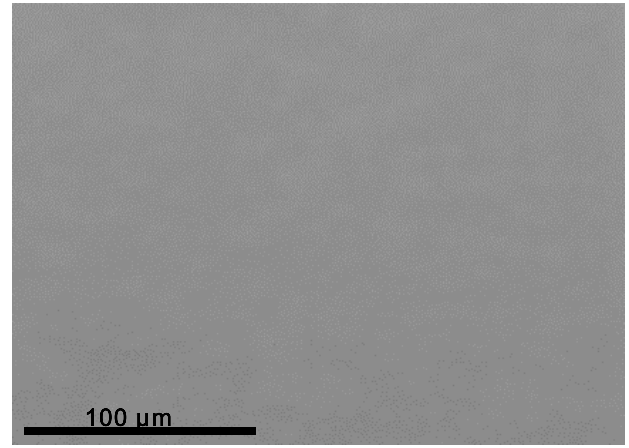
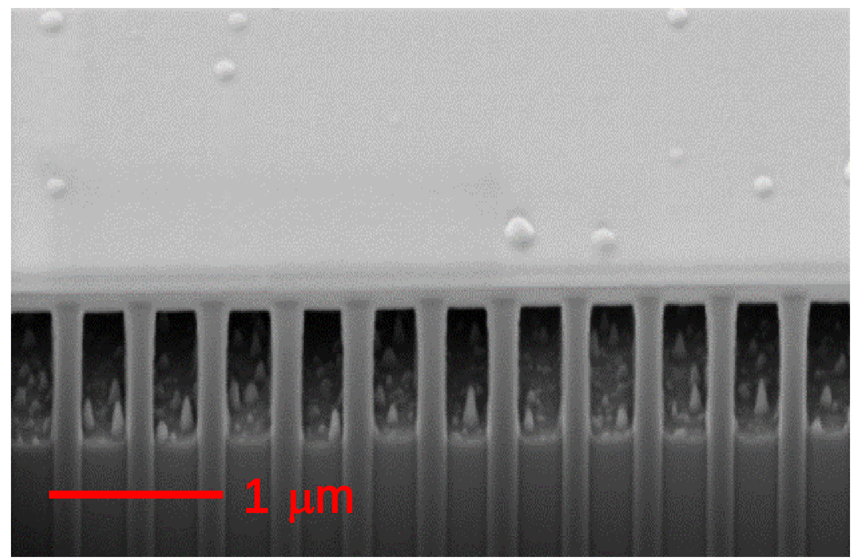
Applications of Nanoporous SiO_2

- diffusion membrane
- High efficiency transmission gratings







Outline

- Introduction
- Coating equipment
- Results
  - Materials
  - Interference Coatings
  - Diffractive Optics
- Outlook

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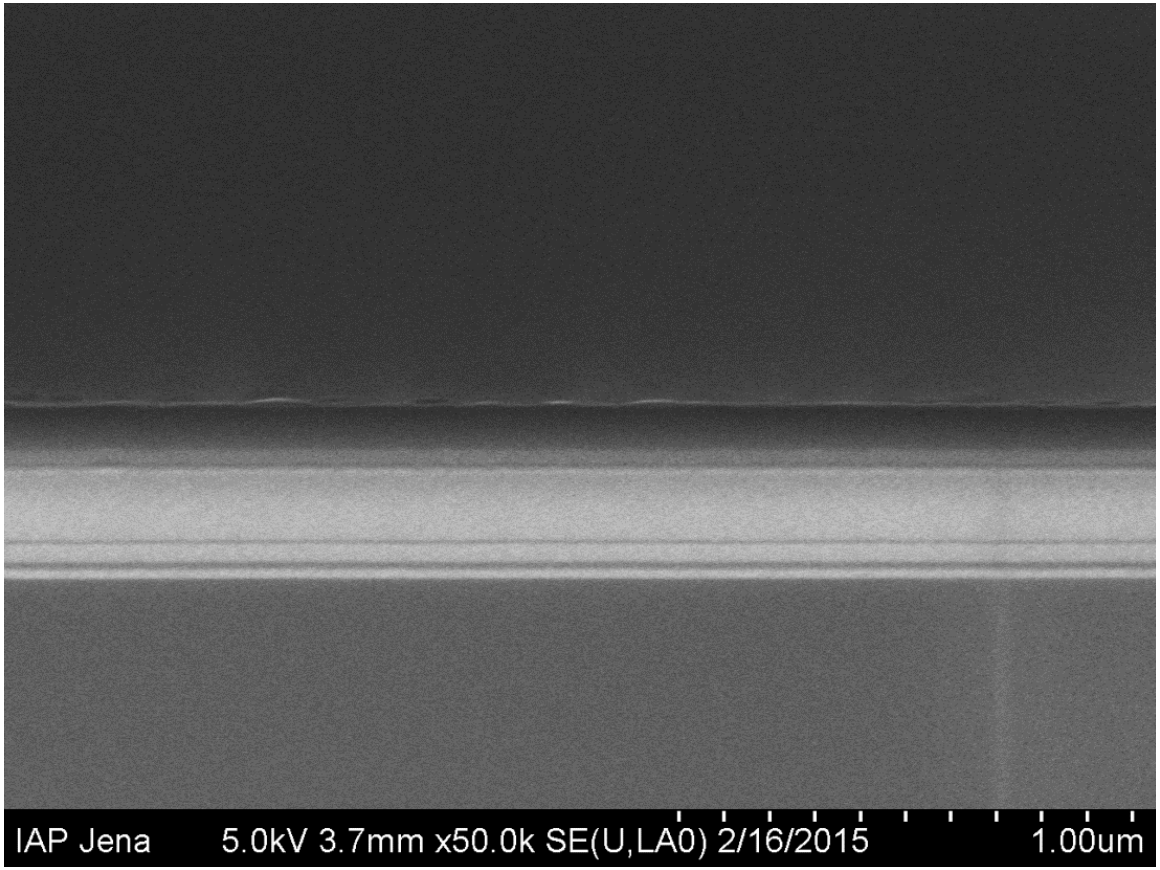
Antireflection Coating SEM Image

- **Total 391 nm**
- SiO_2 97.3
- HfO_2 29.8
- SiO_2 19.5
- HfO_2 153.5
- SiO_2 10.4
- HfO_2 40.5
- SiO_2 19.1
- HfO_2 18.6
- N-SF8 substrate



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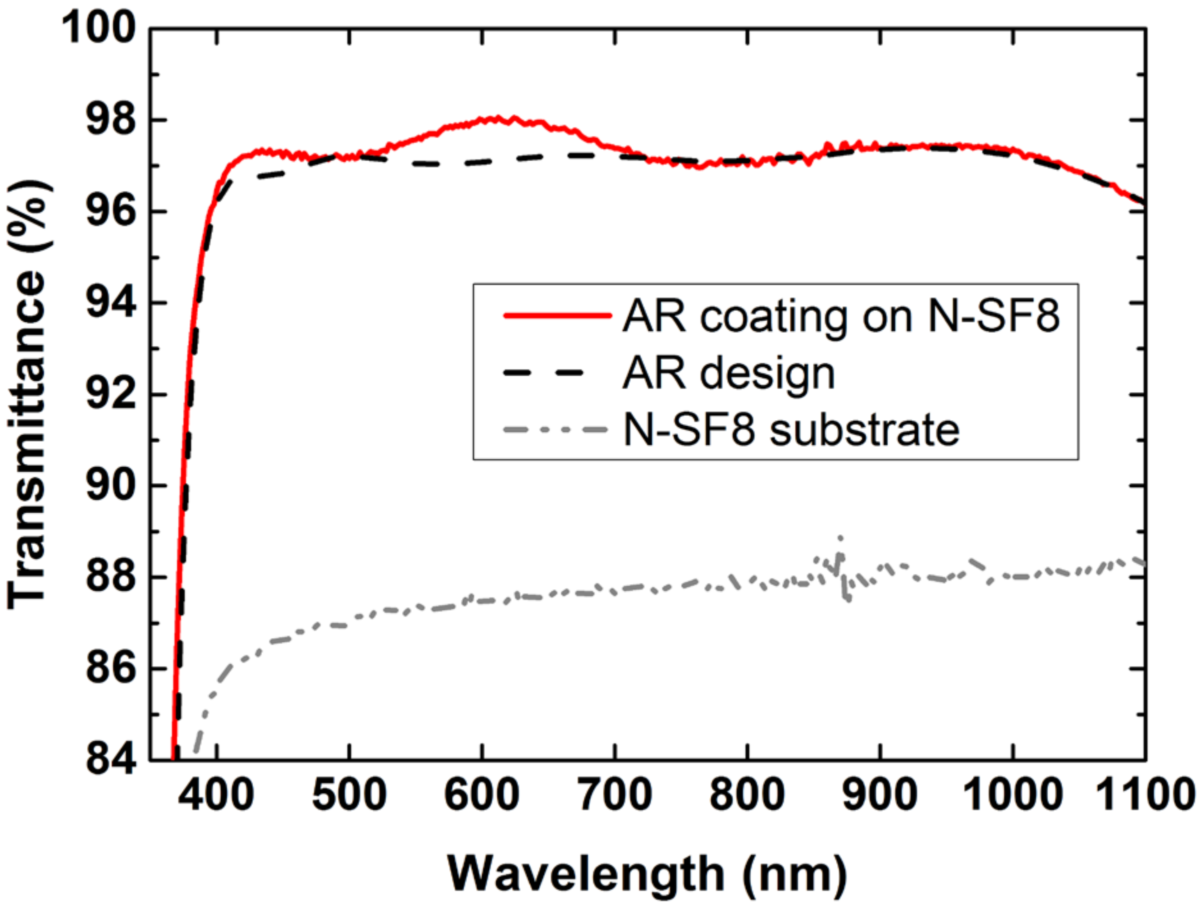
Antireflection Coatings



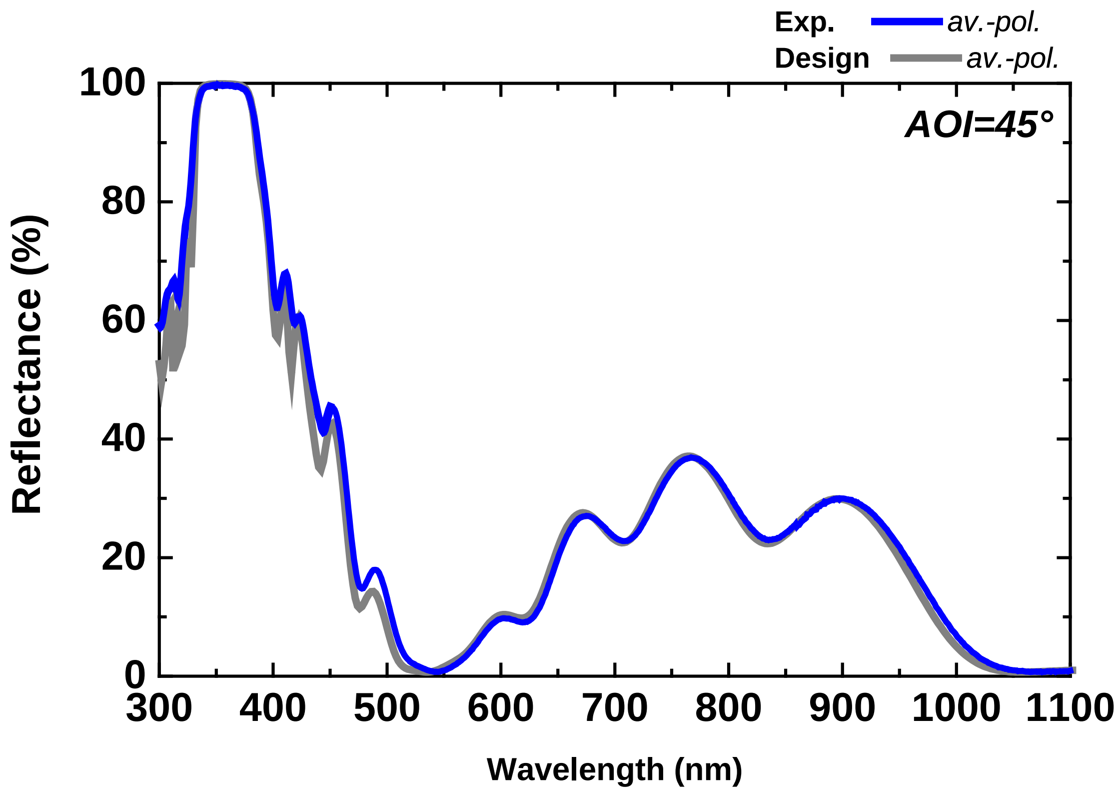
[3] K. Pfeiffer, S. Shestaeva, A. Bingel, P. Munzert, L. Ghazaryan, C. van Helvoirt, W. M. M. Kessels, U. Sanli, C. Grévent, G. Schütz, M. Putkonen, I. Buchanan, L. Jensen, D. Ristau, A. Tünnermann, A. Szeghalmi. Opt. Mater. Express 6 (2016) 660

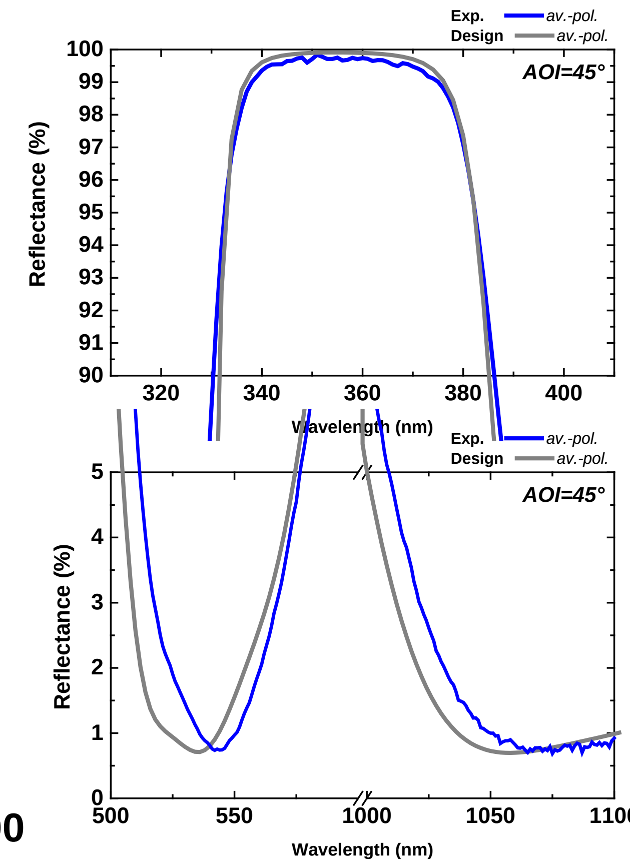
[4] A. Szeghalmi, M. Helgert, R. Brunner, F. Heyroth, U. Gösele, M. Knez. Appl. Opt. 48 (2009) 1727

Dichroic Mirror at 355 nm

- 30 layers [HfO_2(Al_2O_3) and SiO_2]
- ca. 1.9 µm total thickness
- high adhesion coating to substrate
- >99.5% reflectance at 45° AOI @355

Total deposition time: ~4 days
NO optical monitoring





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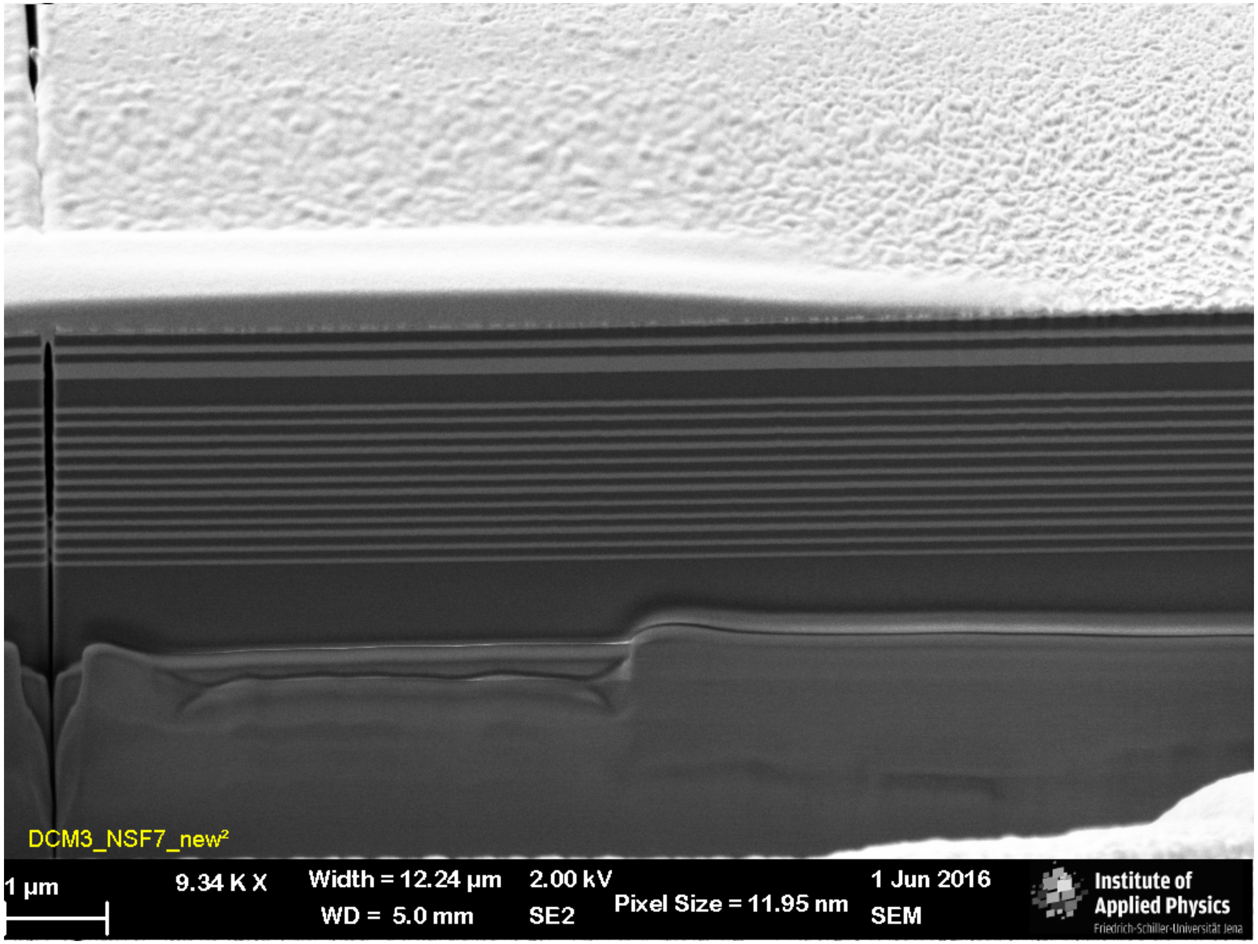
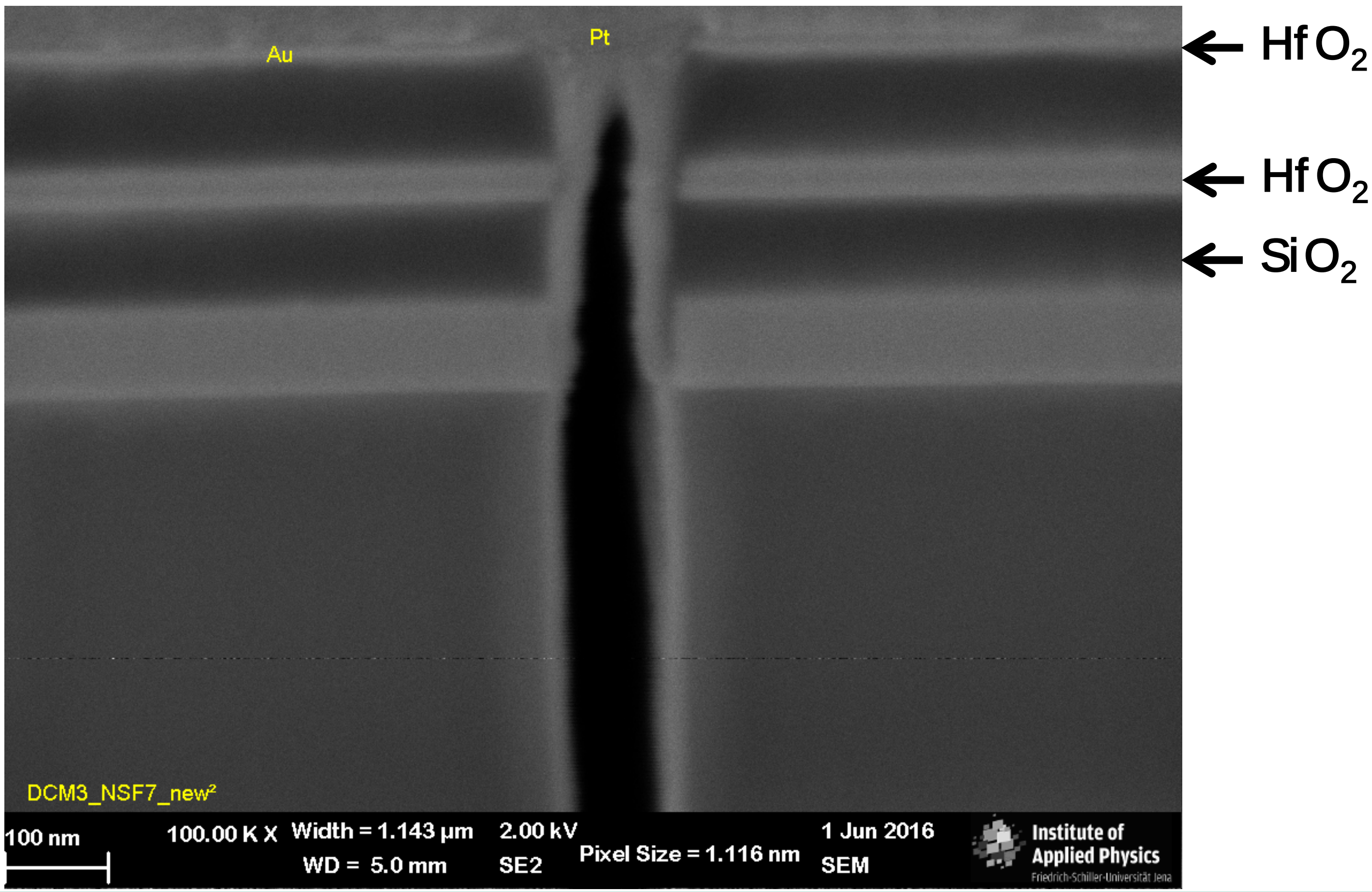
Highly Reflective Dichroic Mirror

- cracking of the coating and substrate



Highly Reflective Dichroic Mirror

- cracking of the coating and substrate
- crack occurs after deposition



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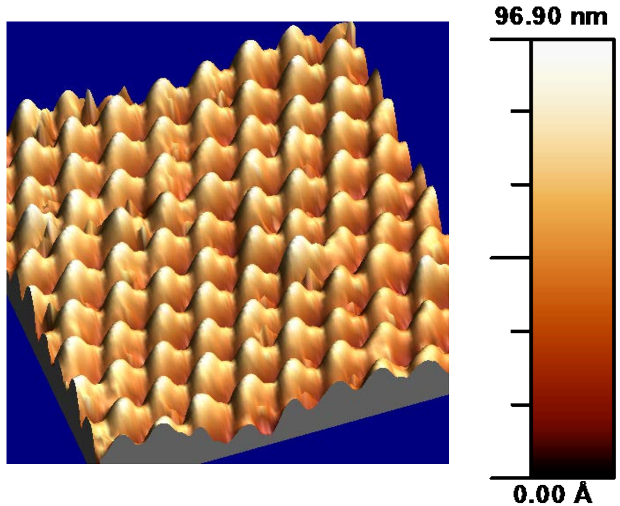
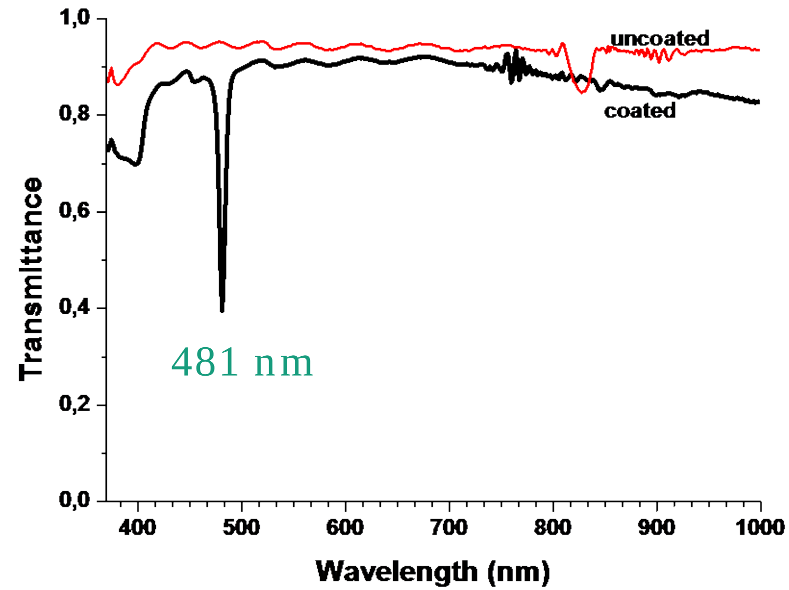
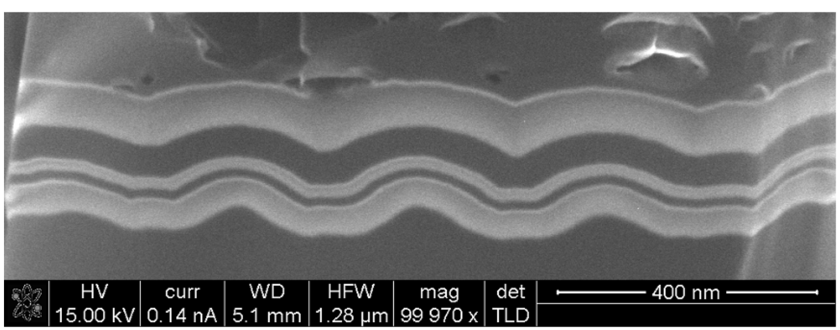
Resonant Waveguides

- nanostructure + ALD coating





C-Paste



PC

Film Thickness
~ 120 nm
(~2 hours deposition)

Narrow Band

A. Szeghalmi, E. B. Kley, M. Knez. J. Phys. Chem. C 114 (2010) 21150
A. Szeghalmi, M. Helgert, R. Brunner, F. Heyroth, U. Gösele, M. Knez. Adv. Funct. Mater. 20 (2010) 2053

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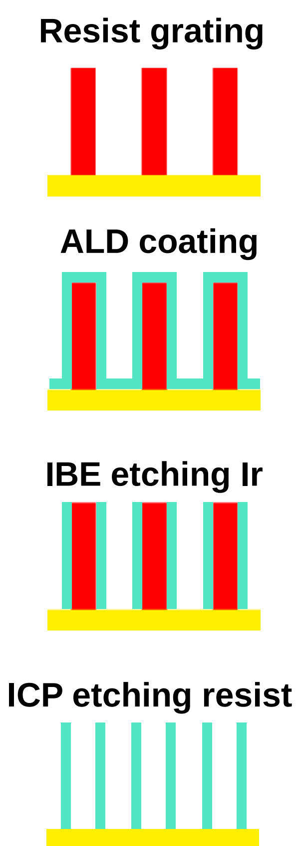
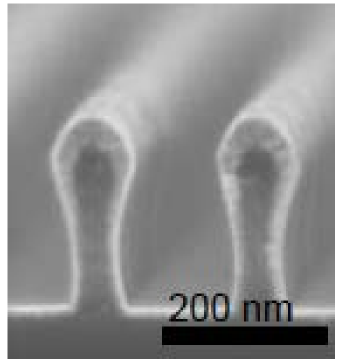
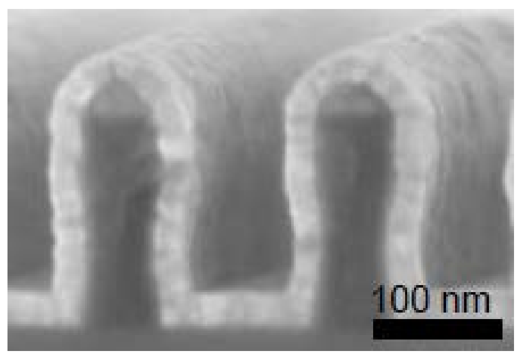
Polarizers



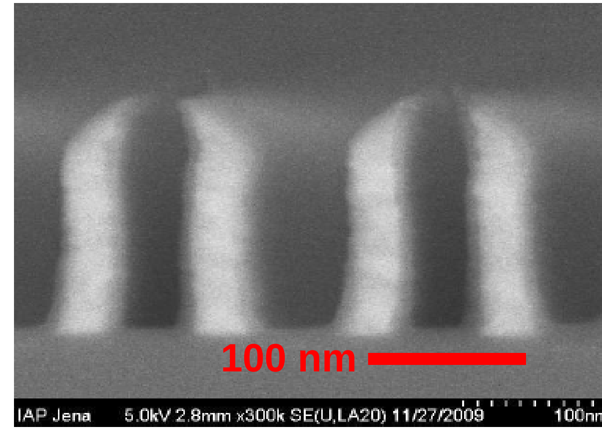
sputtering

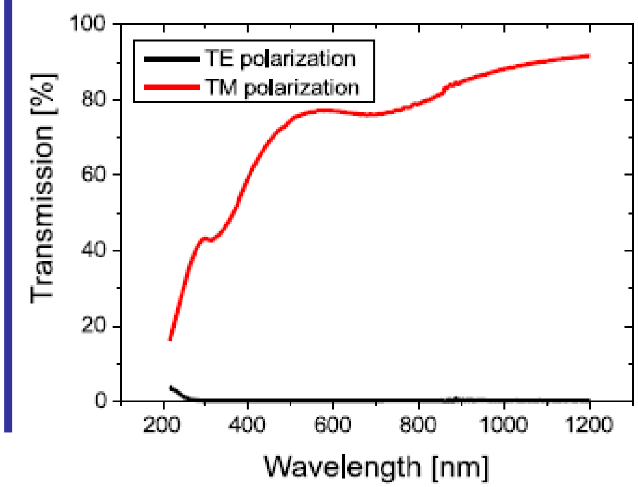


ALD coating



Ir wire grid polarizer

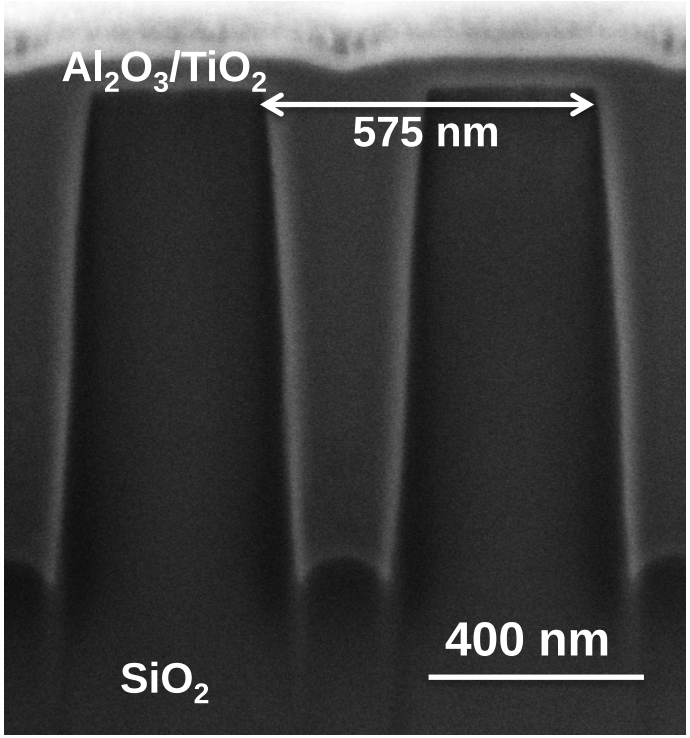


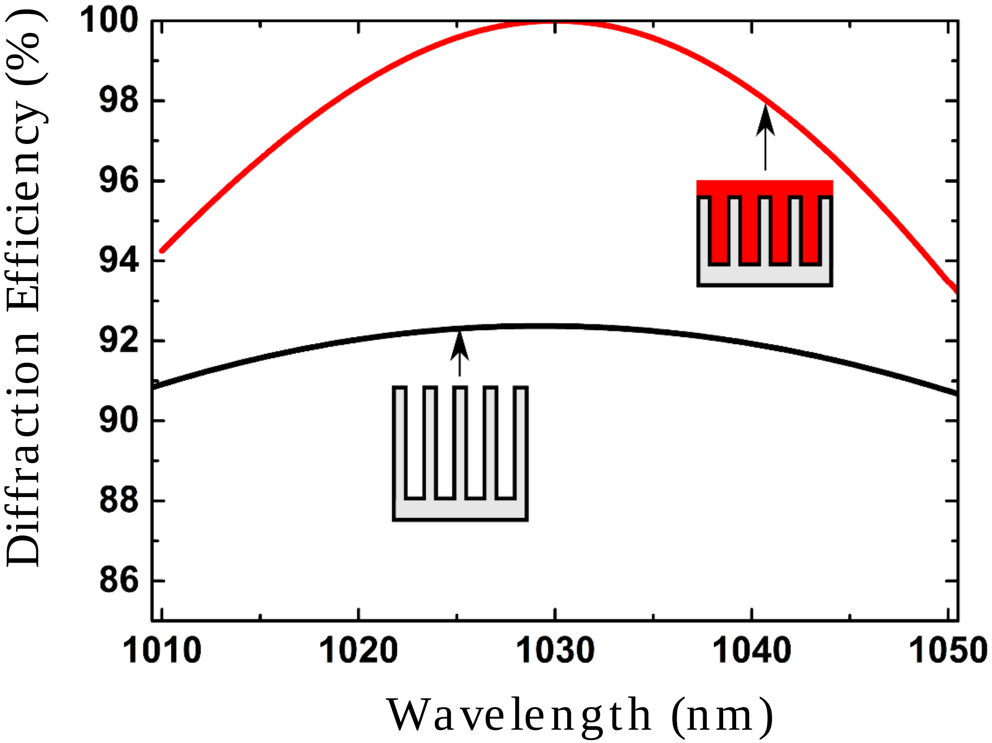


[1] T. Weber, T. Käsebier, A. Szeghalmi, M. Knez, E. B. Kley, A. Tünnermann. Nanoscale Research Lett. 2011, 6, 558
[2] Y. Bourgin, T. Siefke, T. Käsebier, P. Genevée, A. Szeghalmi, E. B. Kley, U. D. Zeitner. Optics Express, 2015, 23, 16628

High Efficiency Transmission Gratings

- measured diffraction efficiency (-1 order) grating for TE-Polarisation 97.5%
- measured diffraction efficiency (-1 order) grating for TM-Polarisation 95%





S. Ratzsch, E. B. Kley, A. Tünnermann, A. Szeghalmi. Materials, 2015, 8, 7805-7812.
S. Ratzsch, E. B. Kley, A. Tünnermann, A. Szeghalmi. Optics Express, 2015, 23, 17955-17965.
S. Ratzsch, E. B. Kley, A. Tünnermann, A. Szeghalmi. Nanotechnology, 2015, 26, 024003 (1-11).

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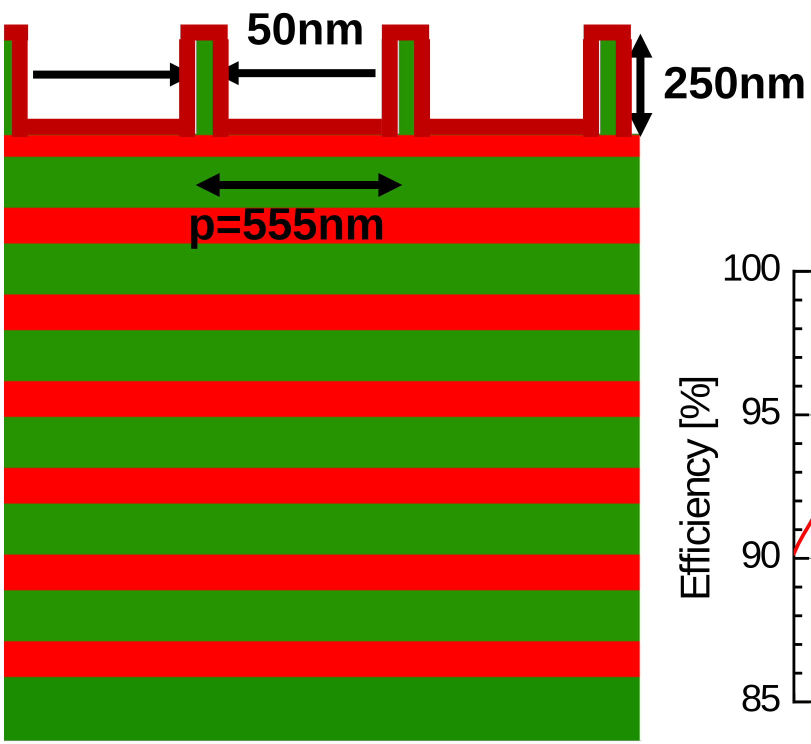
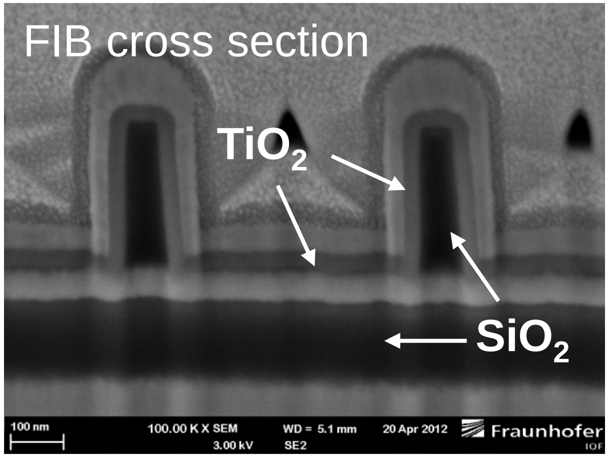
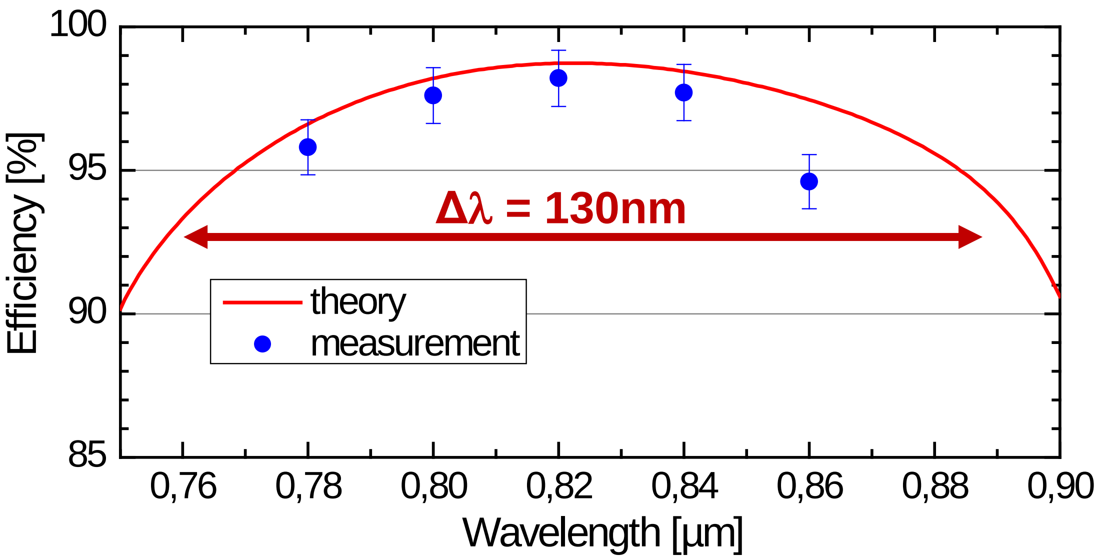
High Index Contrast Grating

- fabricate a low fill-factor grating in SiO_2
- conformal overcoating with ALD
- TiO_2 thickness ca. 44 nm







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ALD Portfolio @Jena (Fraunhofer IOF & University)

ALD CENTRE for OPTICS

- ALD Material Development
- Interference Coating Systems
- Nano and Microstructured Optics

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