Linear and nonlinear absorption in optical coatings

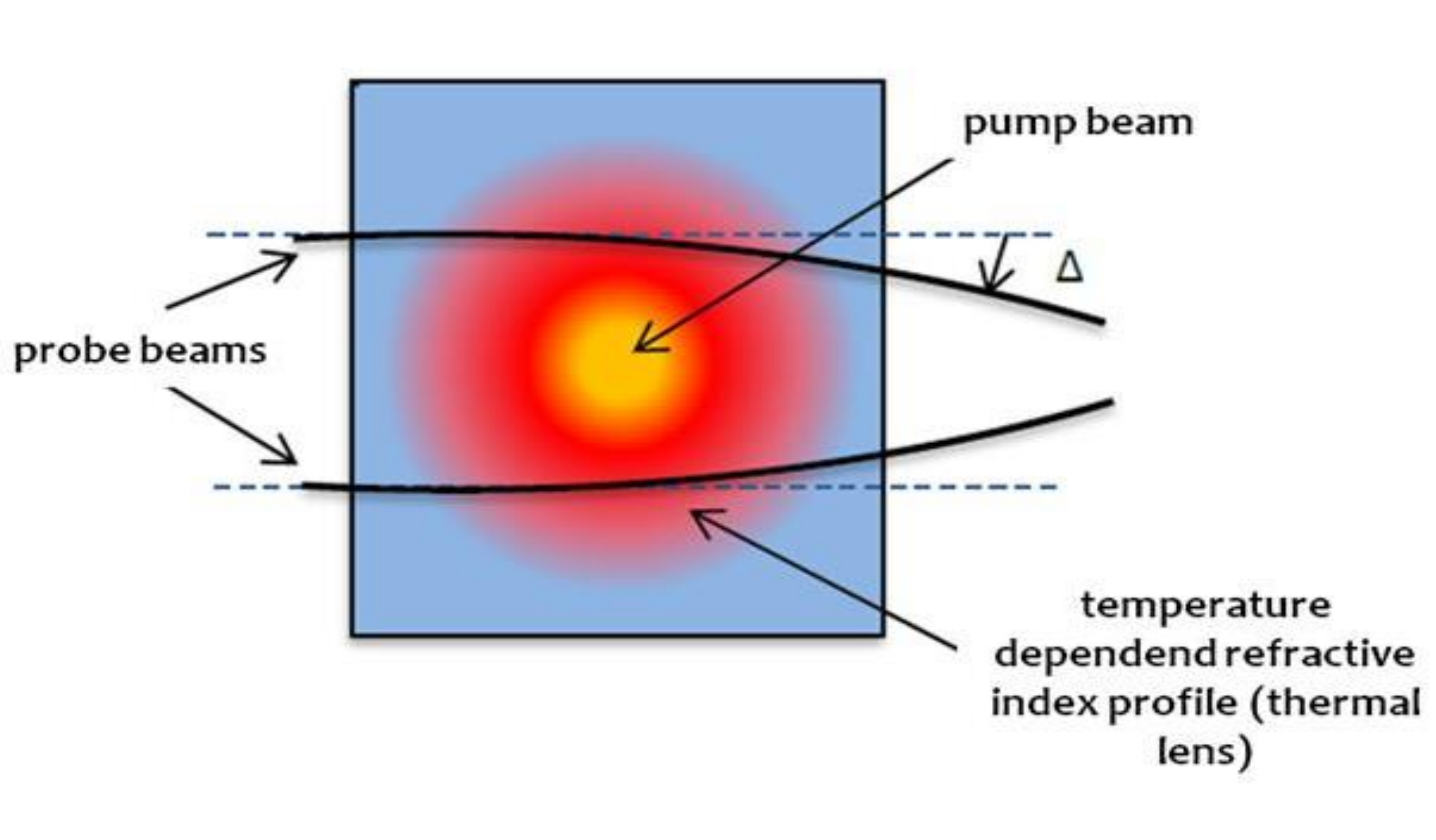
Christian Mühlig

Outline

- ***L**aser **I**nduced **D**eflection (LID) absorption measurement technique*
- Calibration / setup
- Measurement concepts (round substrates)
- Experimental results
- Summary

Laser induced deflection (LID)

Photo-thermal technique with ***transversal*** probe beam guiding
(no crossing between pump and probe beam)



Absorption

↓

Temperature profile

↓

Refractive index profile
(dn/dT; thermal expansion → stress)

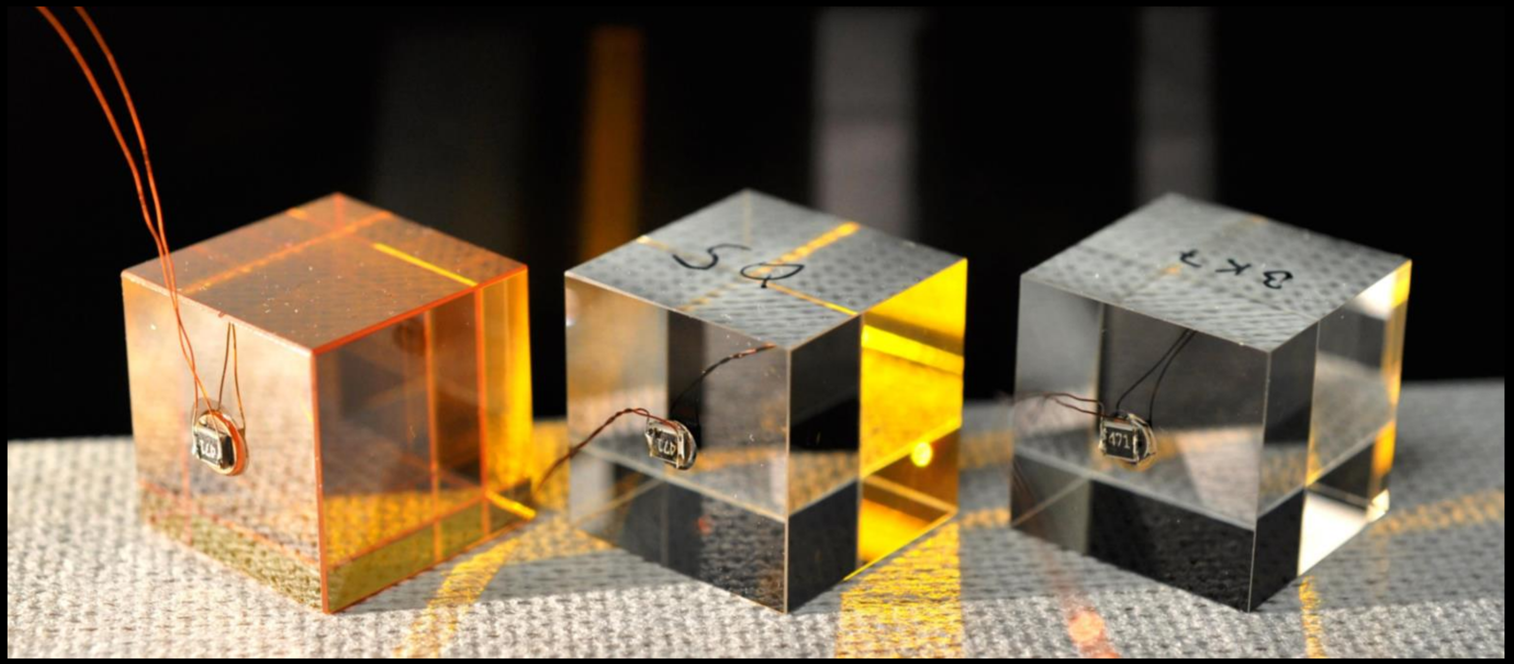
↓

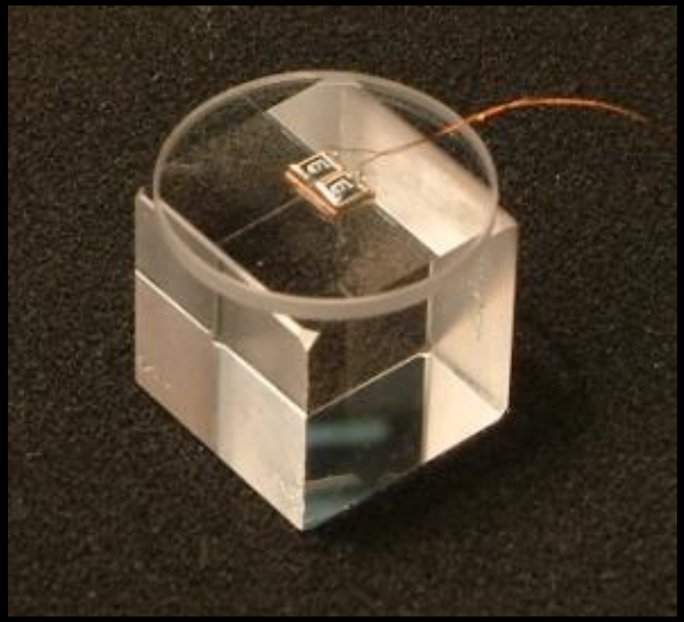
Probe beam deflection
(steady-state)

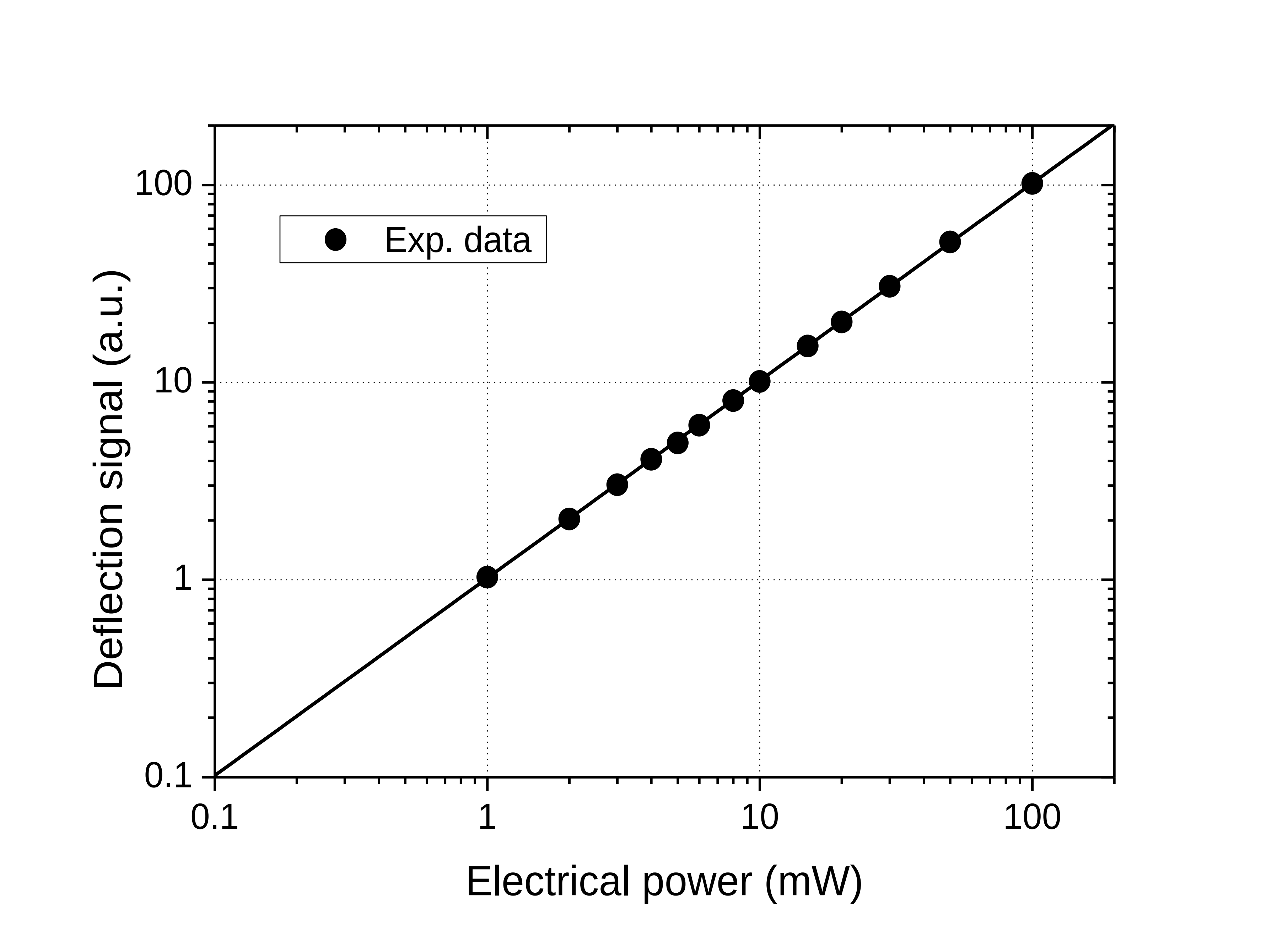
- Probe beam deflection is a ***direct measure*** for the absorbed mean laser power
- Main challenge: How to calibrate for ***reliable*** absolute absorption data?

Calibration for absolute absorption data

- Absorption induced heat is simulated by electrical heating (adaption of calorimetry's calibration)
- ***Absolute data without simulations and knowledge of material's photo-thermal parameters***
- Unique feature for photo-thermal techniques

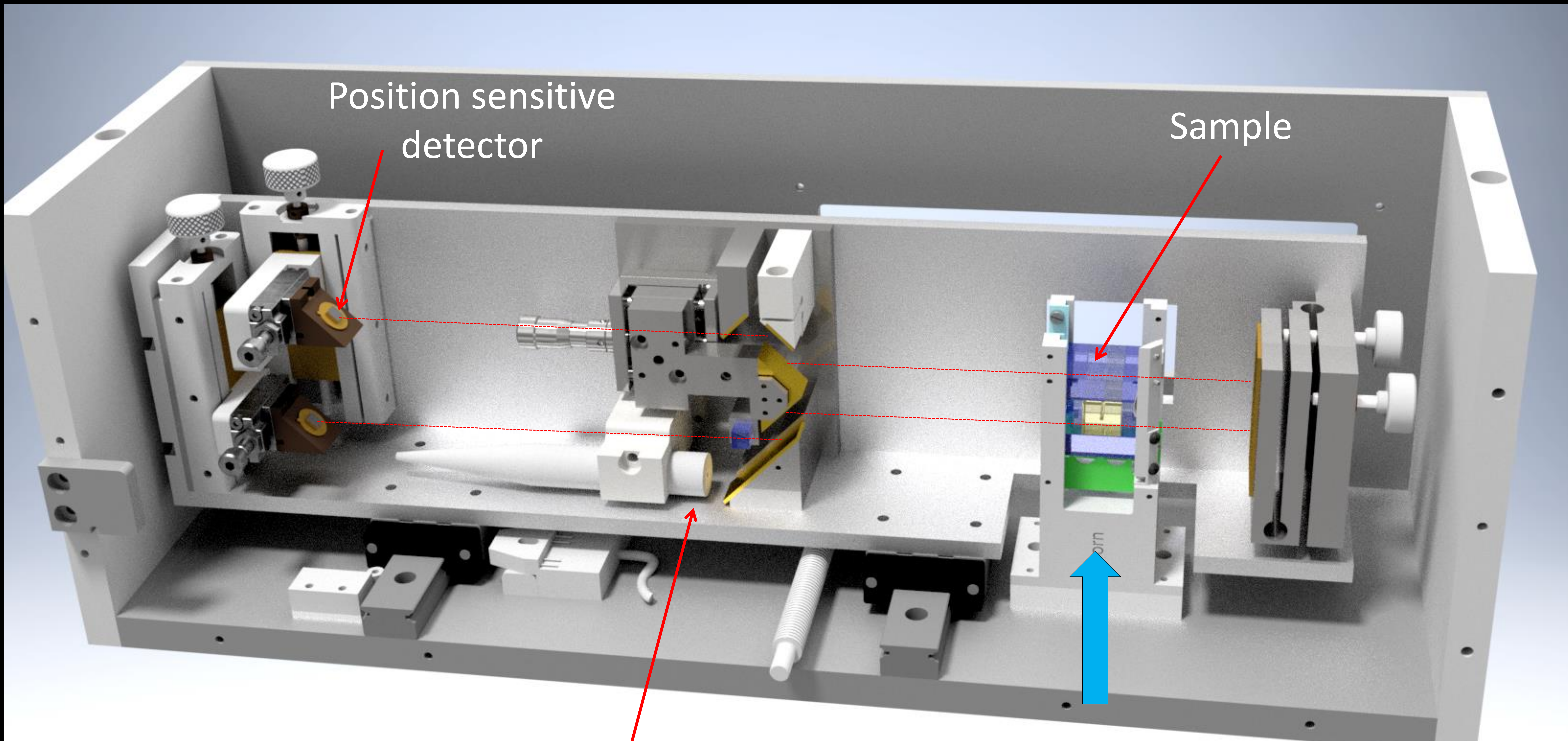






- Linear behavior over several orders of magnitude of absorbed power
- Energy balance measurements (T+R+A+S=1) approve that no systematic error exists

Setup



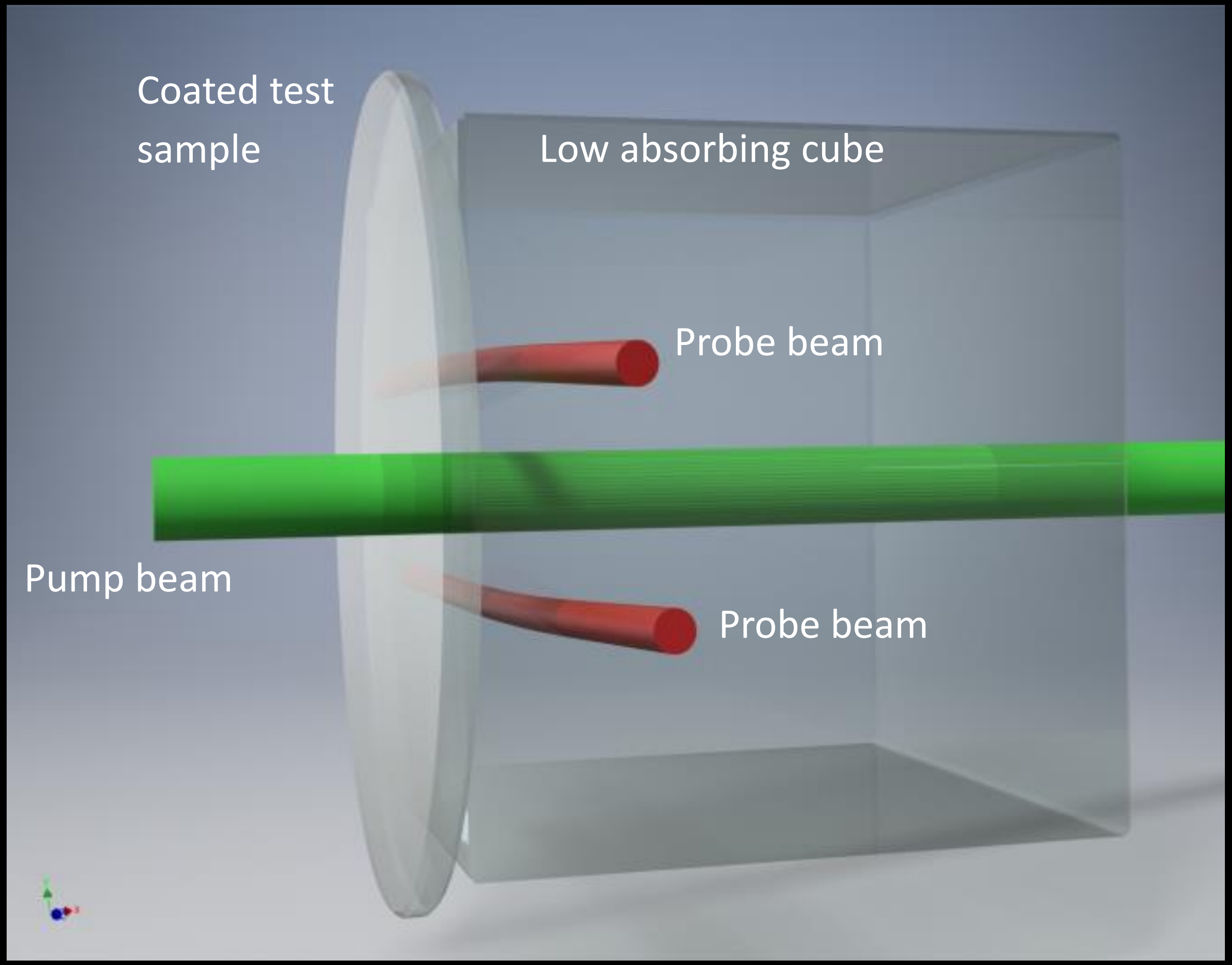
Probe beam unit
(movable to scan over sample length)

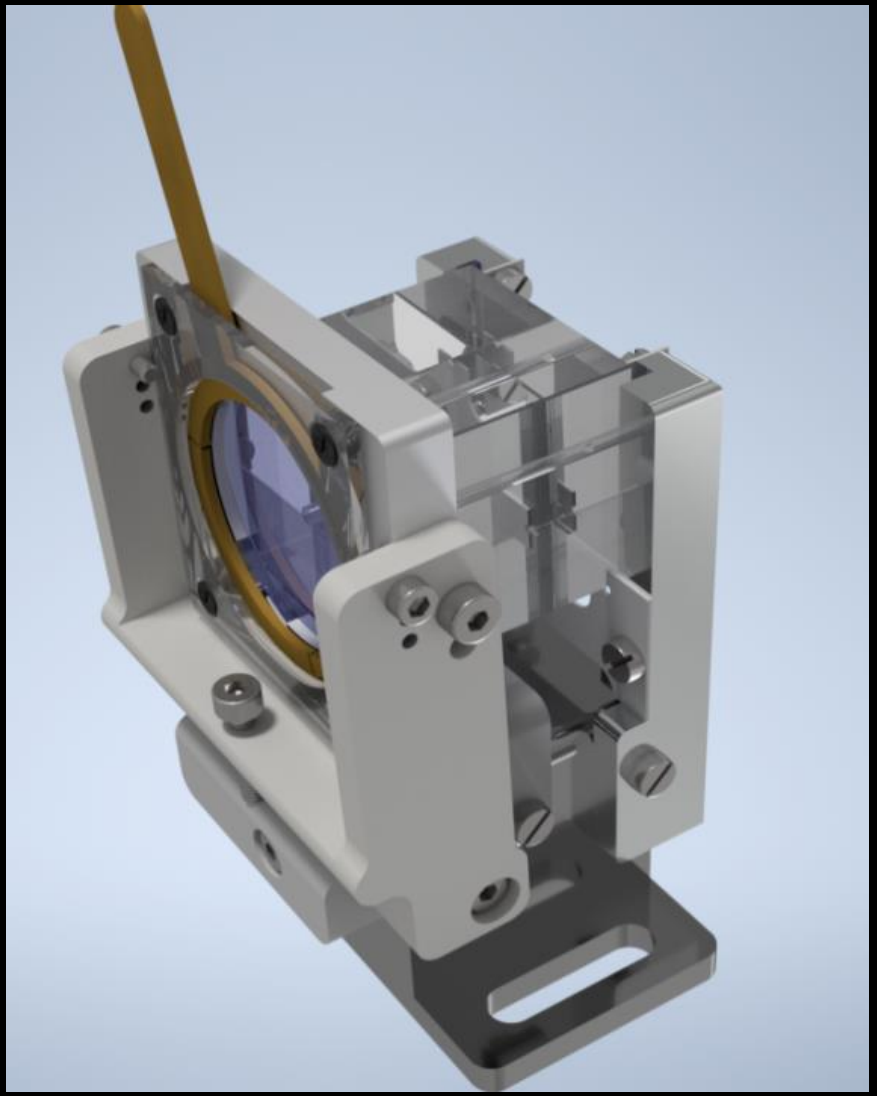
Pump laser

Example for sensitivity (fused silica): 10µW absorbed power

Measurement concepts

Concept for “*thin* disc” samples

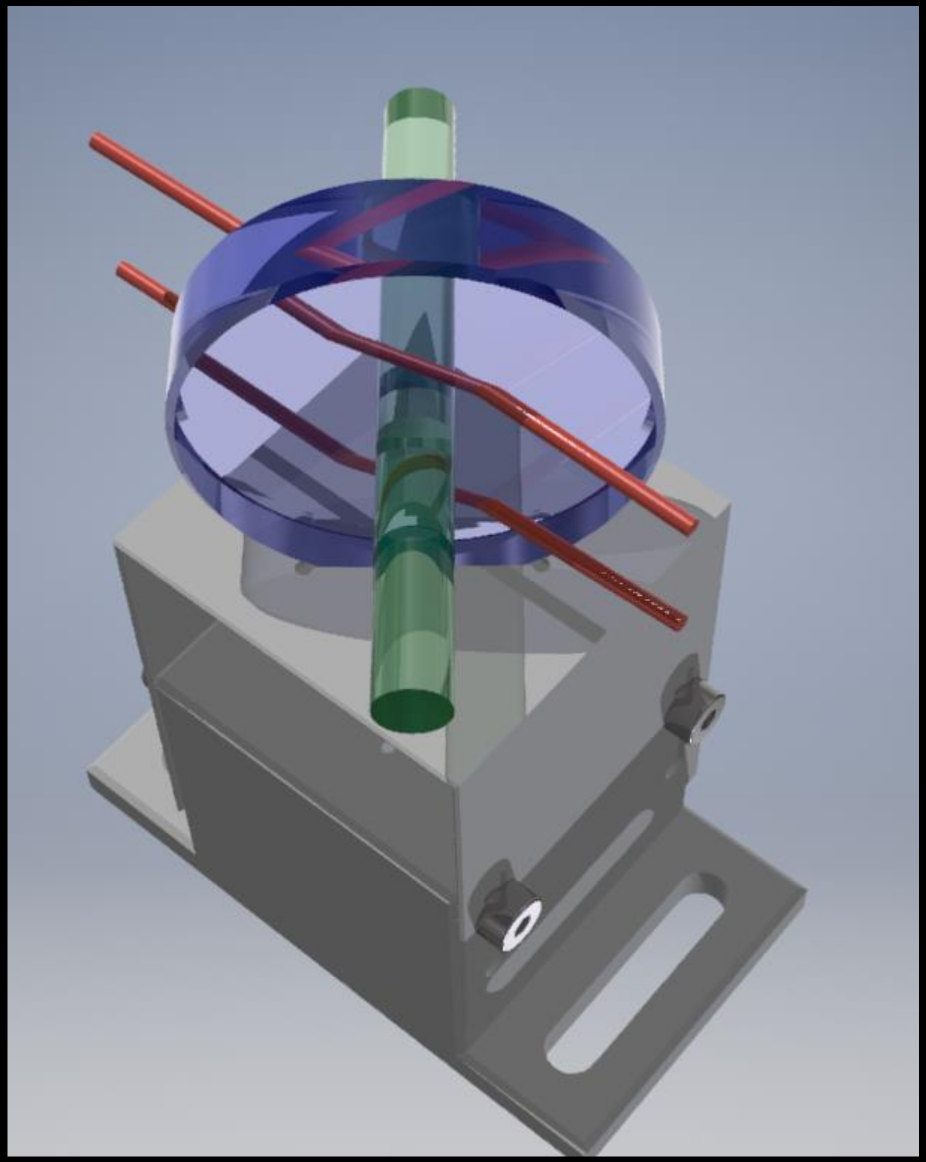


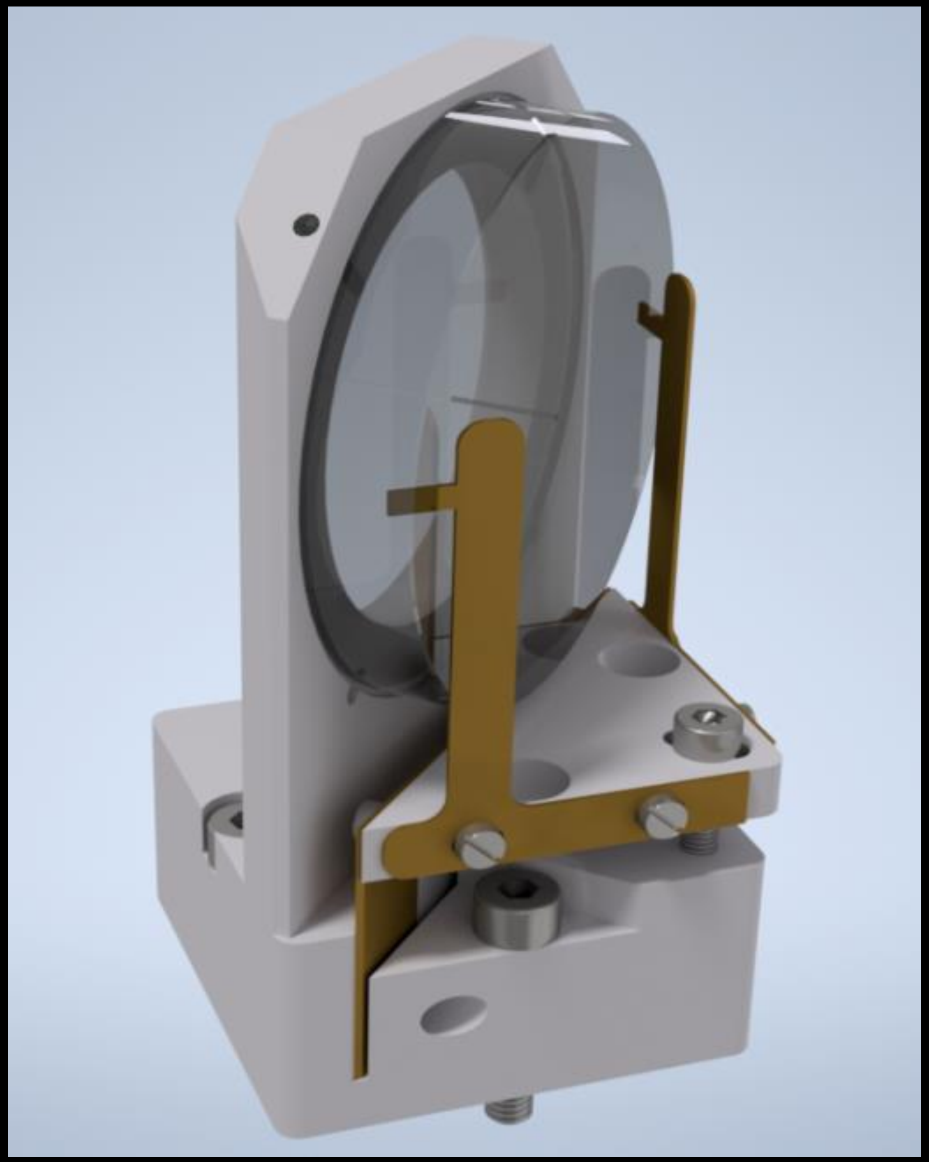


- Coated disc is attached to a low absorbing (cubic) substrate
- Heat transfer from disc to substrate → probe beam deflection *inside* substrate
- If disc material/substrate show measuring signal itself → Measuring coated vs. uncoated thin disc → discrimination of coating absorption
- Double-side coated samples give better accuracy (though no access to individual coating)
- Typical disc size: ∅=1/2”, 1”…2”; thickness ≤2mm (sensitivity decreases with thickness; measuring signal from disc material increases)
- Standard electrical calibration

Measurement concepts

Concept for “*thick* disc” samples

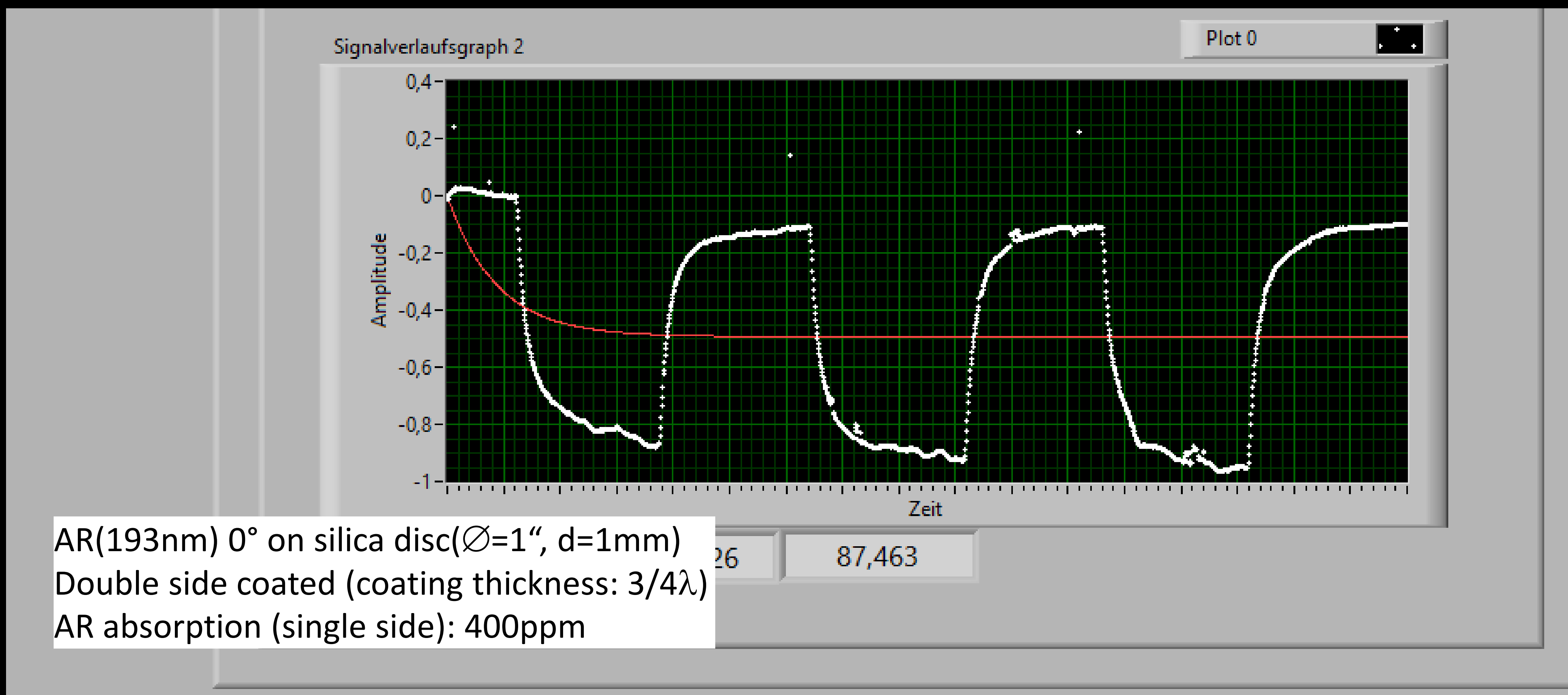


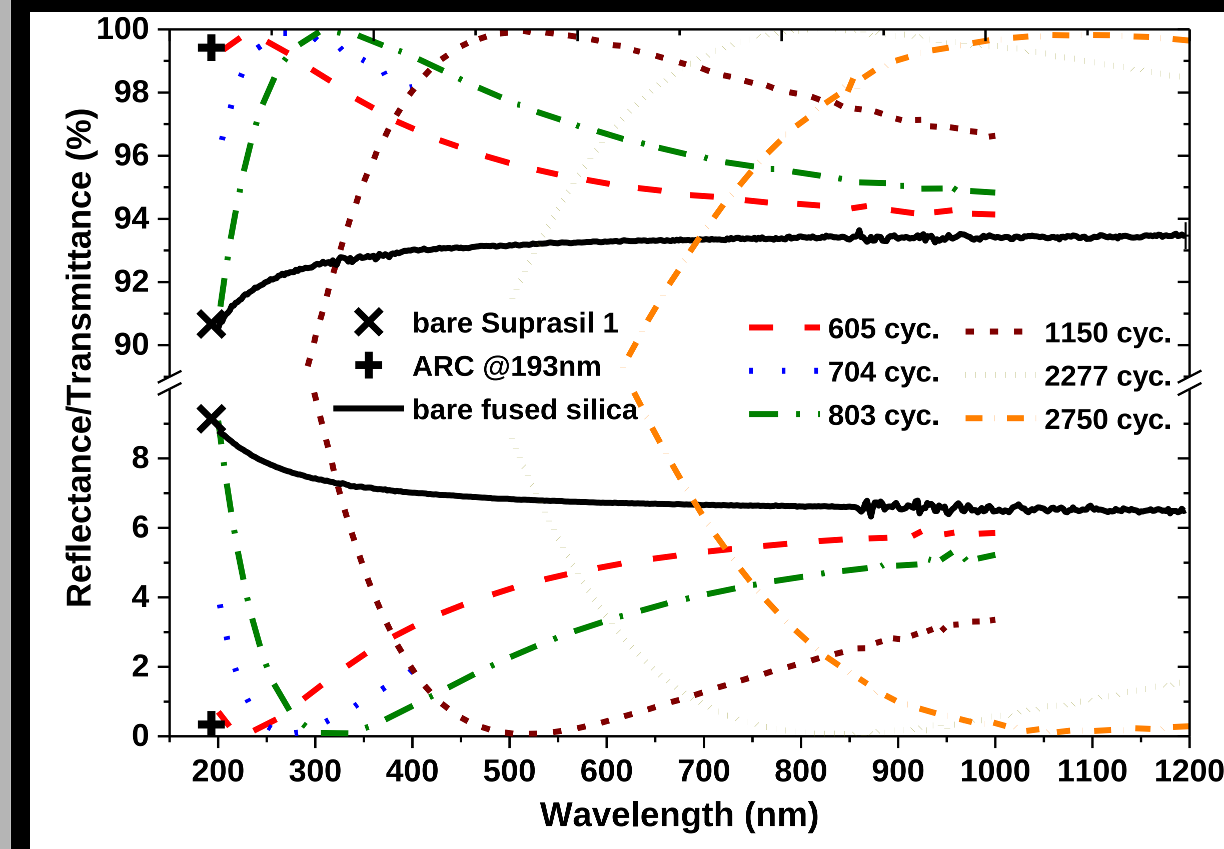


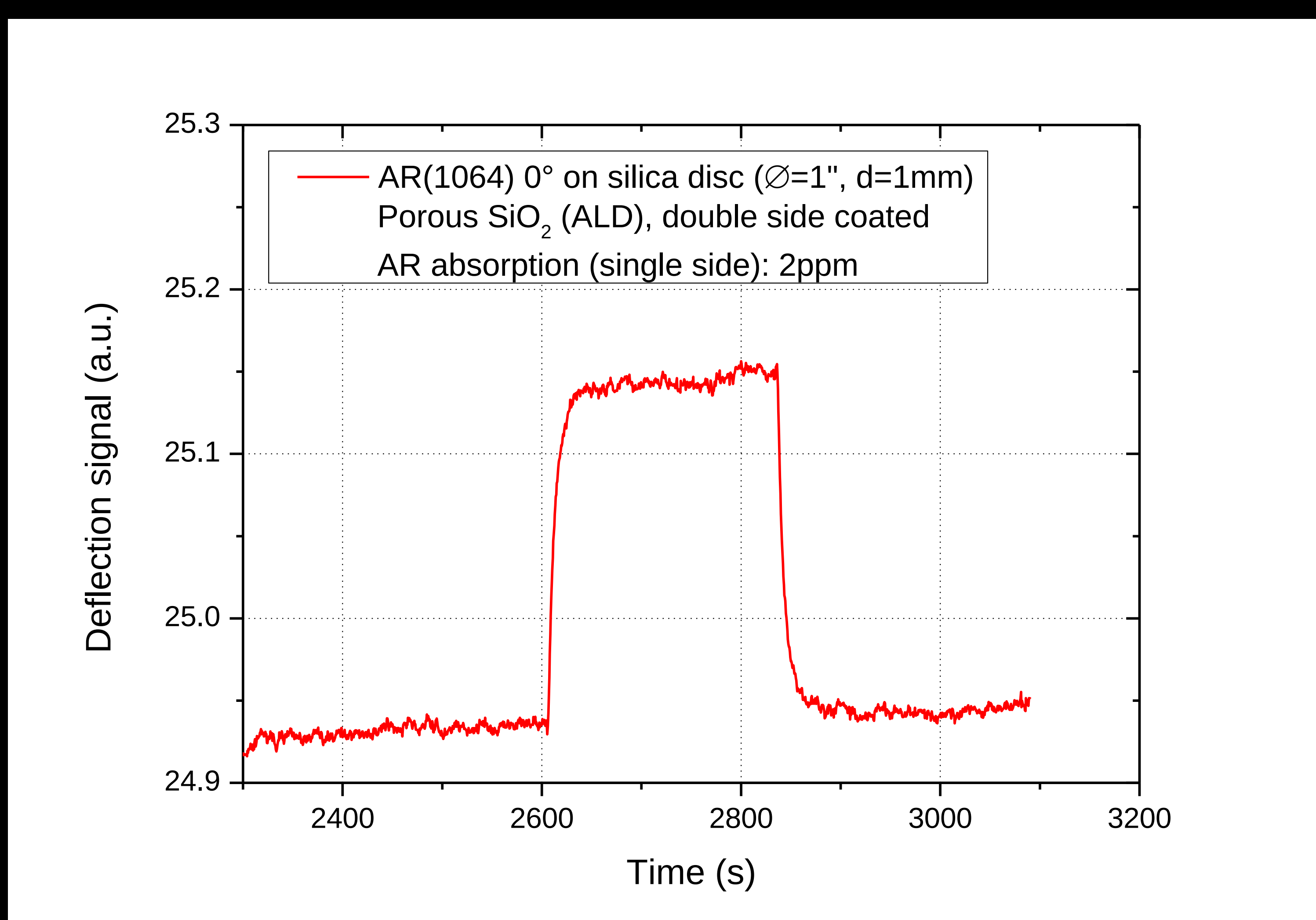
- Pump and probe beams are guided through the same surfaces, but still under 45°/90° to each other
- Easy access to both, AOI=45°and AOI=0° tests
- Double-side coated samples give better accuracy (though no access to individual coating)
- Typical disc size: ∅=1/2”…2”; d=5-10mm (→ excellent option to compare with CRD measurements at same sample)
- Standard electrical calibration

Experimental results

Nanoporous SiO_2 films (ALD) for single layer antireflection coating







- Low absorption (400ppm) for oxide AR coating at 193nm (→ no Al_2O_3)

- Absorption @1064nm (2ppm) is comparable to “common” AR coatings

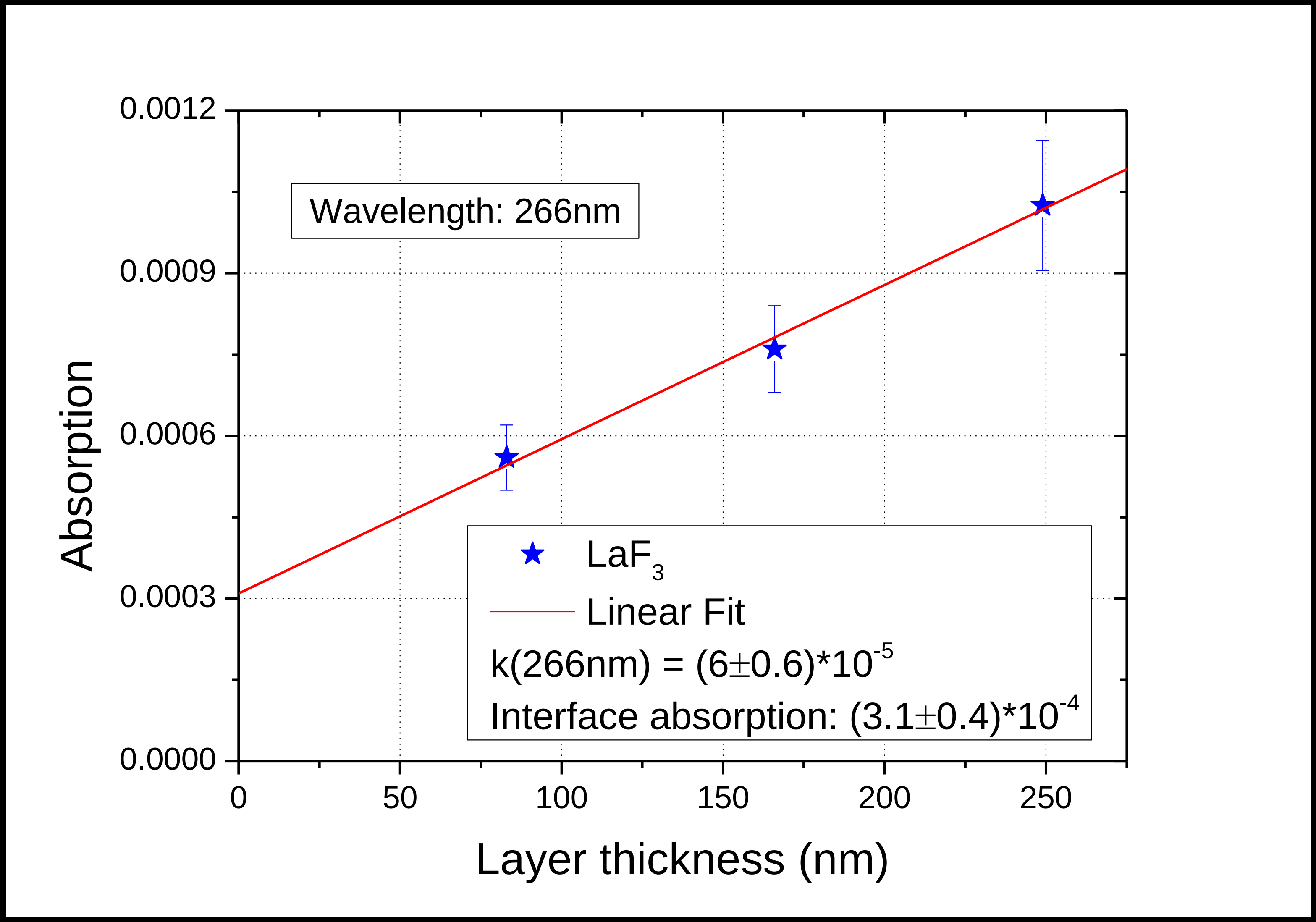
L. Ghazaryan *et. al.* „On the Properties of Nanoporous SiO_2 Films for Single Layer Antireflection Coating “, *Adv. Eng. Mater*. 2019 1801229

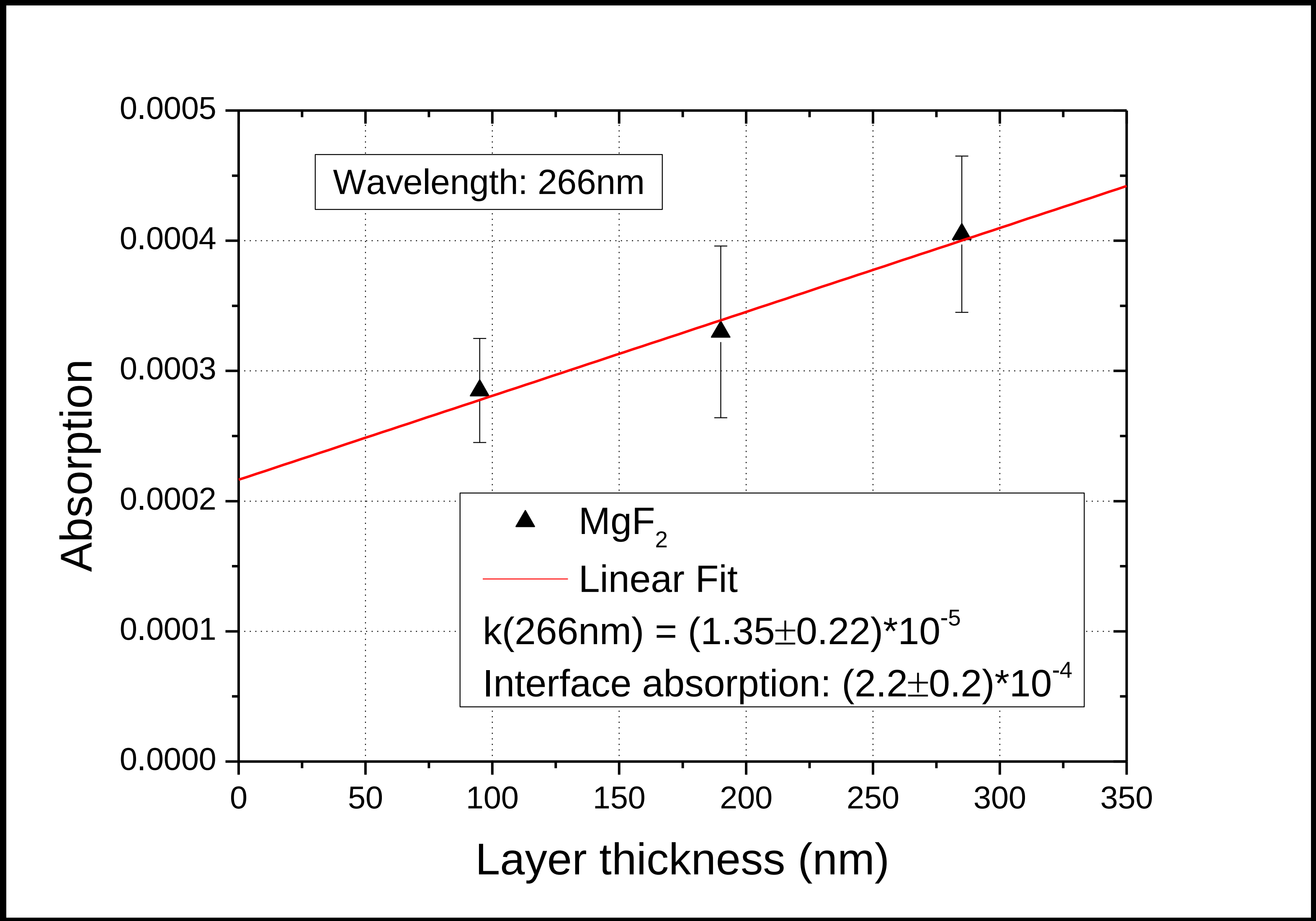
Experimental results

Separation of coating and interface absorption

- **Goal: Access to k-values for thin films**
- LaF_3 and MgF_2 single layers on thin CaF_2 discs → attached to fused silica cube → sensitivity increase by use of thermal lens in fused silica
- Uncoated CaF_2 disc for reference
- Total absorption vs. film thickness → separation of interface and layer absorption
- Important: identical CaF_2 discs, interface and layer properties

→ k-value of thin film is significantly overestimated without separation of interface effect!





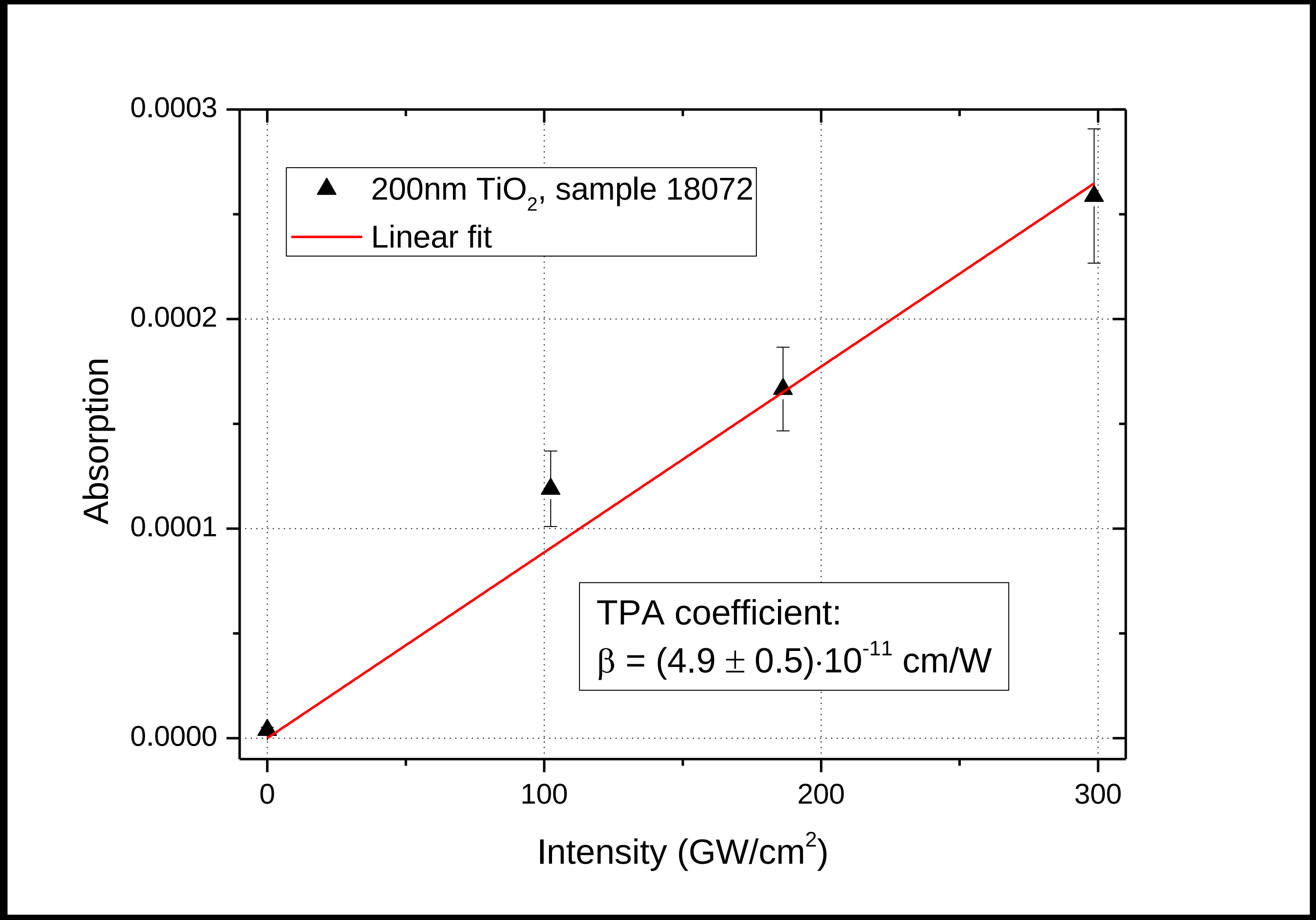
Experimental results

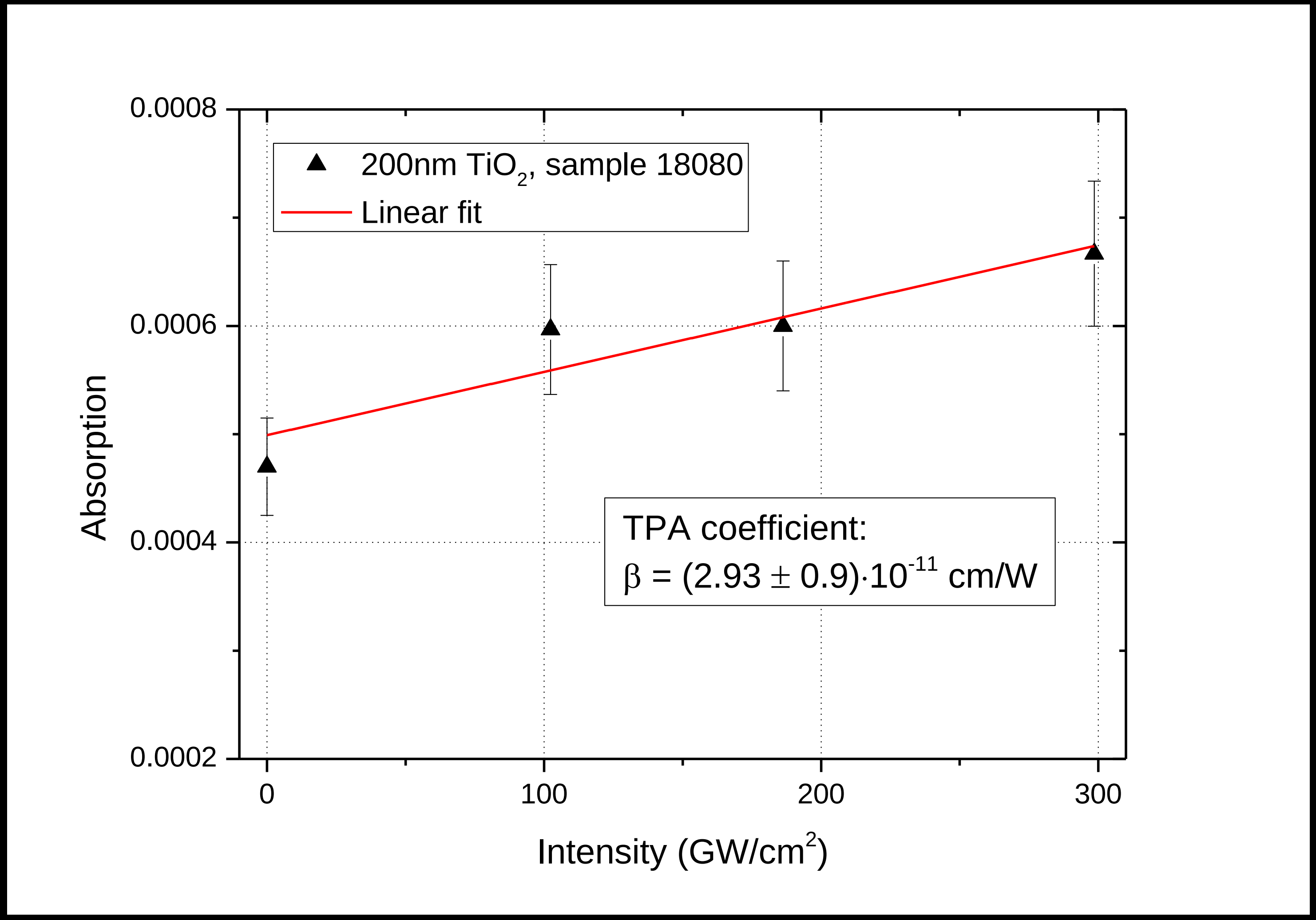
Nonlinear absorption in TiO_2 at 800nm

- **Issue:** Coating properties (e.g. reflectivity) at high intensities alter from simulated data → Nonlinear absorption/refractive index need to be taken into account in coating simulations for high intensity applications

- Single TiO_2 layers (PIAD, d=200nm) on SiO_2 substrate; λ=800nm
- "Zero" intensity → cw semiconductor laser
- High intensities → fs-laser with τ=82fs, f=1kHz (Astrella-V-F-1k, Coherent Inc.)

→ Linear increase of absorption with intensity (Beer's Law)



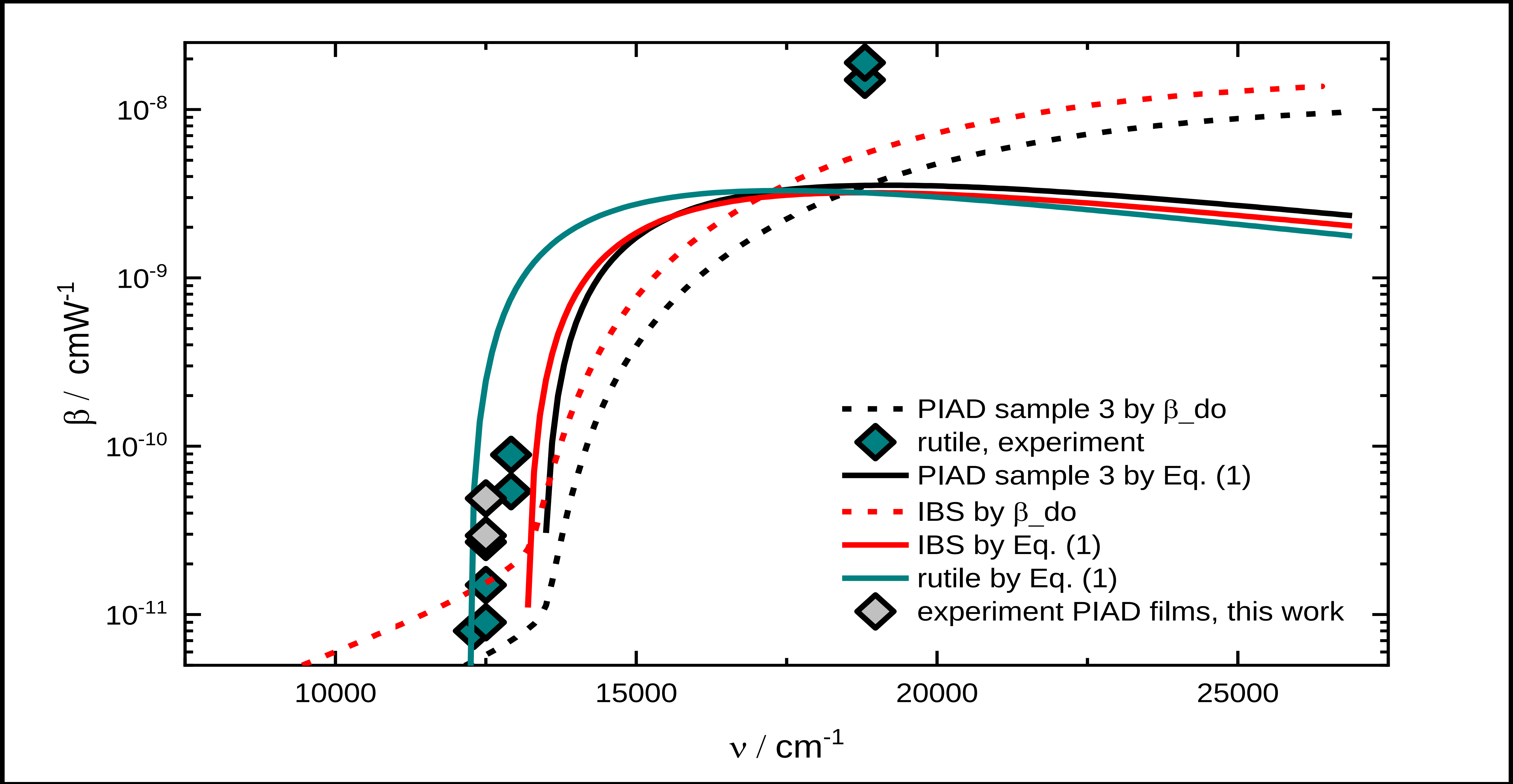


O. Stenzel *et. al.* „Linear and Nonlinear Absorption of Titanium Dioxide Films Produced by Plasma Ion-Assisted Electron Beam Evaporation: Modeling and Experiments ", *Coatings*. 2020, 10, 59, doi:10.3390

Experimental results

Nonlinear absorption in TiO_2 at 800nm

TPA coefficients from rutile and PIAD TiO_2 compared to simulations (Sheik-Bahae; Stenzel *et.al.*)



- Amorphous TiO_2 films show higher TPA coefficients than rutile → agreement with earlier experimental results for fluoride thin films at 193 nm
- Absorption is higher than expected from model → "real" vs. "ideal" thin film structure

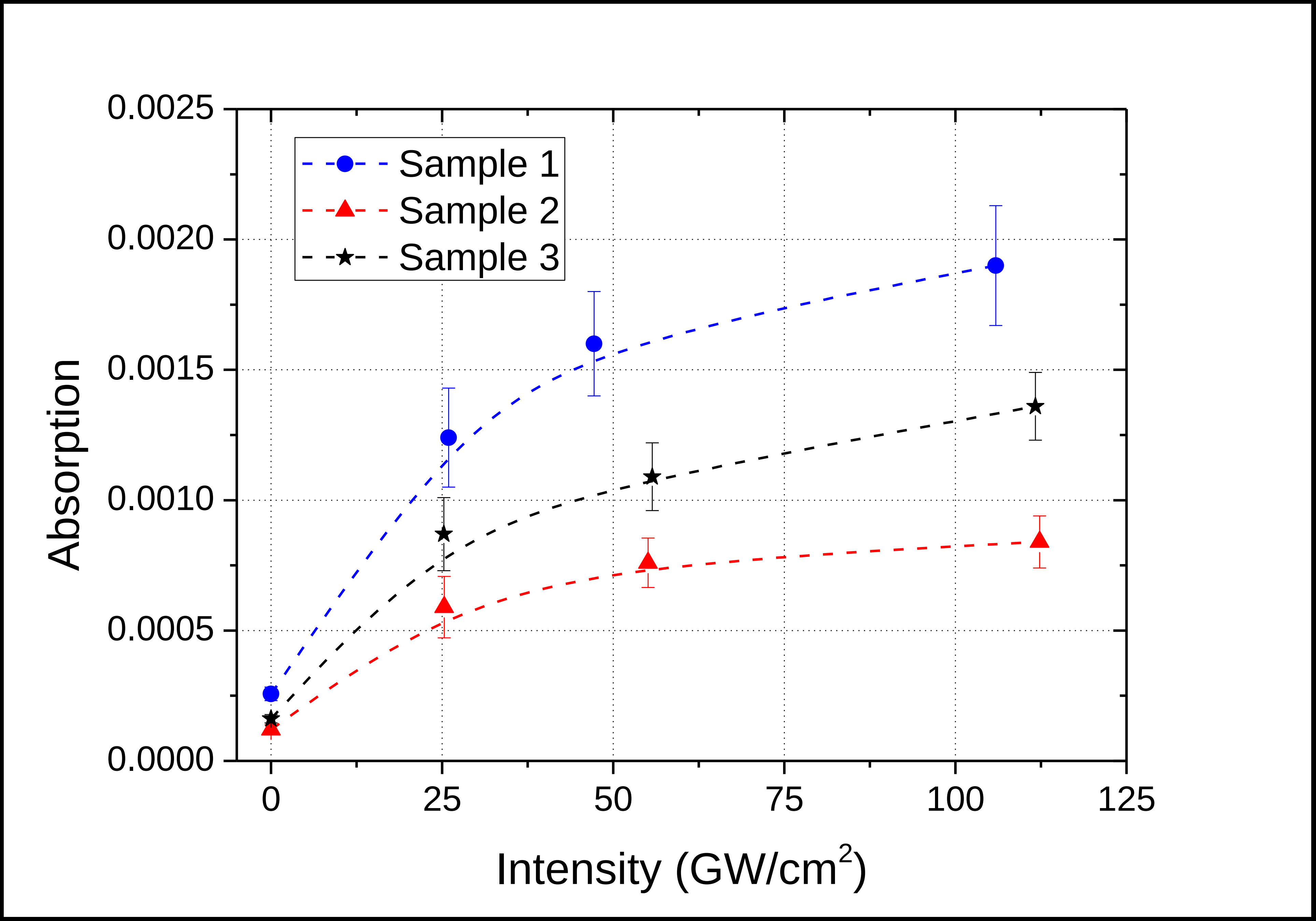
O. Stenzel *et. al.* „Linear and Nonlinear Absorption of Titanium Dioxide Films Produced by Plasma Ion-Assisted Electron Beam Evaporation: Modeling and Experiments ", *Coatings*. 2020, 10, 59, doi:10.3390

Experimental results

Nonlinear absorption in HfO_2 single layers at 400nm

- "Zero" intensity → cw semiconductor laser
- High intensities → fs-laser with τ=58fs, f=1kHz (SHG; Astrella-V-F-1k, Coherent Inc.)

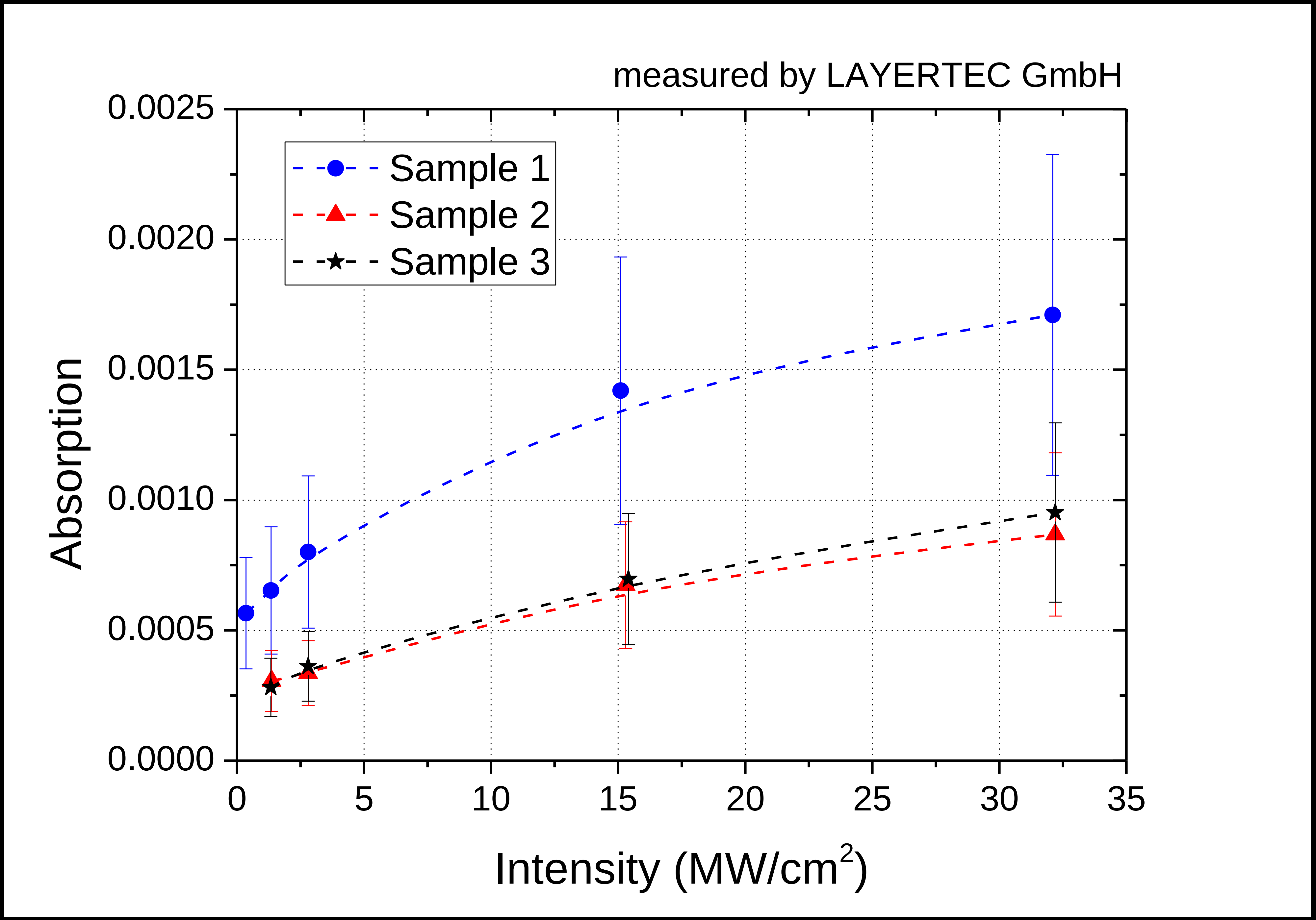
→ **Strong deviation from linear increase of absorption vs. intensity for high intensities**
→ **Determination of TPA coefficient is not straightforward**



Experimental results

Nonlinear absorption in HfO_2 single layers at 355nm

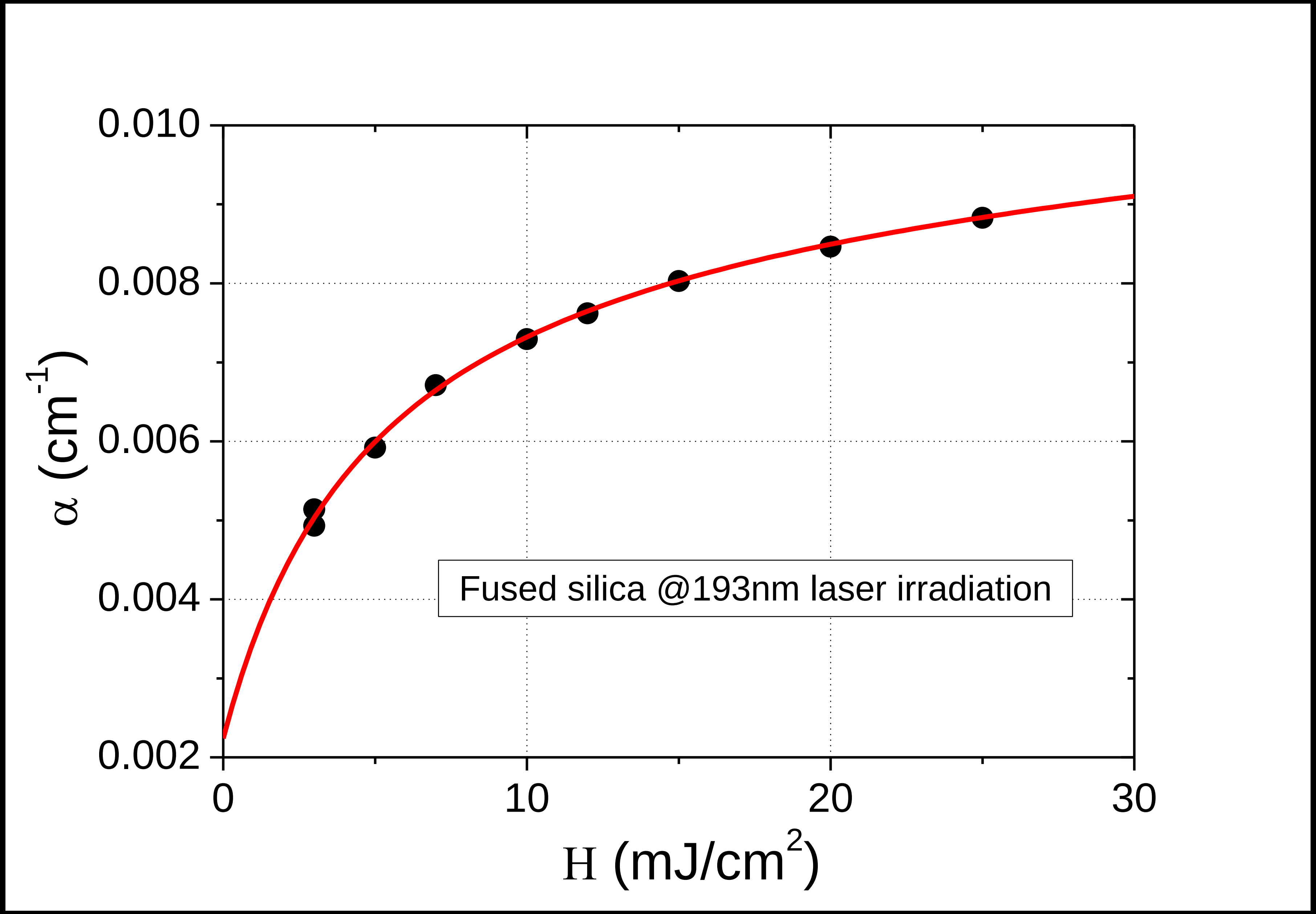
Absorption as function of laser intensity at 355nm (pulse duration: 35ns)



- Confirmation of sub-linear increase of absorption with intensity
- Explanation???

Experimental results

For comparison: Nonlinear absorption in bulk-SiO_2 at 193nm



- Bulk-SiO_2: Very similar absorption behavior which results from laser induced defect generation/annealing in addition to intrinsic two-photon absorption

→ **Similar defect structures in SiO_2 and HfO_2 (e.g. oxygen related defects)**
→ **Assumption: Absorption as function of intensity in HfO_2 is also driven by structural defects in addition to two-photon absorption**

Ch. Mühlig *et. al.* „Coefficients of stationary ArF laser pulse absorption in fused silica (type III) ", *J.-Non-Cryst. Solids*. 353 (2007), 542, doi:10.1016/j.jnoncrysol.2006.10.021

Summary

- **L**aser **I**nduced **D**eflection (LID) technique enables sensitive direct absorption measurements (photo-thermal effect)

- Unique electrical calibration procedure → absolute absorption data without knowledge of material parameters/simulations

- Access to all wavelengths - no restrictions to laser beam quality

- Nonlinear absorption in thin films:
  - TiO_2 @800nm:
    - Linear absorption increase with intensity
    - TPA coefficients in expected range compared to rutile and theoretical models
  - HfO_2 @400nm:
    - Sublinear increase of absorption with intensity
    - Confirmed by measurements @355nm
    - Similar to absorption behavior in bulk-SiO_2 @193nm
    - Assumption: Absorption as function of intensity in HfO_2 is driven by structural defects in addition to two-photon absorption

Acknowledgement

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- Layertec GmbH for measuring nonlinear absorption of HfO_2 at 355nm

- Numerous partners for their trust in our measurement technique and ongoing new challenges.

THANK YOU FOR YOUR ATTENTION!!!

Calibration (electrical)

Energy balance measurements (T+R+A+S) for calibration approval

CaF_2 samples of different quality at 193nm:

Sample	T [3cm] [%]	R (Fresnel) [%]	A [1cm] [%]	TS [3cm] [%]	T+R+TS+A [3cm] [%]
A	91.18	7.73	0.74	0.32	99.97
B	90.26	7.73	1.50	0.24	99.73
C	84.85	7.73	4.14	3.60	100.32

Transmission Measurements (IPHT) | Theoretical value | LID Measurements (IPHT) | TS Measurements (IOF)

→ No systematic error for individual characterization techniques