

Optical properties of titania-tantala coatings at cryogenic temperatures

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Motivation

The behavior of GWD mirrors at cryo temperatures might be different from that at room temperature:

- T-dependent properties: coating, substrate
- Ambiental conditions: 'ice', adsorbates

Information about the properties of the coatings at cryo temperatures is limited.

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A **new setup for cryo-optic measurements** is being developed at the Università di Genova – INFN Genova, with the aim to **systematically study the optical properties** of coatings and substrates at cryo temperatures.

The setup



Cryostat with optical access (500 to 4.2 K)

- small inner volume
- can be used with nitrogen or helium
- high vacuum (10^{-6} mbar) to mitigate ice formation
- 2 sets of windows

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Spectroscopic ellipsometer (190-2500 nm)

- Large spectral range available (UV-vis-IR) with very good spectral resolution (1 datapoint every nm)
- Fully automated measurement

The samples

I) Crystalline silicon substrate

Silicon is the most suitable substrate for ellipsometry measurements

II) Titania-tantala coating, NOT annealed

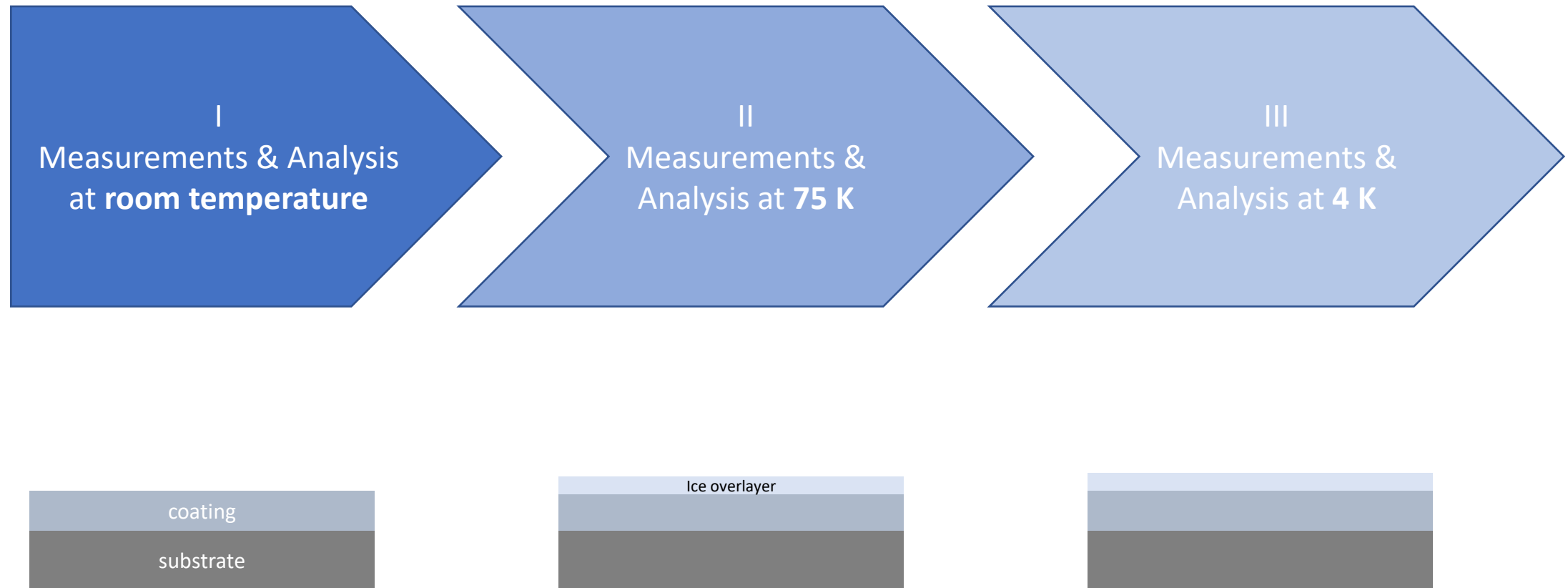
- Ti/(Ti+Ta): 0.21
- Thickness: 503 nm
- Substrate: silicon wafer

III) Titania-tantala coating, annealed

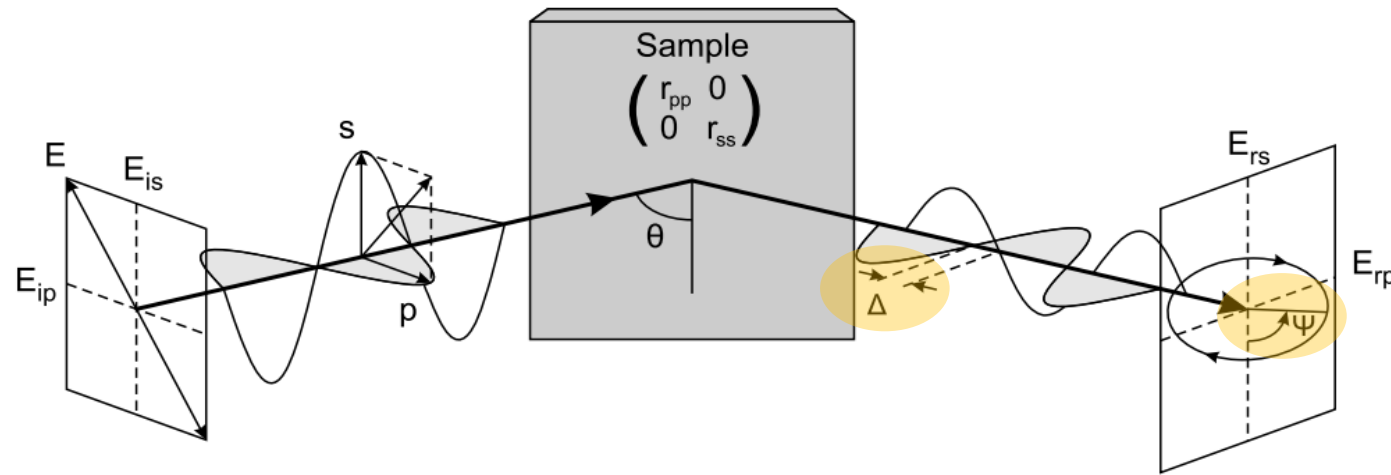
- Ti/(Ti+Ta): 0.21
- Thickness: 513 nm
- Substrate: silicon wafer

Titania-tantala coatings produced and annealed (10 h, 500 °C) at LMA

Strategy



Ellipsometry: the basics



M. Canepa

$$\frac{\tilde{r}_p}{\tilde{r}_s} = \tan \Psi \exp(i \Delta)$$

$$\Psi = \tan^{-1} \left(\frac{|r_p|}{|r_s|} \right)$$

$$\Delta = \delta_{rp} - \delta_{rs}$$

A model is required to extract information of interest (thickness, refractive index, energy gap...) from the ellipsometry measurements

In very thin transparent films, the Delta variation is proportional to the film thickness, while Psi is almost unchanged. In particular, **Delta decreases for increasing film thickness**

Cooling the samples: real-time monitoring

Cooling rate & initial pressure have effects over the ice formation:

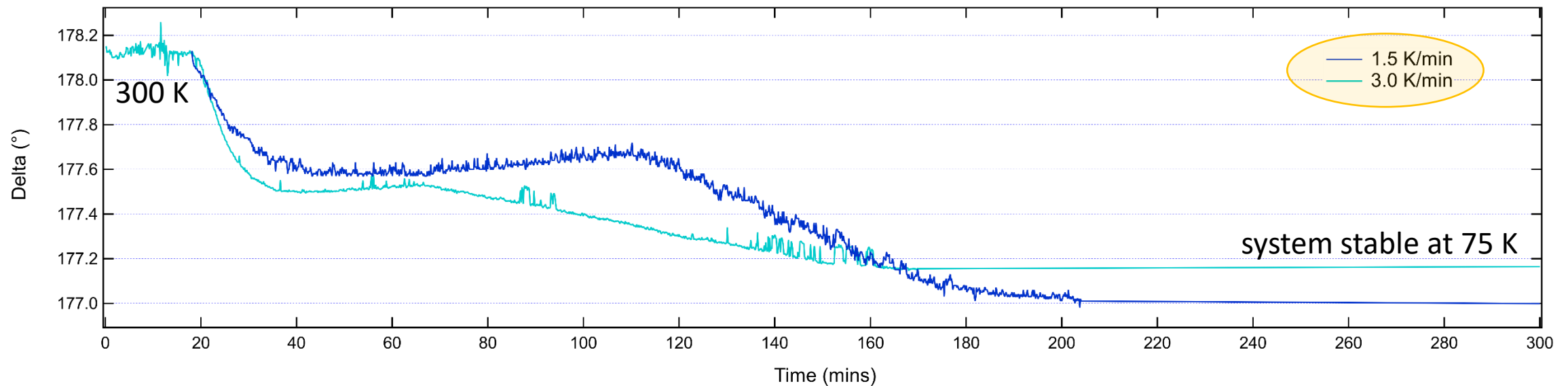
- Thickness
- Time required to stabilize at the lowest temperature

Cooling the samples: real-time monitoring

Cooling rate & initial pressure have effects over the ice formation:

- Thickness
- Time required to stabilize at the lowest temperature

Higher cooling rates yielded **thinner overlayers** and quicker stabilization. Delta variation is not linear during cooling.



The ice issue: general remarks

Ice is a material that can exhibit many different forms (structure, density...) depending on the conditions in which it is formed.

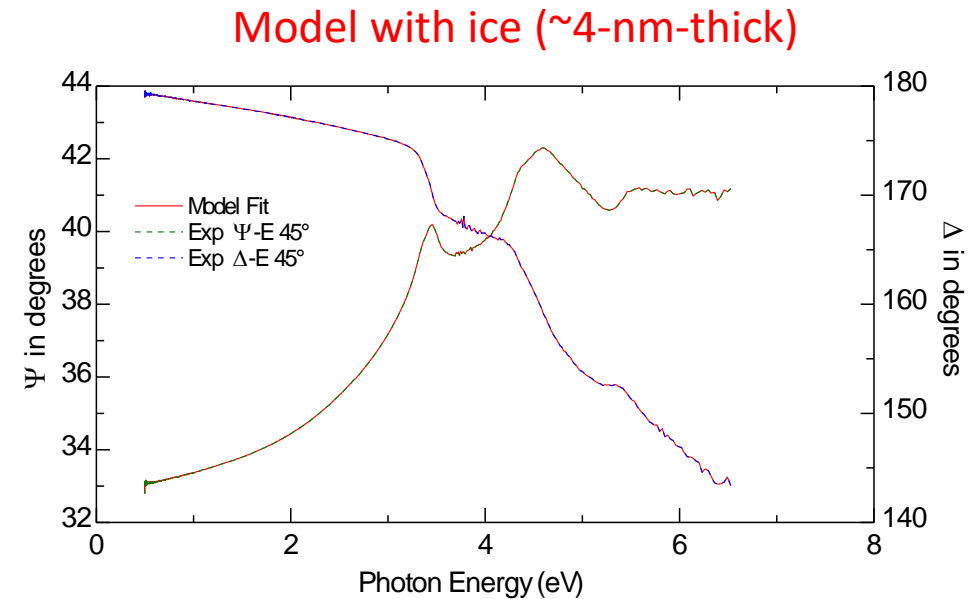
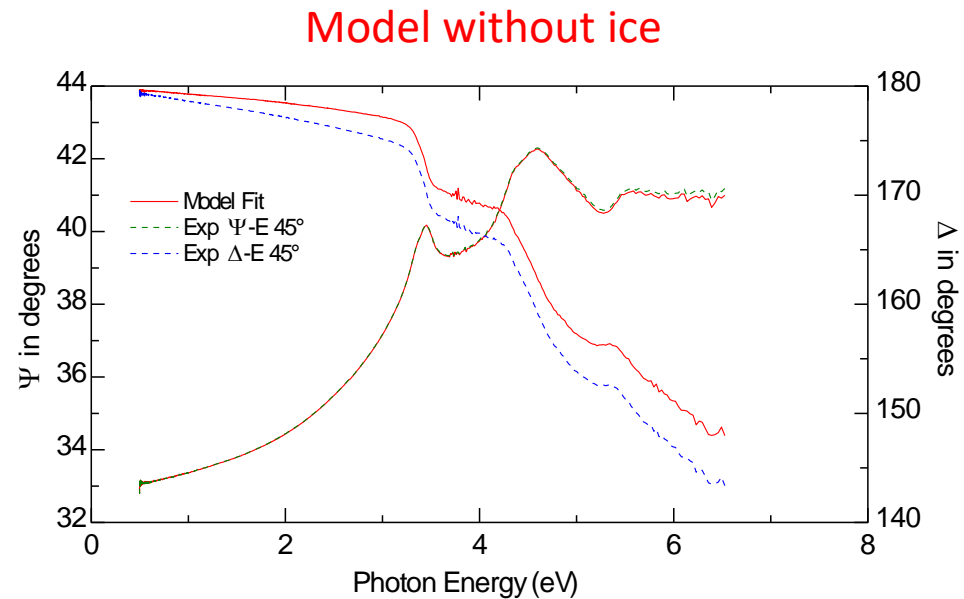
Form	Density $\left(\frac{g}{cm^3}\right)$	Temperature Range (K)
I_h	0.92	> 160
I_c	0.93	136 - 160
LDA	0.94	15 - 136
HDA	1.17	< 15

J. M. Labello, PhD Thesis University of Tennessee, 2011

Relevant facts:

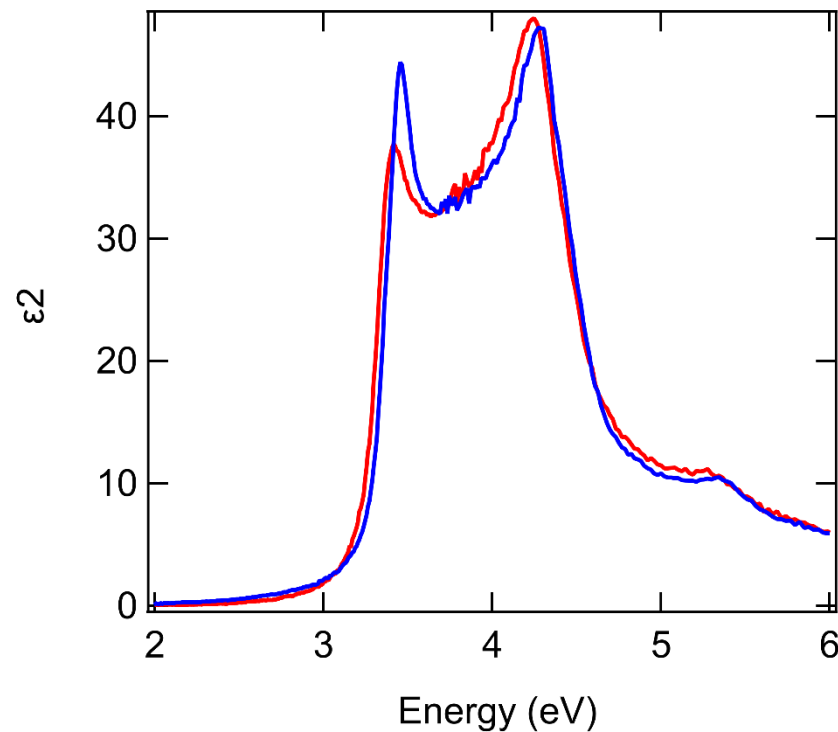
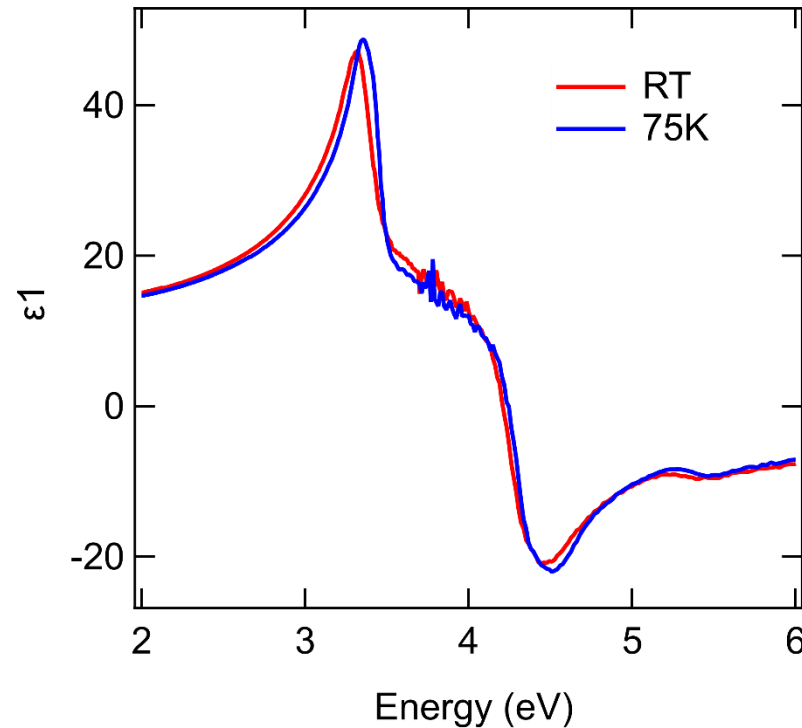
- I_h is the ice found in nature (the one most studied)
- I_h and I_c have 'practically identical' optical properties
- I_h has very low absorption coefficient ($<10^{-5}$ around $1 \mu m$). Birefringence is very slight.

The ice layer: modelling



- The presence of ice can be effectively modeled with a Cauchy formula. Once the ice layer is properly taken into account, **it no longer affects the analysis of the underlying coating.**

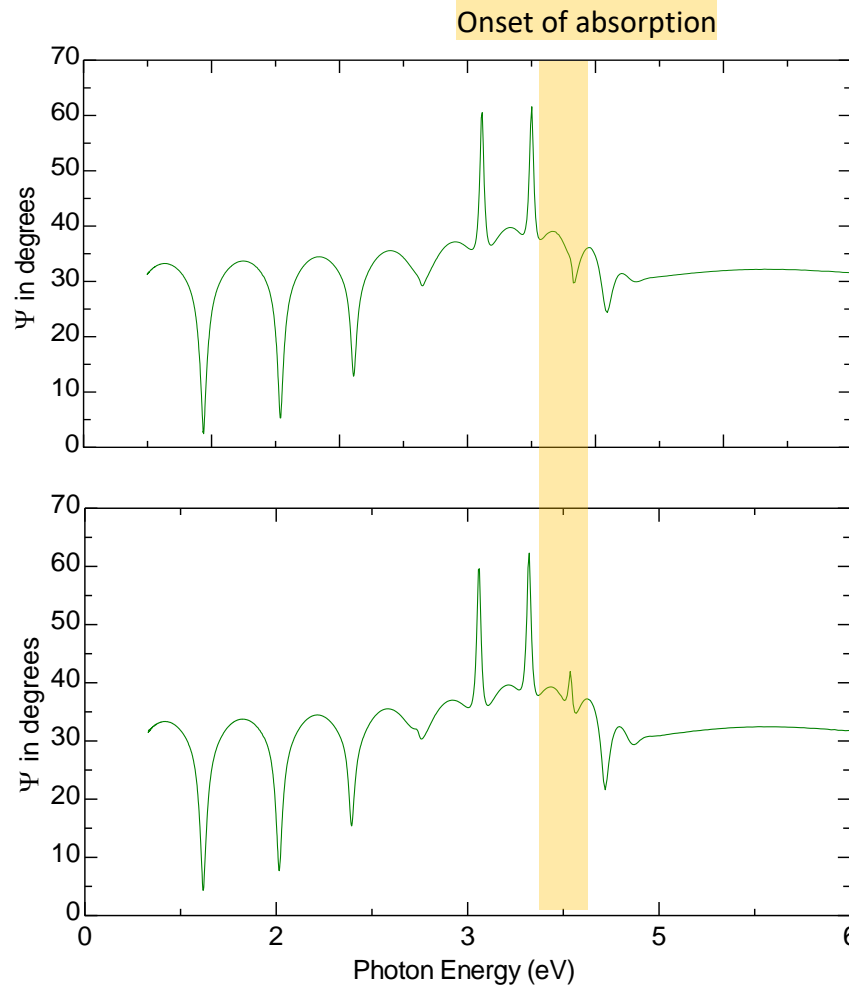
Substrate T-induced variations



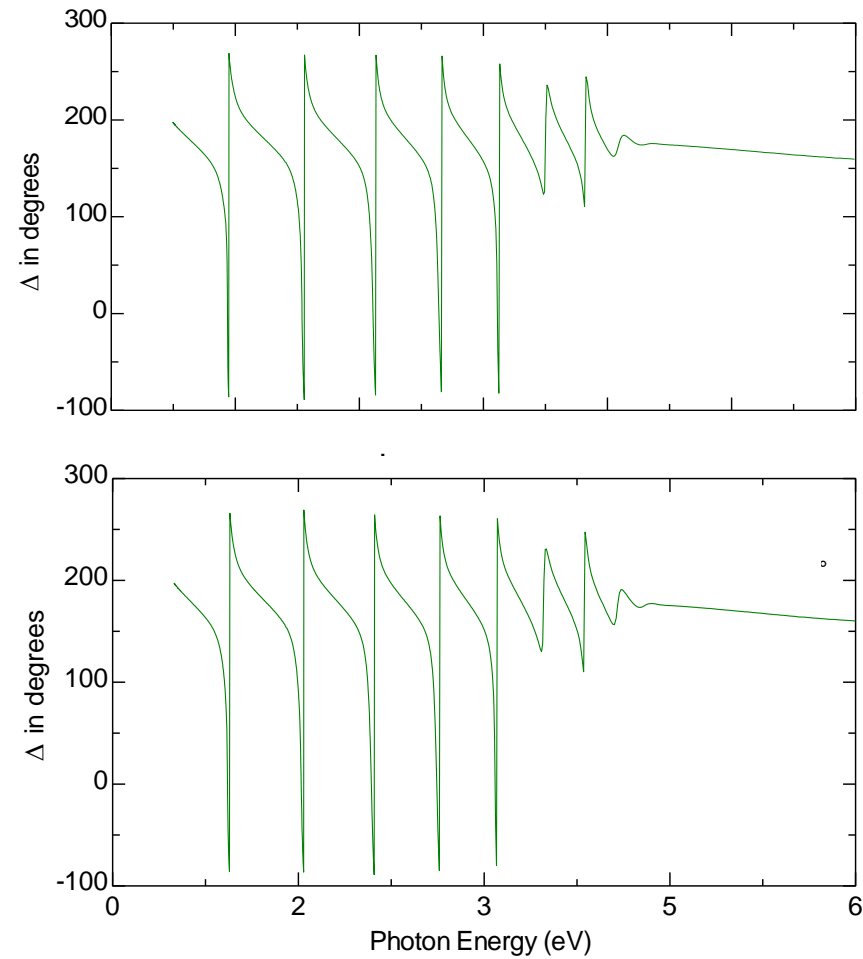
- The dielectric function of the substrate shows **relatively large variations with the temperature** (compatible with the literature, see for example Lautenschlager Phys.Rev. B 36 1987)
- **Very small variations between 75 K and 4 K** (not reported here)
- Dielectric function determined through a point-by-point fit - excellent agreement with the experimental data.

Titania-tantala coatings: SE data at room temperature

NOT
annealed



annealed

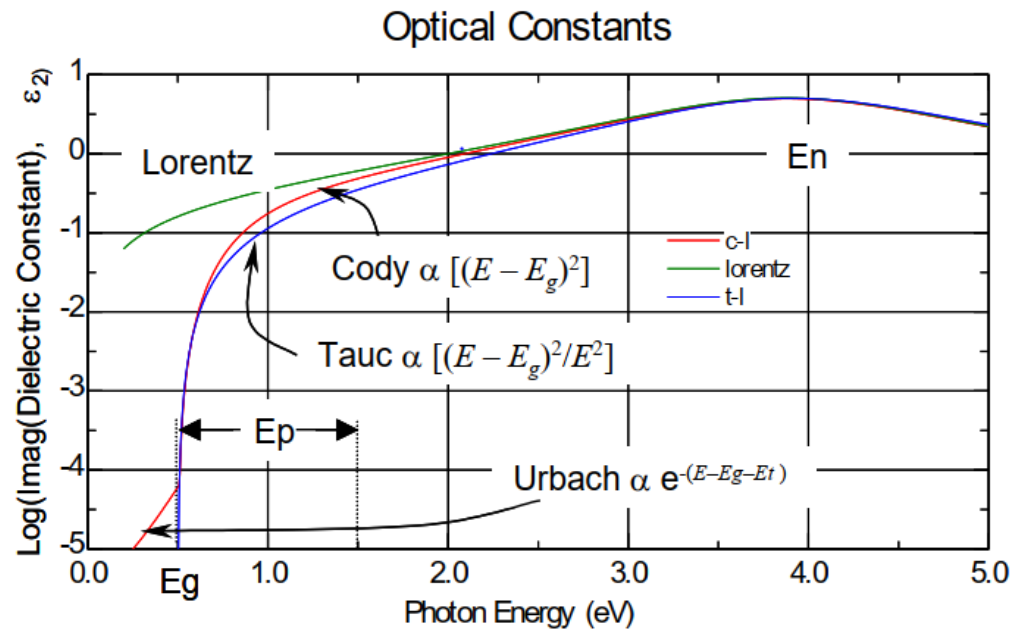


Modelling the optical properties of titania-tantala coatings

The optical properties of Titania-tantala coatings can be effectively modelled by using a **Cody-Lorentz model (CL)**

[A. Amato et al., Sci. Rep. 2020]

[A. Amato, M. Magnozzi et al., in preparation]



The CL model explicitly takes into account the **Urbach Tail**, related to the disorder in the coating.

The energy spread of the Urbach tail is indicated through the **Urbach Energy E_U** .

This can be described as a sum of three terms:

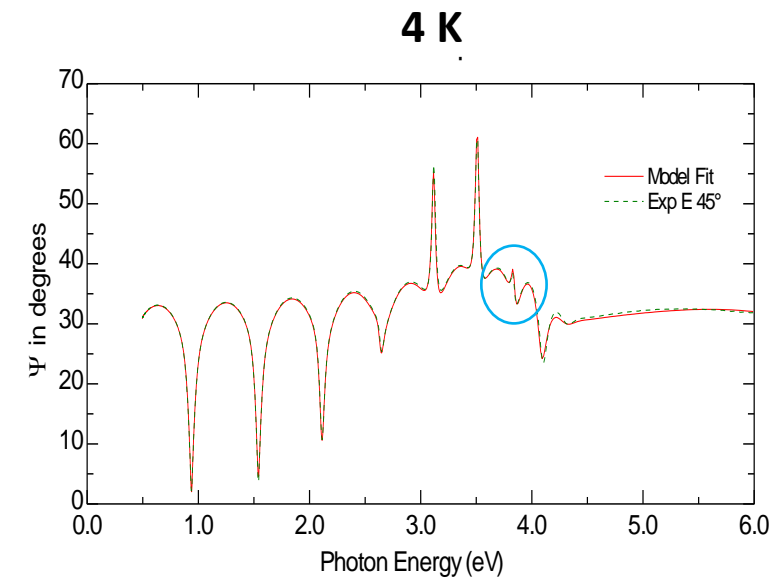
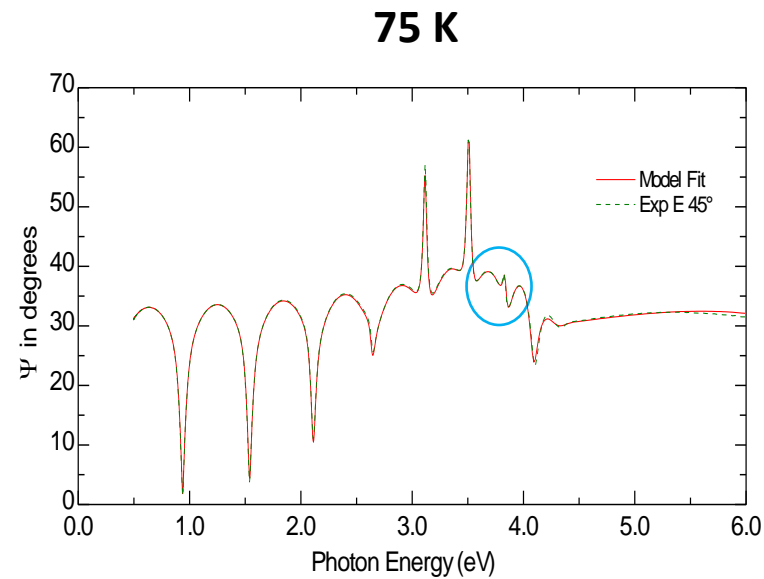
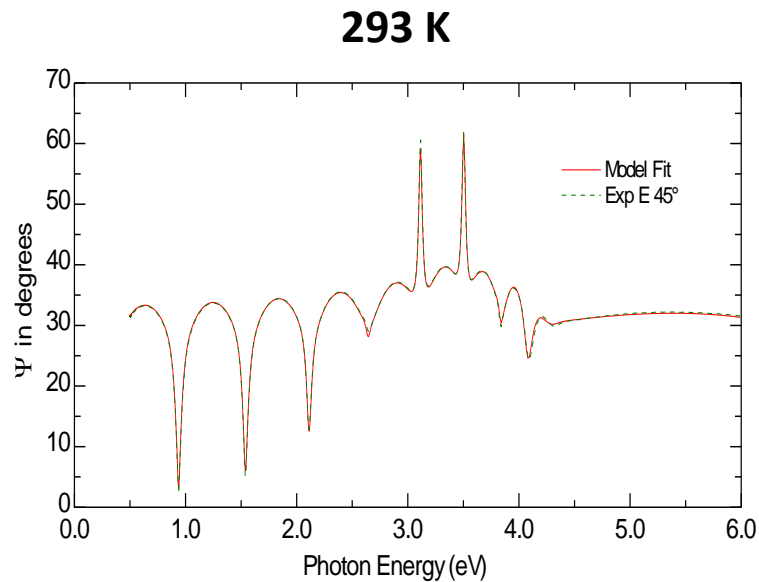
$$E_U = (E_U)_T + (E_U)_X + (E_U)_C$$

T=temperature-induced disorder

X=structural 'disorder'

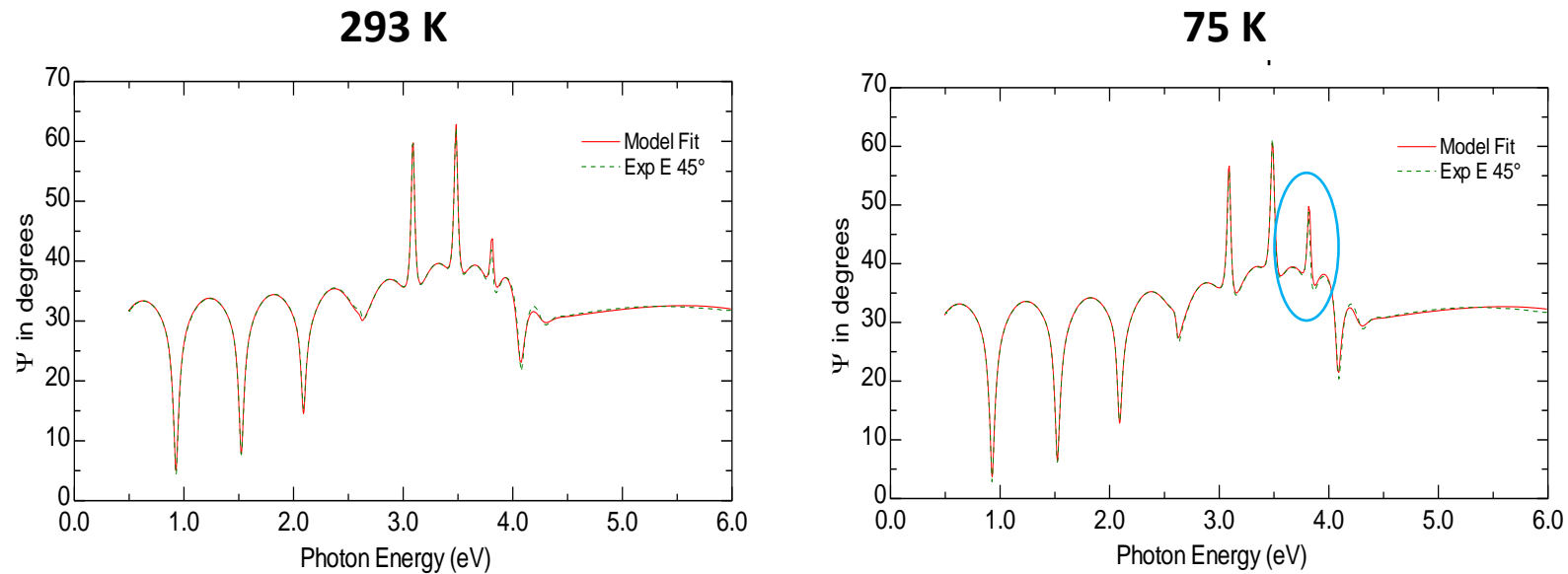
C=compositional 'disorder'

Model fitting at different temperatures: not annealed titania-tantala



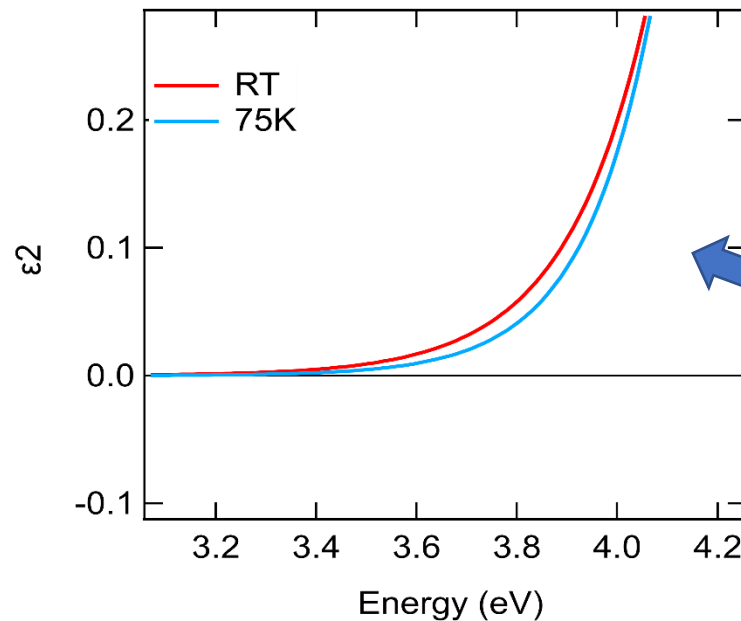
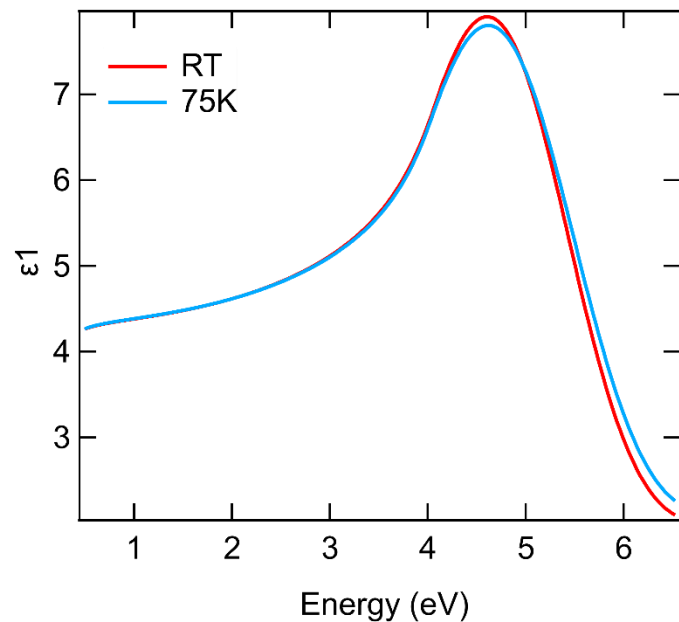
- The fit remains good at different temperatures.
- Sizable variations in the region of the onset of absorption: Urbach energy, energy gap

Model fitting at different temperatures: annealed titania-tantala

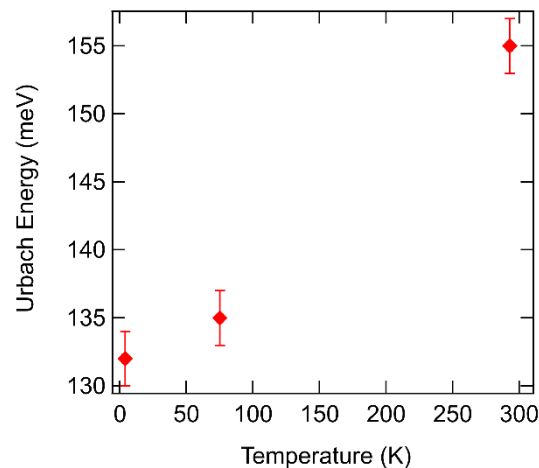


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Dielectric function comparison: not-annealed coating



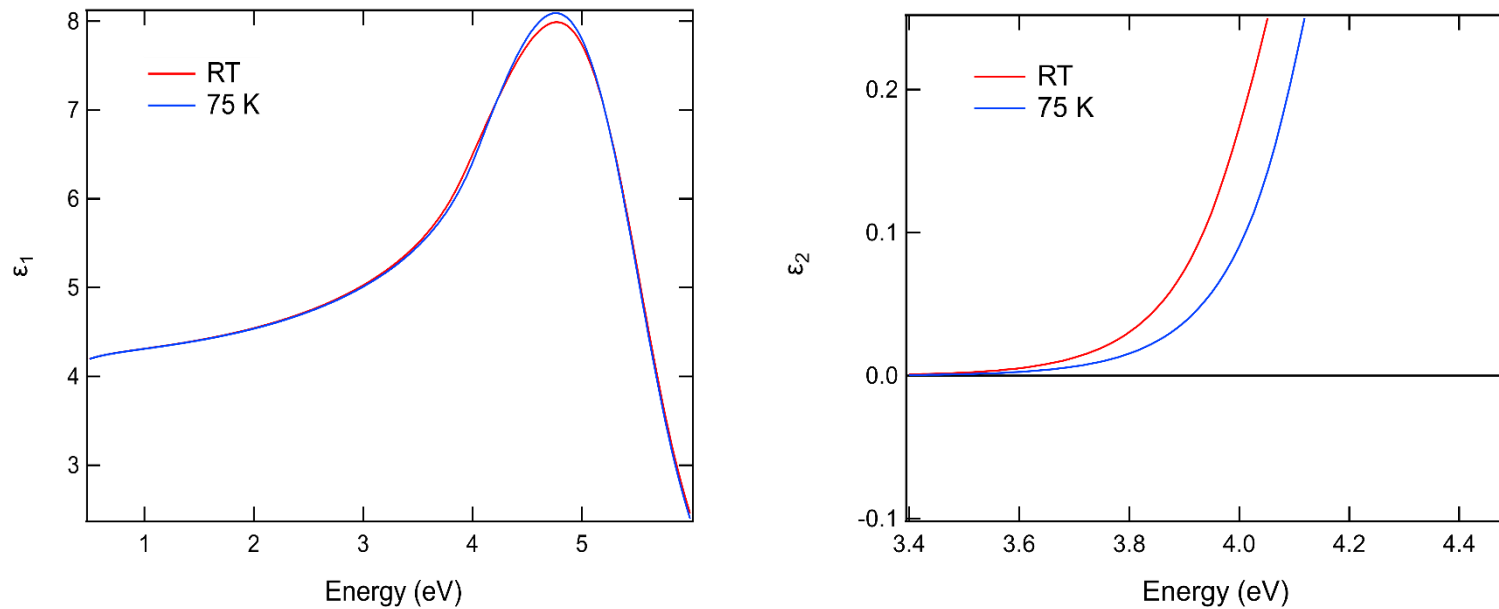
Reduction of the Urbach tail at low temperature



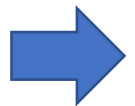
Temperature (K)	Urbach Energy (meV)
293	155
75	135
4	132

At 4 K, the Urbach energy of this sample is **reduced by ~15%**

Dielectric function comparison: annealed coating



No changes detected in the Urbach energy – T-induced variations are mainly described by a shift in the energy gap



The onset of absorption show different T-induced variations in annealed and not annealed coatings

Refractive index: silicon substrate

Silicon substrate	n @ 1064 nm	n @ 1550 nm	n @ 2000 nm
300 K	3.57(2)	3.48(8)	3.45(1)
75 K	3.52(4)	3.44(5)	3.40(7)
4 K	3.52(4)	3.44(6)	3.40(7)

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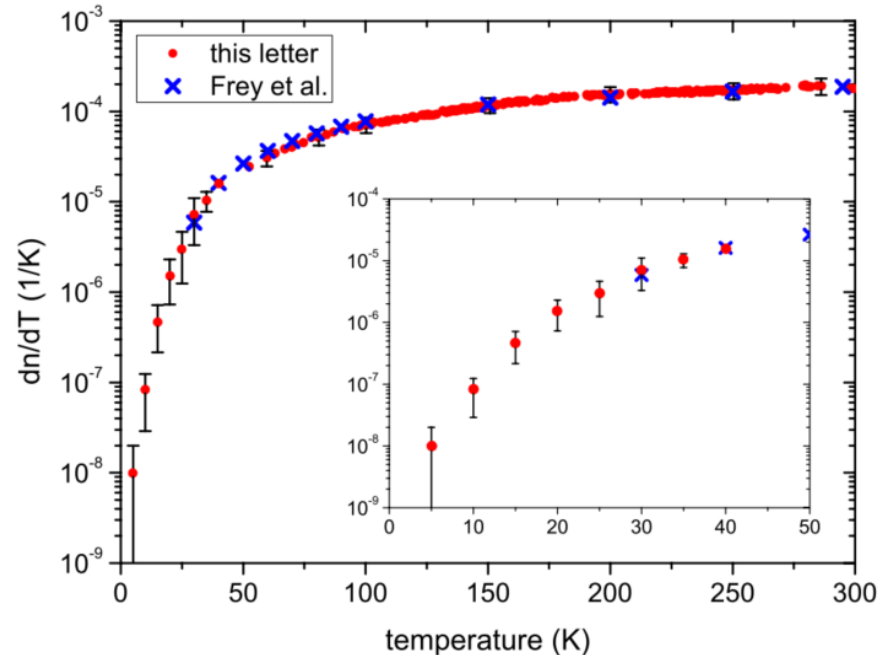
} **300 K to 75 K:**
n decreases by ~0.05

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300 K to 75 K:
n decreases by ~ 0.05

75 K to 4 K:
no variations detected
(thermo-optic coefficient of silicon
becomes really small at low T)



Komma et al., *Appl. Phys. Lett.* 101, 041905, 2012

Refractive index: titania-tantala

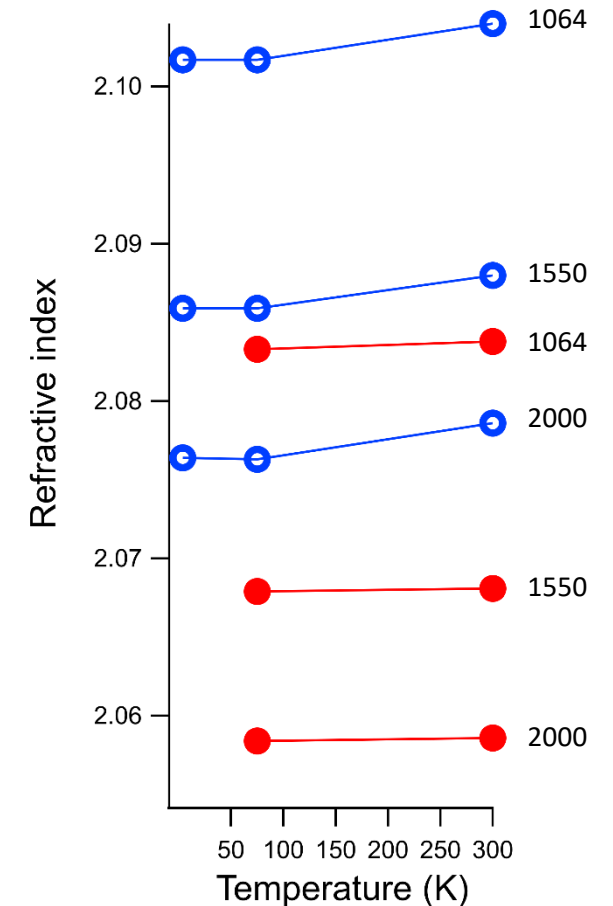
NOT annealed	n @ 1064 nm	n @ 1550 nm	n @ 2000 nm
300 K	2.10(40)	2.08(80)	2.07(86)
75 K	2.10(17)	2.08(59)	2.07(63)
4 K	2.10(17)	2.08(59)	2.07(64)

annealed	n @ 1064 nm	n @ 1550 nm	n @ 2000 nm
300 K	2.08(38)	2.06(81)	2.05(86)
75 K	2.08(33)	2.06(79)	2.05(84)

Refractive index: titania-tantala

NOT annealed	n @ 1064 nm	n @ 1550 nm	n @ 2000 nm
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The refractive index has **little** or **very little dependence on temperature** for not-annealed and annealed sample, respectively

Summary



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- Cryo setup for spectroscopic ellipsometry

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- Nanometric ice layer detected and modelled

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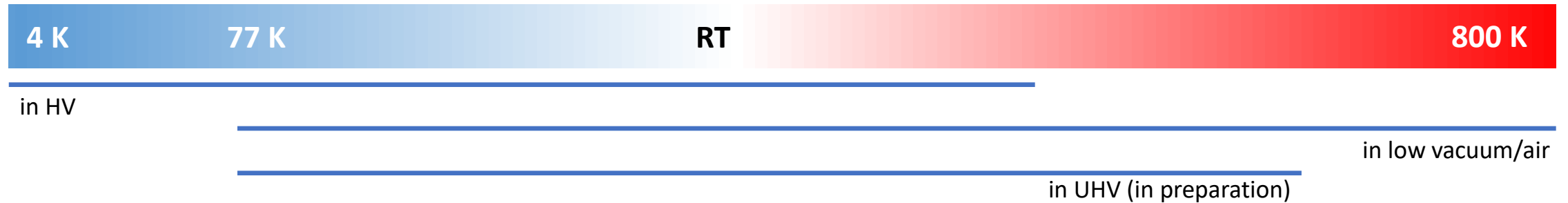


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- Nanometric ice layer detected and modelled
- Different behavior of annealed and not annealed coatings in the region of the onset of absorption
- T-induced variations in the refractive index: small in not-annealed coating, smaller in annealed ones
- Possibility to study other kinds of coatings in a variety of temperature and pressure conditions

Perspectives: T-dependent *in situ* optical characterization



Possibility to study **annealing and cooling in real time** with spectroscopic ellipsometry

In collaboration with F. Bisio (CNR-SPIN), Ermes Peci (UniGe)





Thanks

- Maurizio Canepa Università di Genova
- Gianluca Gemme INFN Genova
- Francesco Bisio CNR-SPIN
- Massimo Granata LMA
- Christophe Michel LMA
- Laurent Pinard LMA