Spectroscopic Ellipsometry on Ta2O5/SiO2 films

(CoaCh update on optical characterization)

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Aim:
1) to find the relation between coating mechanical and optical properties
2) to understand how mechanical/optical properties depend on chemistry/ structure/morphology of the coating.

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Structure / Morphology

- Topmost surface
-Film (thickness uniformity, degree of amorphization)
- Inner interface (mixing)
- Substrate

Optical Properties

Dielectric function vs wvl

Mechanical properties

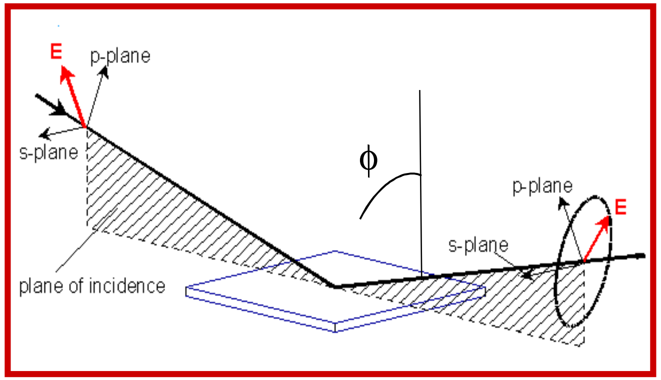
Q

Losses

Young’s modulus

Spectroscopic Ellipsometry

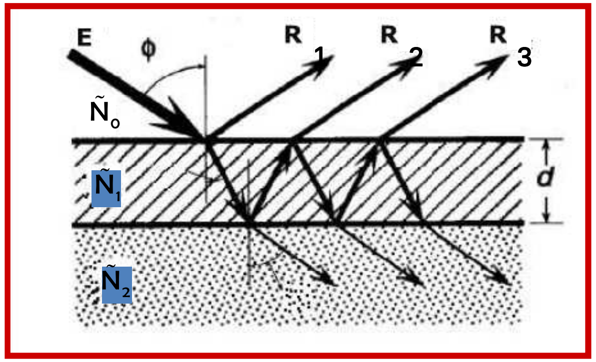
from isotropic samples



$$\begin{pmatrix} E_p^{out} \\ E_s^{out} \end{pmatrix} = \begin{pmatrix} R_p & 0 \\ 0 & R_s \end{pmatrix} \begin{pmatrix} E_p^{in} \\ E_s^{in} \end{pmatrix}$$

$$\rho = \frac{R_p}{R_s} = \left|\frac{R_p}{R_s}\right| e^{i(\delta_{rp} - \delta_{rs})} = tg\Psi e^{i\Delta}$$

Ambient / film / substrate

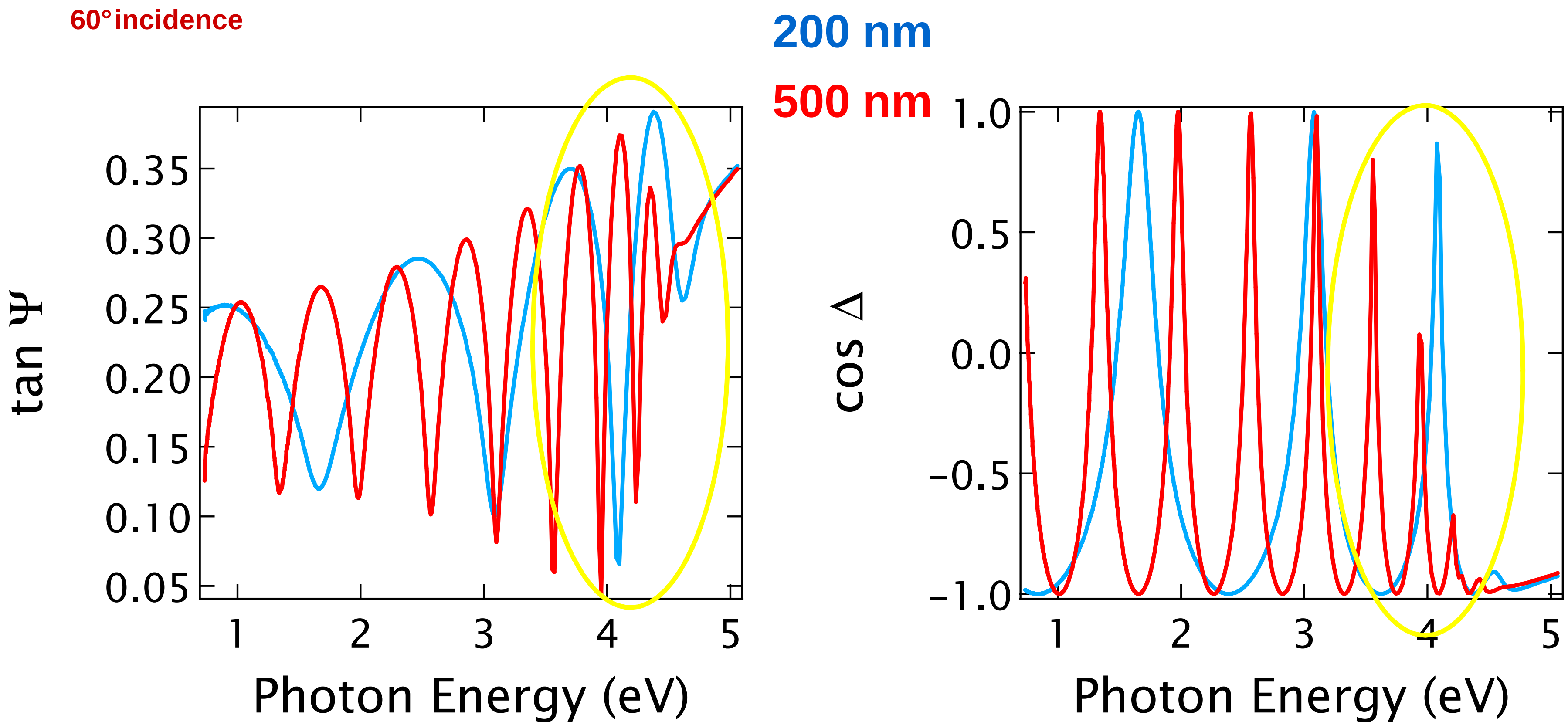


$$\rho = tg\Psi e^{i\Delta} = \frac{R_p}{R_s} = \frac{r_{0,1}^p + r_{1,2}^p e^{-2i\beta}}{1 + r_{0,1}^p r_{1,2}^p e^{-2i\beta}} \cdot \frac{1 + r_{0,1}^s r_{1,2}^s e^{-2i\beta}}{r_{0,1}^s + r_{1,2}^s e^{-2i\beta}}$$

$$\beta = 2\pi \frac{d}{\lambda} \sqrt{\tilde{N}_1^2 - \tilde{N}_0^2 \sin^2\Phi}$$

Non destructive

Ta_2O_5 **(ATFilms)** on SiO_2 corning substrates



$$\rho = \rho(\tilde{N}_0, \tilde{N}_1, \tilde{N}_2, d, \lambda, \phi)$$

Complex function of 9 real arguments

Comparison with simulations needs a model of the optical system

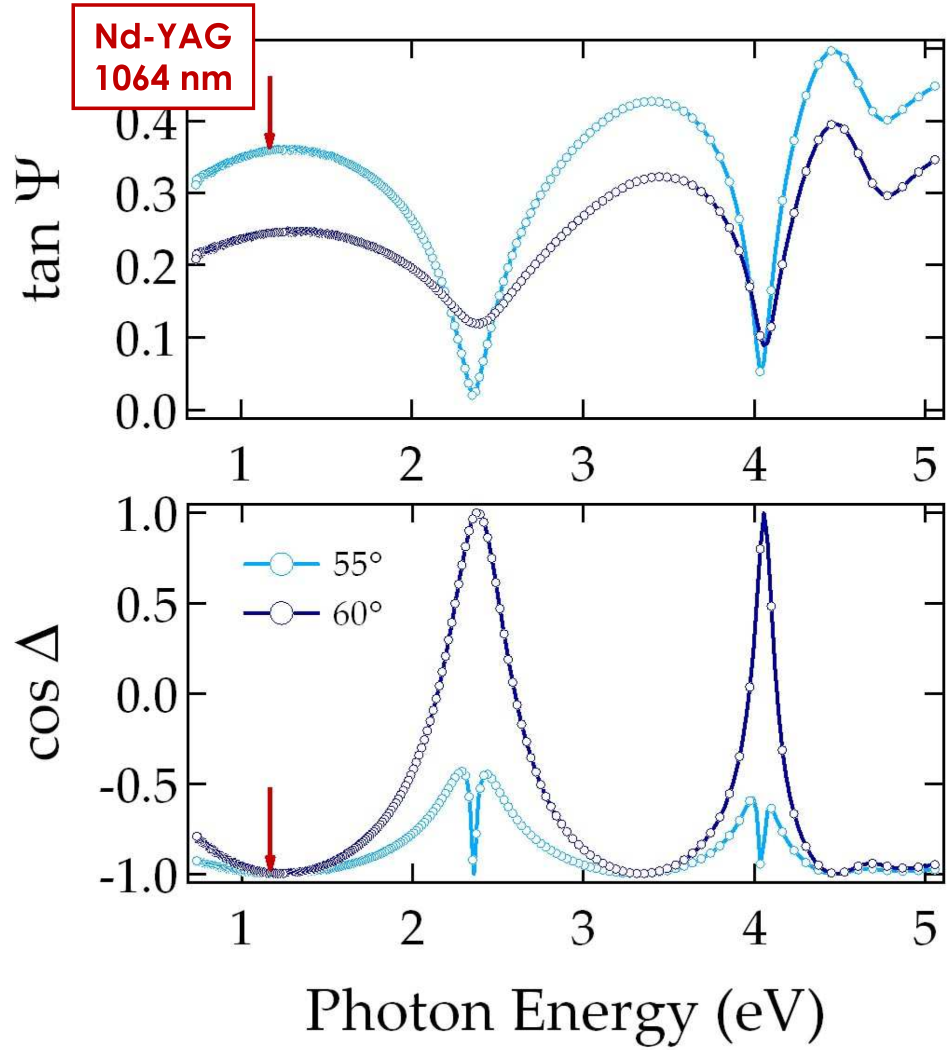
If ambient and substrate properties are known with precision you really need "only" n,k and thickness of the film

Is your film really ideal ?
- **Surface ?**
- **Substrate/film interface?**
- **bulk defects?**

Minimizing MSE

$$MSE = \frac{1}{2N-M}\sum_{i=1}^{N}\left[\left(\frac{\Psi_i^{mod}-\Psi_i^{exp}}{\sigma_{\Psi,i}^{exp}}\right)^2+\left(\frac{\Delta_i^{mod}-\Delta_i^{exp}}{\sigma_{\Delta,i}^{exp}}\right)^2\right]=\frac{1}{2N-M}\chi^2$$

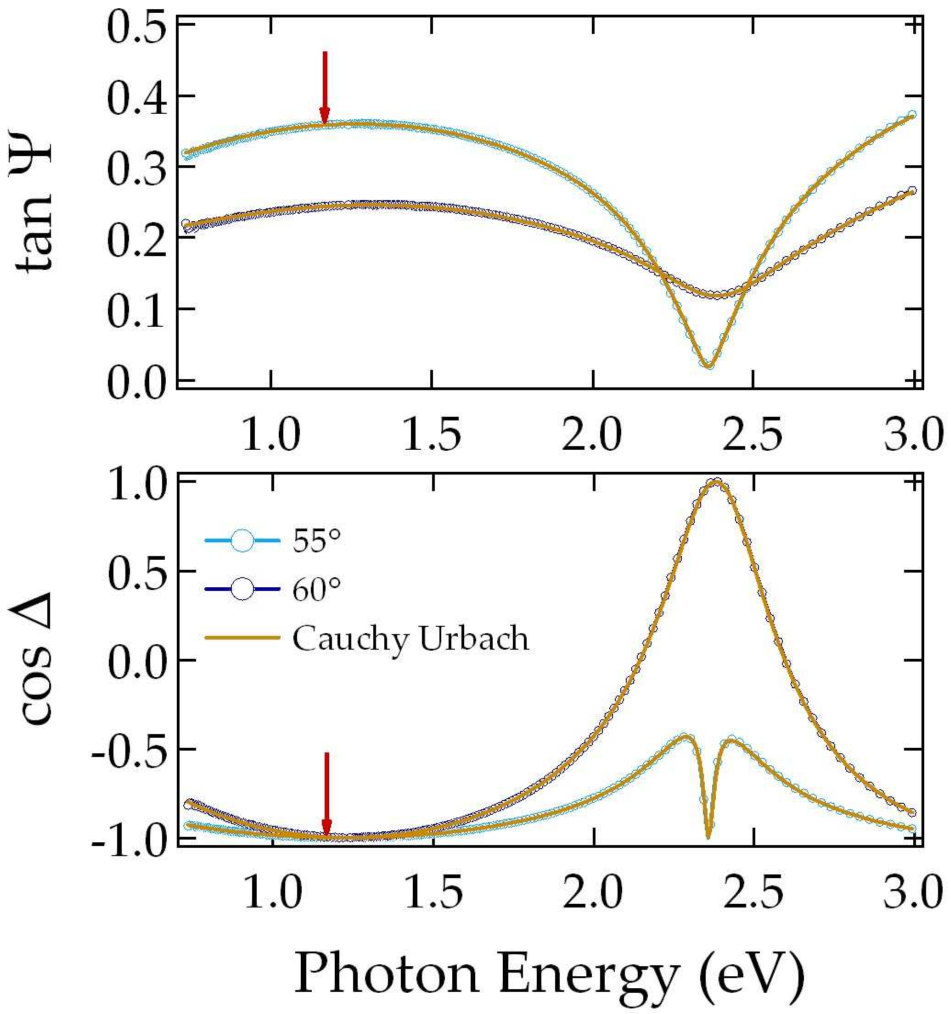
Ta_2O_5 **(LMA)** on Suprasil(Heraeus)
nominal thickness: 131 nm



Below
Above
Suprasil Brewster's Angle

Transparency region: thickness and n

No surface, no interface…



Ta_2O_5
Suprasil 311

Cauchy – Urbach model

$$n = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4}$$

$$k = D \cdot e^{F(h\nu - G)}$$

Ta_2O_5 thickness ~ **134 nm**

Ta_2O_5 n@1064nm = **2.057**

Ta_2O_5 k@1064nm ~ 8 10^{-4}

MSE = 5.05

R. Flaminio et al.
Class.Quantum Grav. **27** (2010) 084030

Optical index ~ 2.06

Monolayer absorption ~ 1.2 ppm (i.e. k ≈ 2 10^{-7})

1. Extend the analysis to the whole energy range: 0.75-5 eV (245-1700 nm)

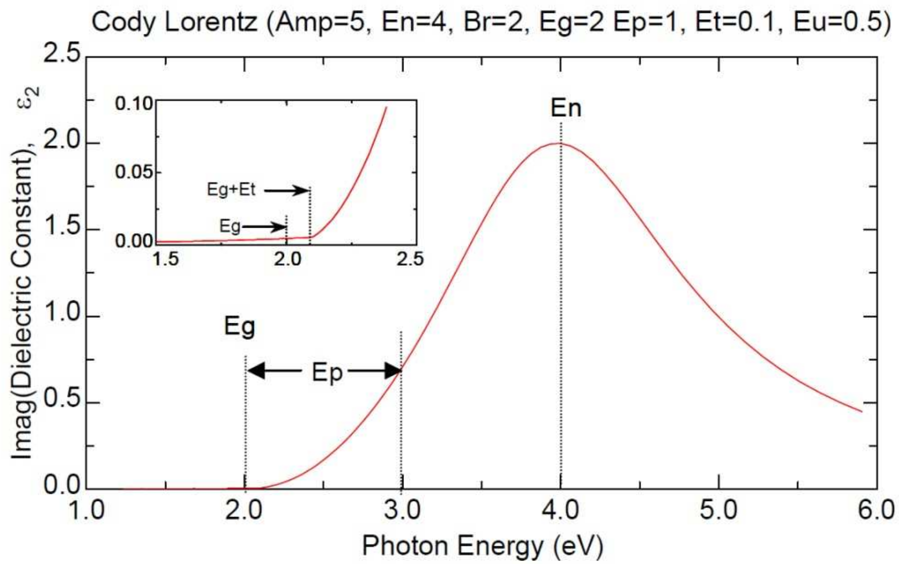
Amorphous oxides: Cody-Lorentz model + Urbach tail

[A. S. Ferlauto et al., Journal of Applied Physics 92 (2002)]

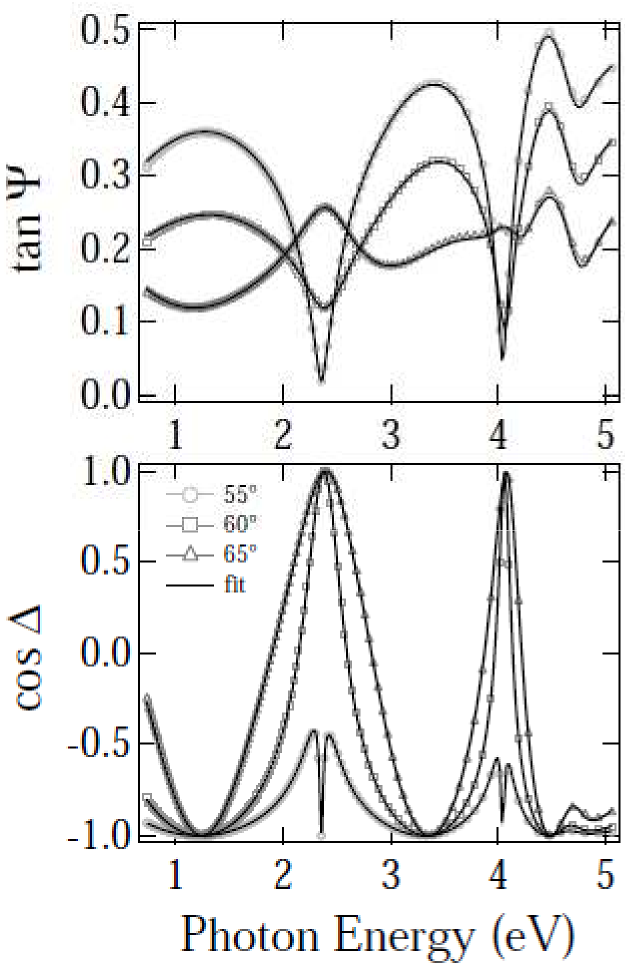
KK consistent

$$\varepsilon_2(E) = \begin{cases} \left(\dfrac{E_1}{E}\right) e^{\left[\frac{(E-E_g-E_t)}{E_U}\right]} & 0 < E \le (E_g + E_t) \\[2ex] \dfrac{(E-E_g)^2}{(E-E_g)^2 + E_p^2} \dfrac{A E_0 \Gamma E}{[(E^2-E_0^2)^2 + \Gamma^2 E^2]} & E > (E_g + E_t) \end{cases}$$

$$\varepsilon_1(E) = \varepsilon_1(\infty) + \frac{2}{\pi} P \int_{E_g}^{\infty} \frac{\xi \varepsilon_2(\xi)}{\xi^2 - E^2} d\xi$$



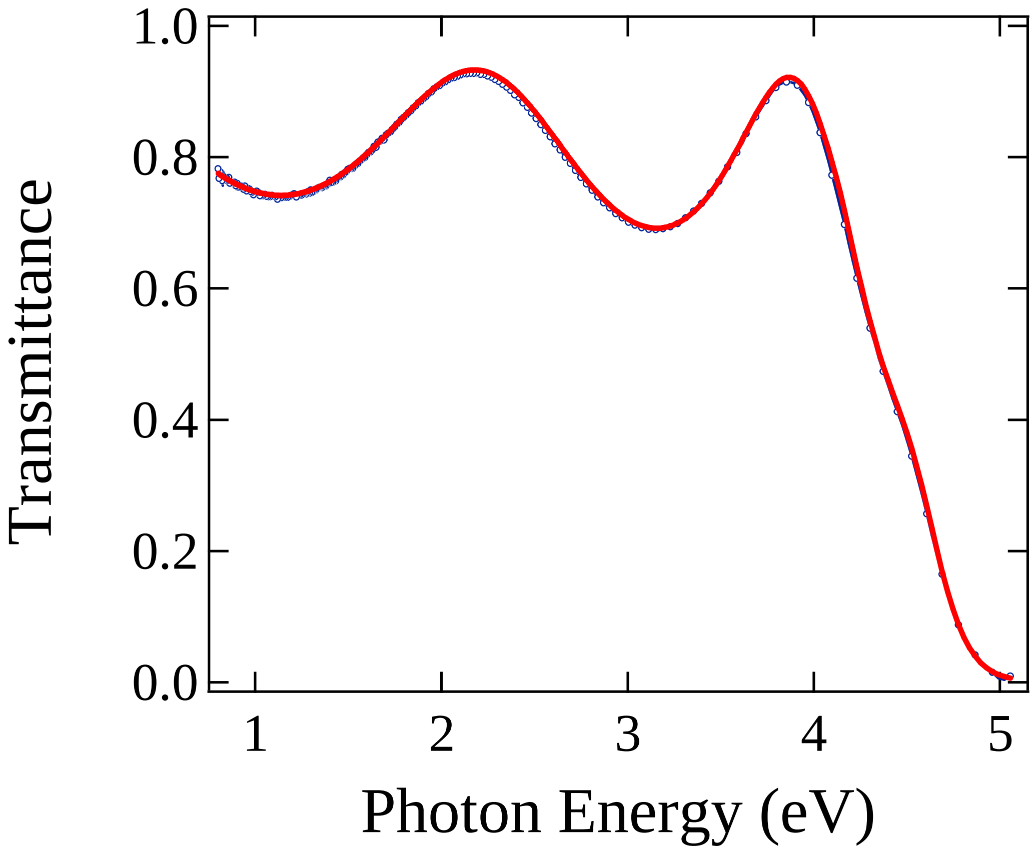
2. The "simplest model" : Roughness layer + transition layer



Roughness layer **~ 0.25 nm**
Ta_2O_5 ~ **131 nm**
Intermix 50:50 **~ 6 nm**
Suprasil 311

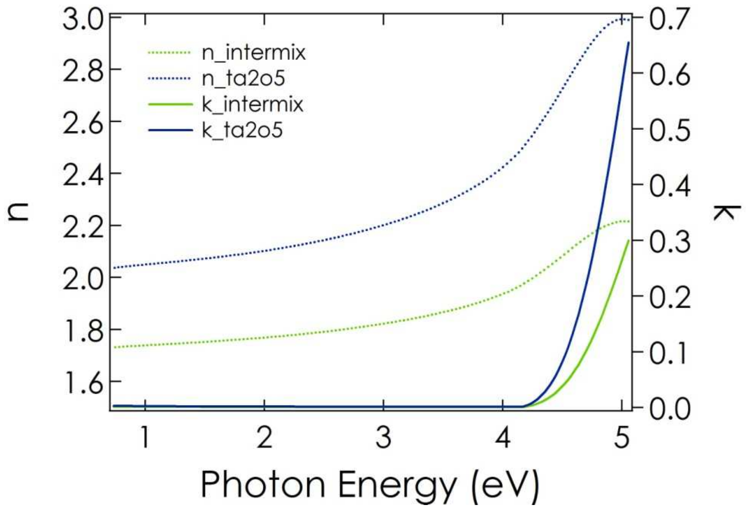
MSE =10.15

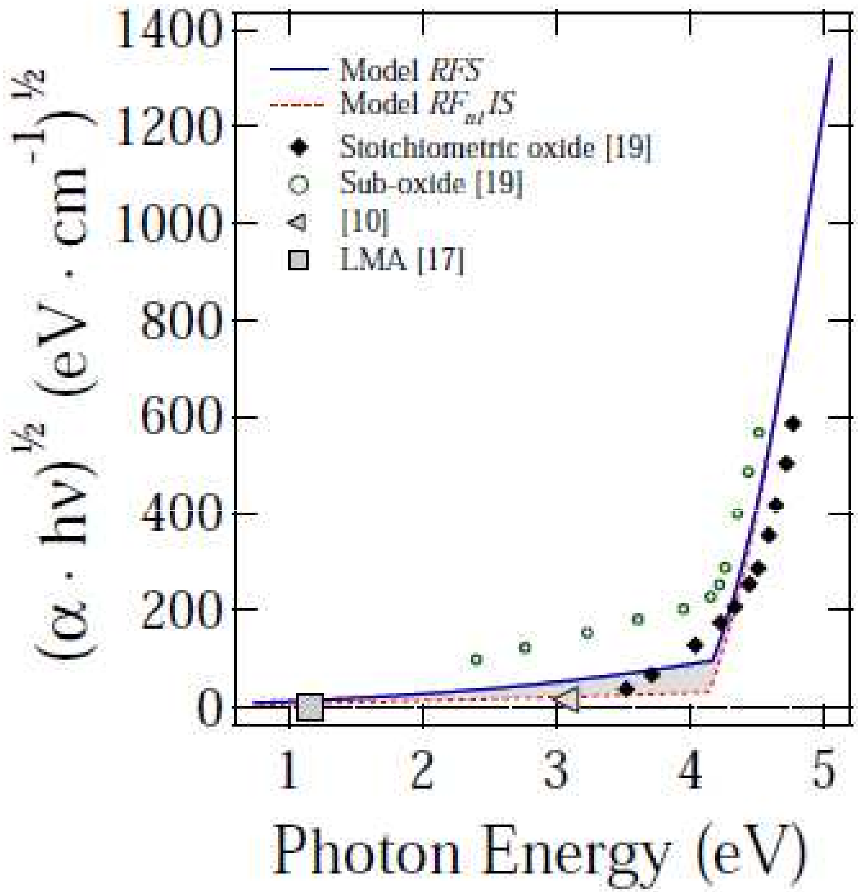
The model fits transmittance measurements too



Tauc plot

M. Prato et al. to appear on Thin Solid Films





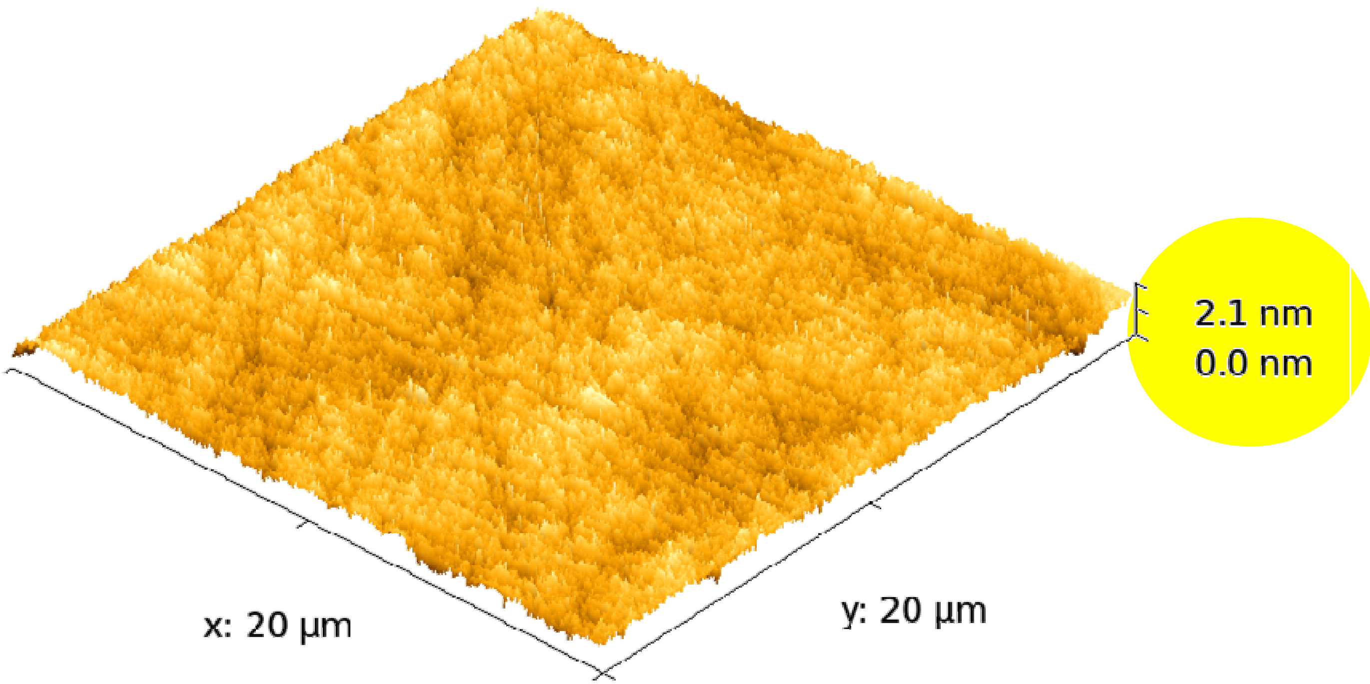
n = 2.057
@1064 nm

Gap 4.11-4.17 eV

Sub-stoichiometric

Surface layer Morphology

Atomic Force
Microscopy (AFM)



RMS roughness ~ 0.2 nm

Surface region chemical composition XPS

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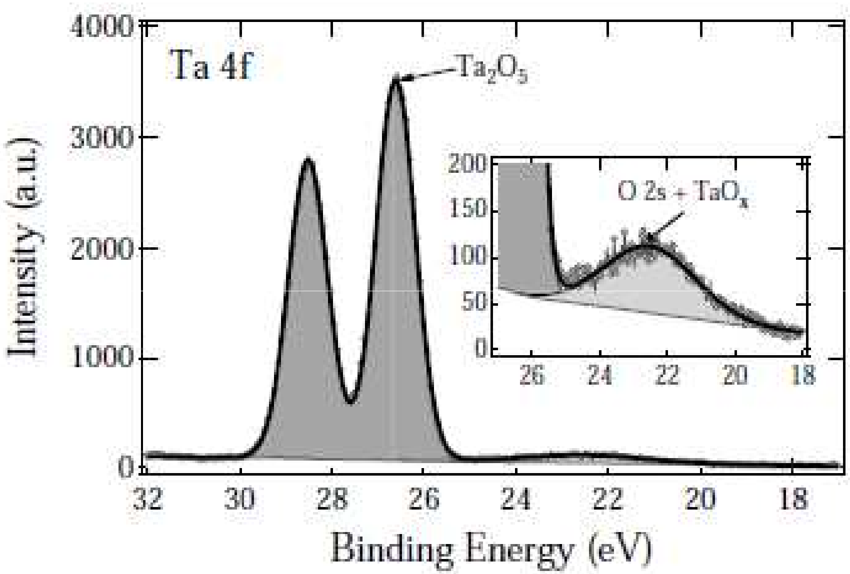


The low BE peak in the XPS spectrum is likely due to O2s photoelectrons and to non-stoichiometric Ta_2O_5 species, possibly including also metallic Ta (a few %).

Need for higher resolution: synchrotron

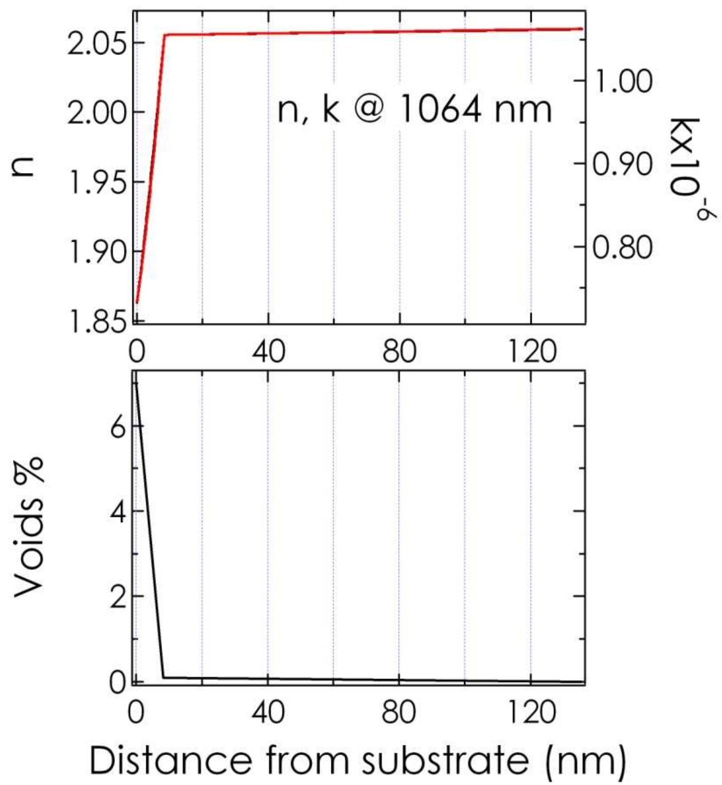
3. From "transition" to interface layer: graded distribution of voids

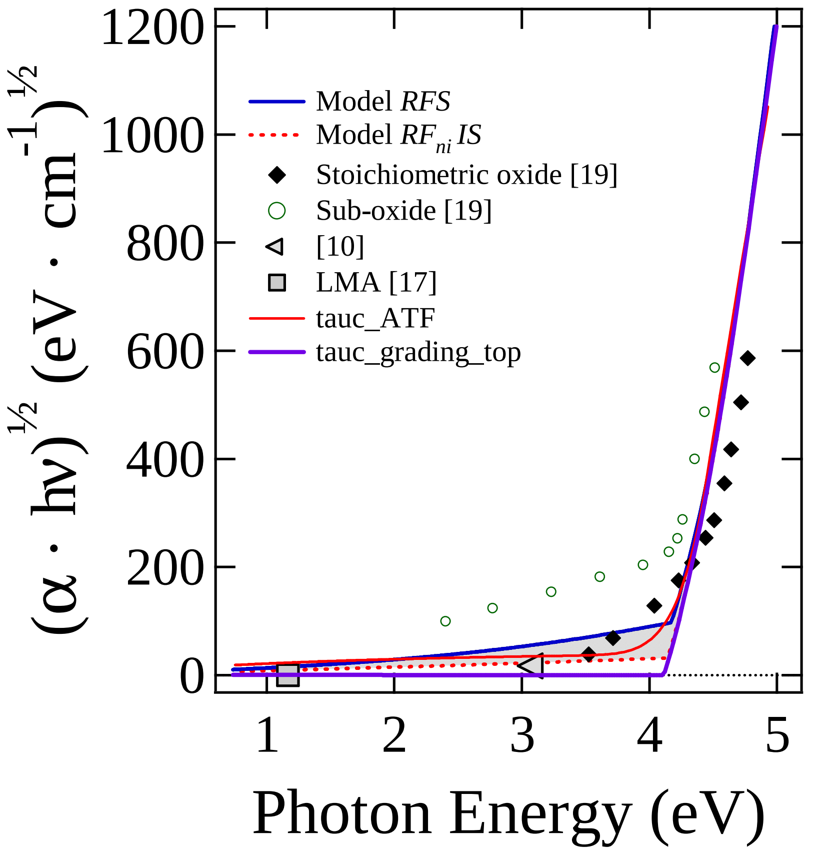
Roughness layer ~ **0.4 nm**
Ta_2O_5 ~ **136 nm** (density increasing with thickness)
Suprasil 311

MSE (voids) = 8.96

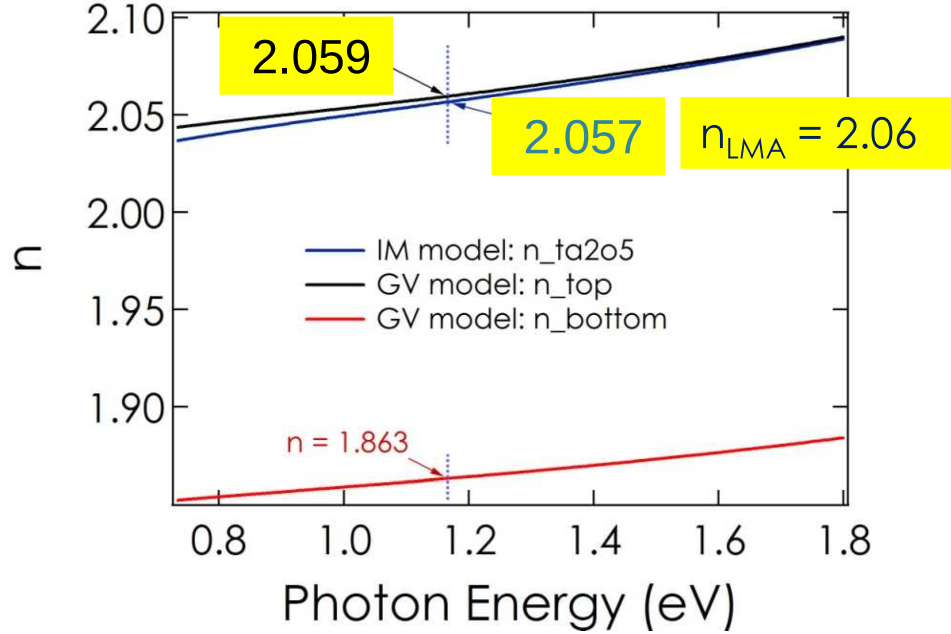
vs

MSE (IM) =10.15

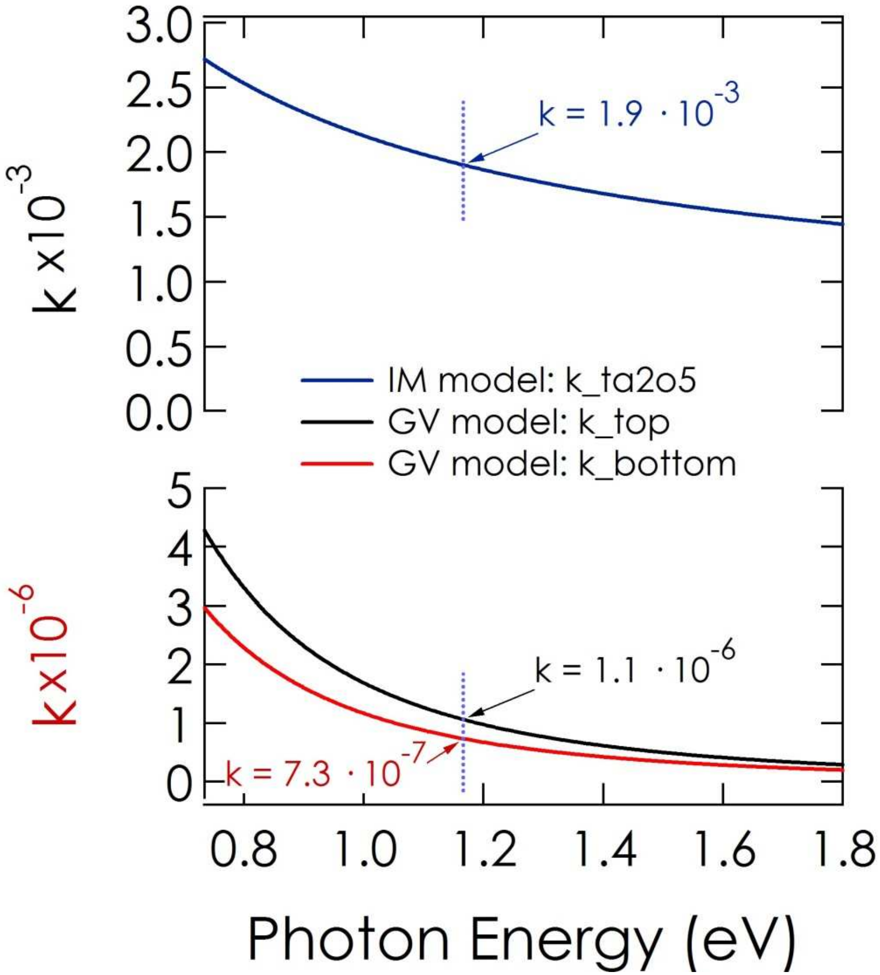




1200
1000
800
600
400
200
0
1
2
3
4
5
Photon Energy (eV)
(α · hν)^½ (eV · cm^-1)^½
Model RFS
Model RF_ni IS
Stoichiometric oxide [19]
Sub-oxide [19]
[10]
LMA [17]
tauc_ATF
tauc_grading_top



2.10
2.05
2.00
1.95
1.90
0.8
1.0
1.2
1.4
1.6
1.8
Photon Energy (eV)
n
2.059
2.057
n_LMA = 2.06
IM model: n_ta2o5
GV model: n_top
GV model: n_bottom
n = 1.863



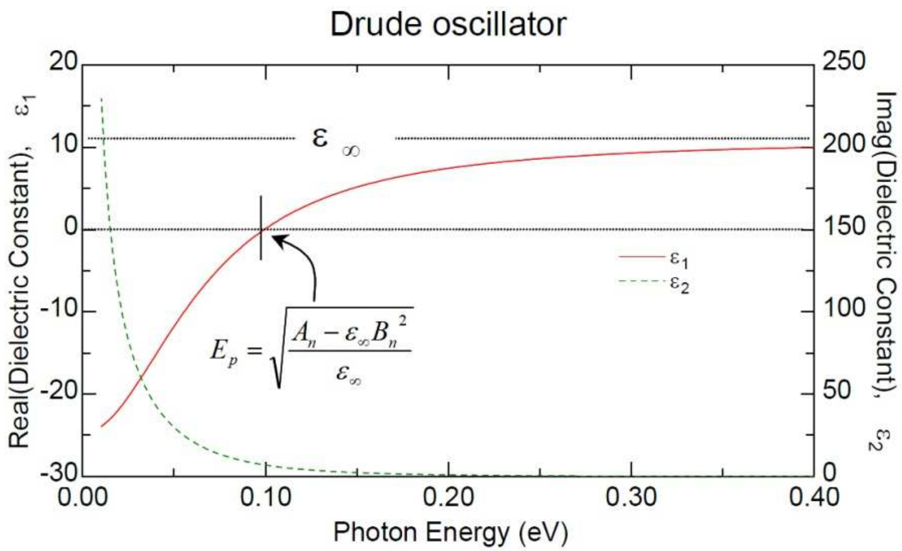
Need for more sensitive methods
Photothermal Deflection Spectroscopy
on the same samples

Improving (not so much) the SE interpretation

4. Including defect-related free carriers absorption @ lowest energy

Free carriers: Drude oscillator

$$\tilde{\varepsilon}(E) = \varepsilon_\infty - \frac{A}{E^2 + iBE}$$



Conclusions/Perspectives

SE:

- **gap value + n value + XPS : non-stoichiometric oxide species**
- **Intermixing @ interface <u>and/or</u> diffused voids can improve the interpretation of the optical response of the Ta_2O_5 layer.**

- **Converging on Q-meas. vs optical measurements (SE + PTDS for k @1064 nm)**
- **SE investigations (broader energy range in the UV) and Higher resolution XPS (synchrotron) needed to improve models**
- **Interface layer: measurements on samples of reduced thickness Independent info by TEM**
- **…..**
- **SE/Transmission @ at cryogenic temperatures**

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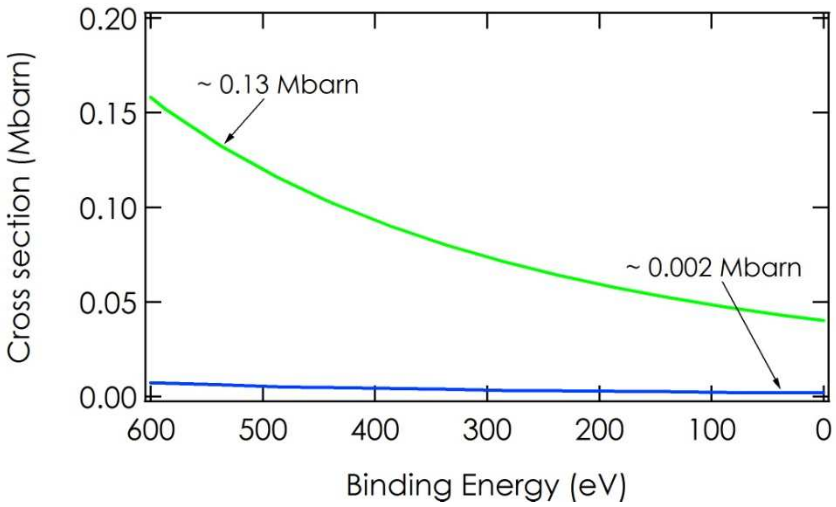
Improving the XPS interpretation



O1s vs. O2s cross-sections

http://ulisse.elettra.trieste.it/services/elements/WebElements.html