X THE PLACE OF USEFUL LEARNING

Development of mixed oxide coatings for future detectors

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NMIS National Manufacturing Institute Scotland

Photonics Scotland

EPOC extreme performance in optical coatings

University of Strathclyde Glasgow

University of Glasgow

HELIA PHOTONICS

UNIVERSITY OF THE WEST of SCOTLAND UWS

UKRI Science and Technology Facilities Council

Gooch & Housego

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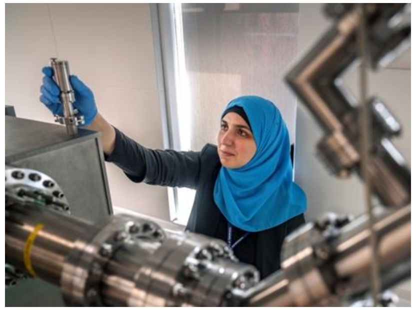

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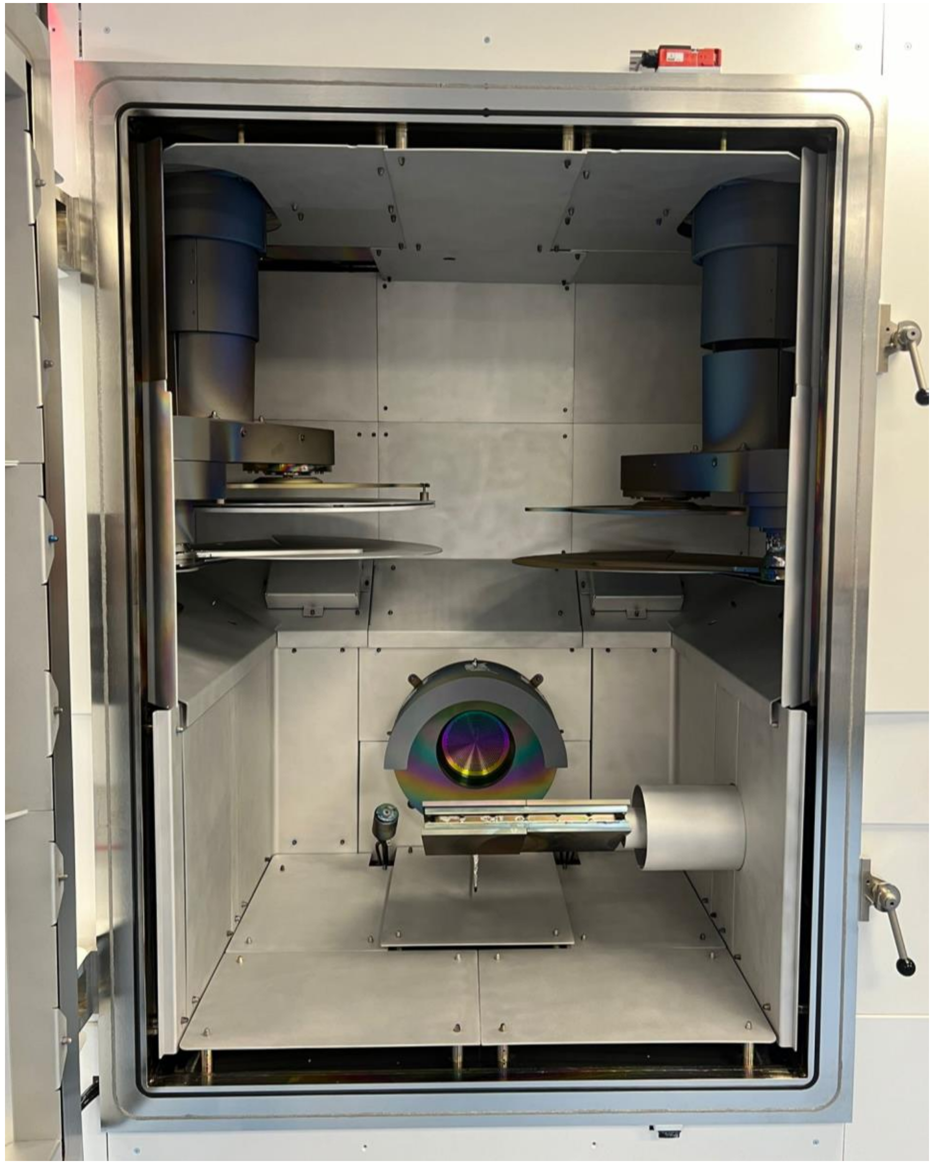
Extreme Performance in Optical Coatings (EPOC)

- <1% uniformity in 62 cm diameter without masks
- RF gridded ion sources with monolithic Ti grids: main source + assist source
- Process gases: Ar, N_2, O_2, H
- Background gases: O_2, N_2
- Process controlled by broadband optical monitoring
- PCI (1064, 1550, +), CRD (880,1064, 1550), spectrophotometry and microscopy (mapping), automated ultrasonic bath.



Brownian (thermal) noise: internal energy dissipation in the test masses

Using the fluctuation-dissipation theorem, assuming a Gaussian beam and negligible losses in the substrate we can approximate the thermal noise power spectral density as:

$$\boxed{S_x(f) \approx \frac{2k_B T}{\pi^2 f} \frac{d}{w^2 Y} \phi \left(\frac{Y'}{Y} + \frac{Y}{Y'} \right)}$$

d : coating thickness

w : beam radius (<< test mass radius)

ϕ : coating mechanical loss angle (= Q^{-1})

Y : substrate Young's modulus

Y' : coating Young's modulus

- Note that the thermal noise is temperature and thickness dependent.
- $S_x(f)$ can be measured in multilayers (*Gras, S., & Evans, M. (2018). "Direct measurement of coating thermal noise in optical resonators". Physical Review D, 98(12), 122001.*)

Mechanical loss angle is our key parameter

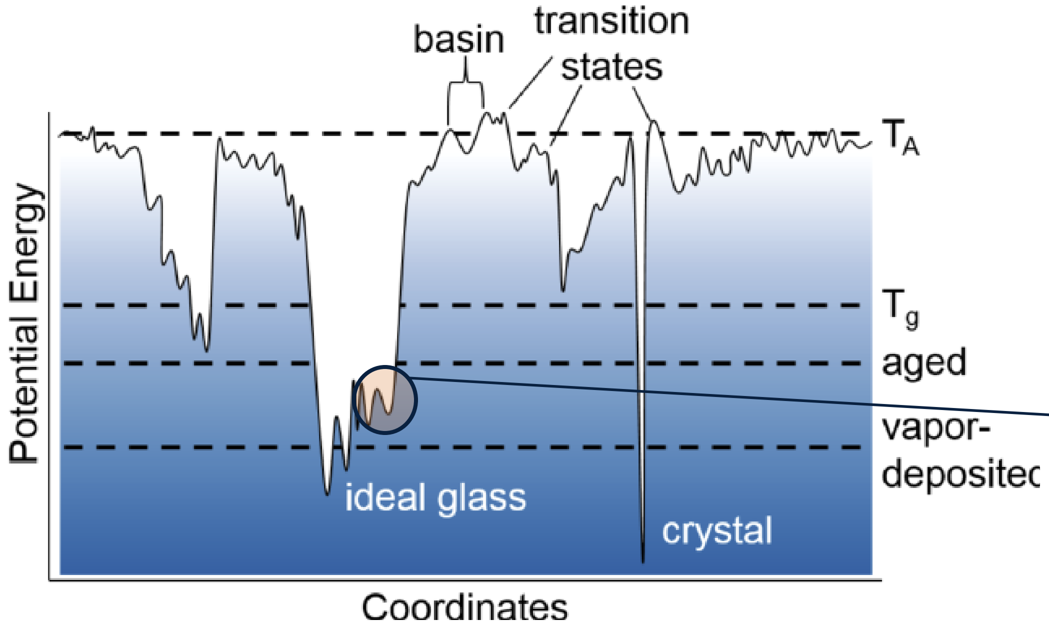
$$S_x(f) \approx \frac{2k_B T}{\pi^2 f} \frac{d}{w^2 Y} \boxed{\phi} \left(\frac{Y'}{Y} + \frac{Y}{Y'} \right)$$

ϕ : coating mechanical loss angle (= Q^{-1})

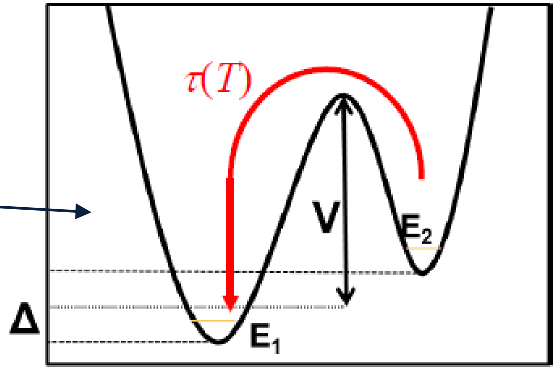
- Characteristic of the material (single layer or multilayer).
- Frequency and temperature dependent.
- Can be measured by *ringdown* experiments (+ elastic theory considerations). *Vajente, G., et. al. Review of Scientific Instruments 88.7 (2017): 073901.*
- Quotient between real and imaginary part of the material's Young's modulus.
- Measure of the energy loss in the optical cavity.
- <u>We can determine the mechanical loss angles for the high and low index materials and then estimate the thermal noise of the mirror (not perfect but good approximation).</u> *Hong, T., et. al. Phys. Rev. D 87, 082001 (2013).*

Mechanical loss relates to low energy excitations

We conceptualize these excitations as **two-level systems** and therefore changes in the atomic structure lead to elastic dissipation.



$$Q^{-1} = N \frac{\gamma^2}{Y k_B T} \frac{\omega\tau}{1 + \omega^2\tau^2} sech^2\left(\frac{\Delta}{k_B T}\right)$$



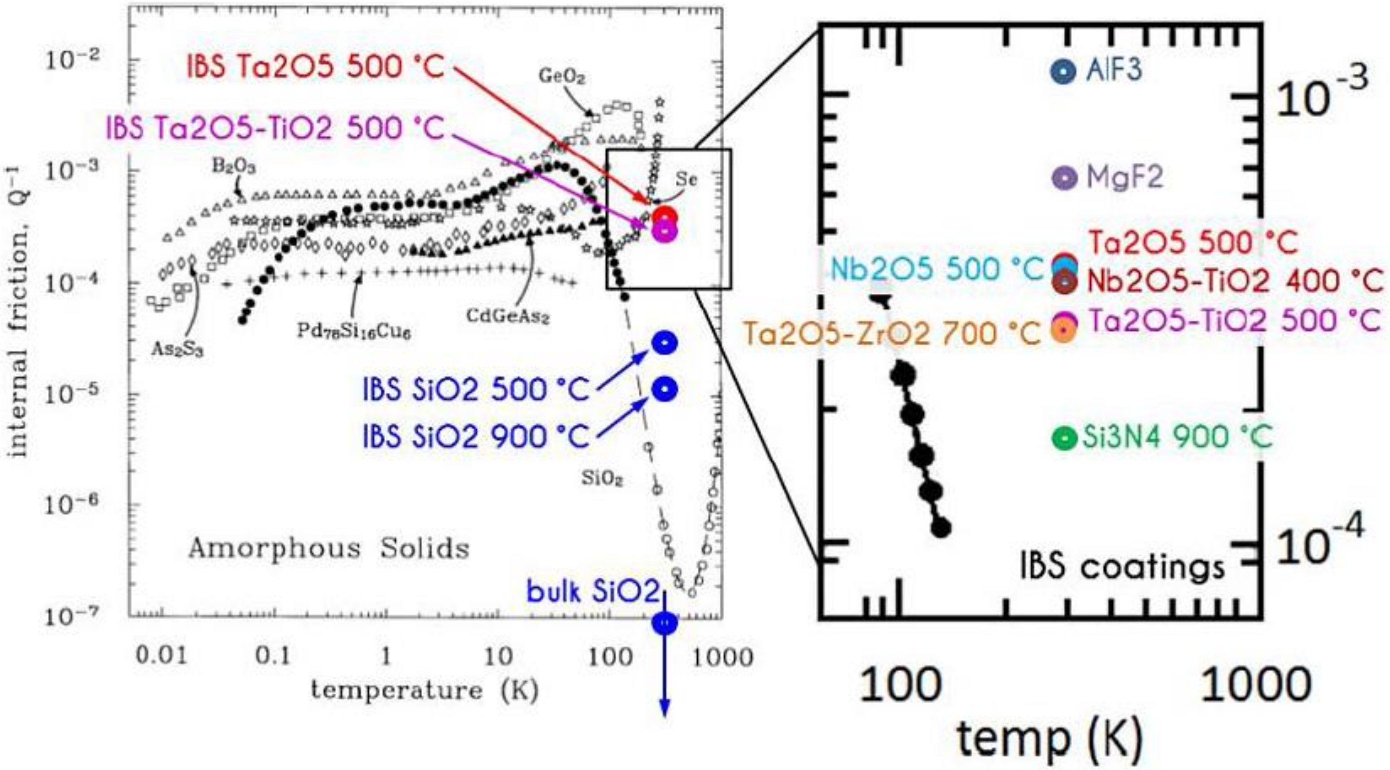
$$\tau = \tau_0 \, sech\left(\frac{\Delta}{2k_B T}\right) e^{V/k_B T}$$

- Materials have a distribution of TLSs.
- Low temperatures ⟶ quantum tunneling.
- T > 5 K ⟶ thermally activated transitions.

Ediger, Mark D. "Perspective: Highly stable vapor-deposited glasses." The Journal of chemical physics 147.21 (2017): 210901.

What we know about mechanical loss in binary oxides and doped Ta_2O_5

High index materials have the highest loss angles



Granata, M., et al. "Progress in the measurement and reduction of thermal noise in optical coatings for gravitational-wave detectors." OIC conference, OSA, 2019.

- Bulk silica has a much lower loss angle than silica coatings.
- High index materials have loss angles at least an order of magnitude higher than silica.

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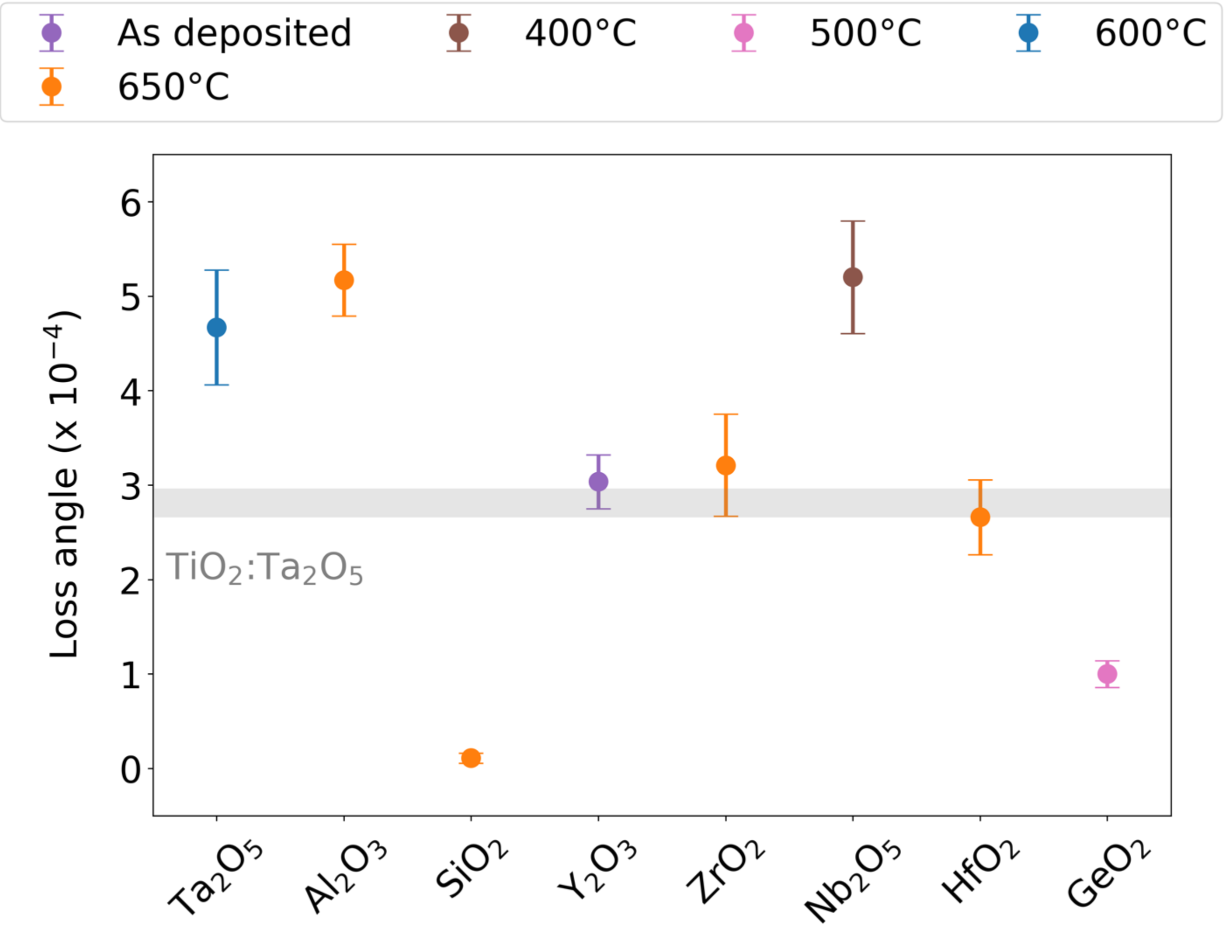
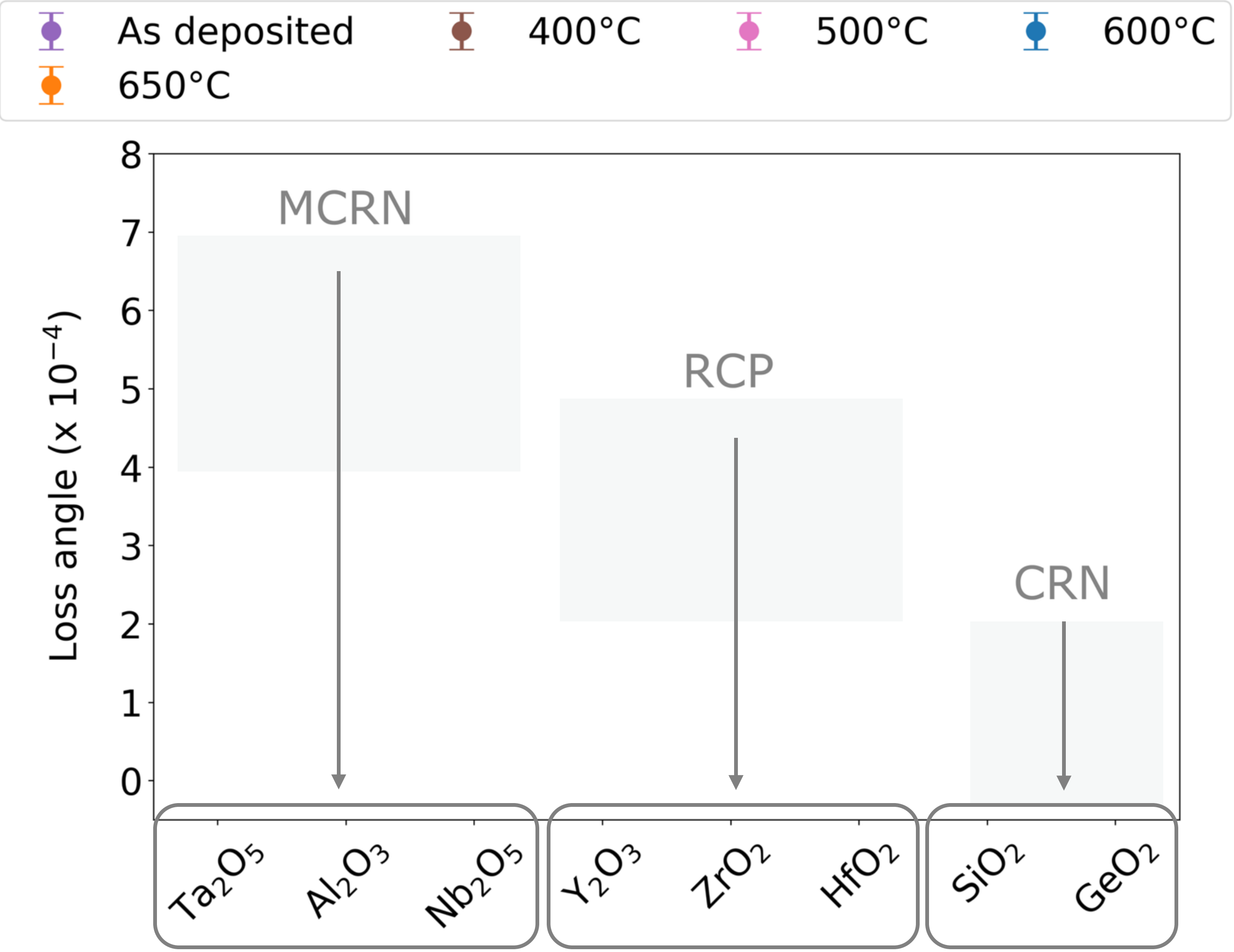
Mechanical loss of binary oxide thin films



- Lowest mechanical loss corresponds to SiO_2 y GeO_2 (but these are low index materials).
- HfO_2 is not a good candidate due to its Young's modulus.
- No obvious correlation between crystallization temperature and mechanical loss.

Fazio, Mariana A., et al. "*Comprehensive study of amorphous metal oxide and Ta2O5-based mixed oxide coatings for gravitational-wave detectors.*" PRD 105 102008 (2022).

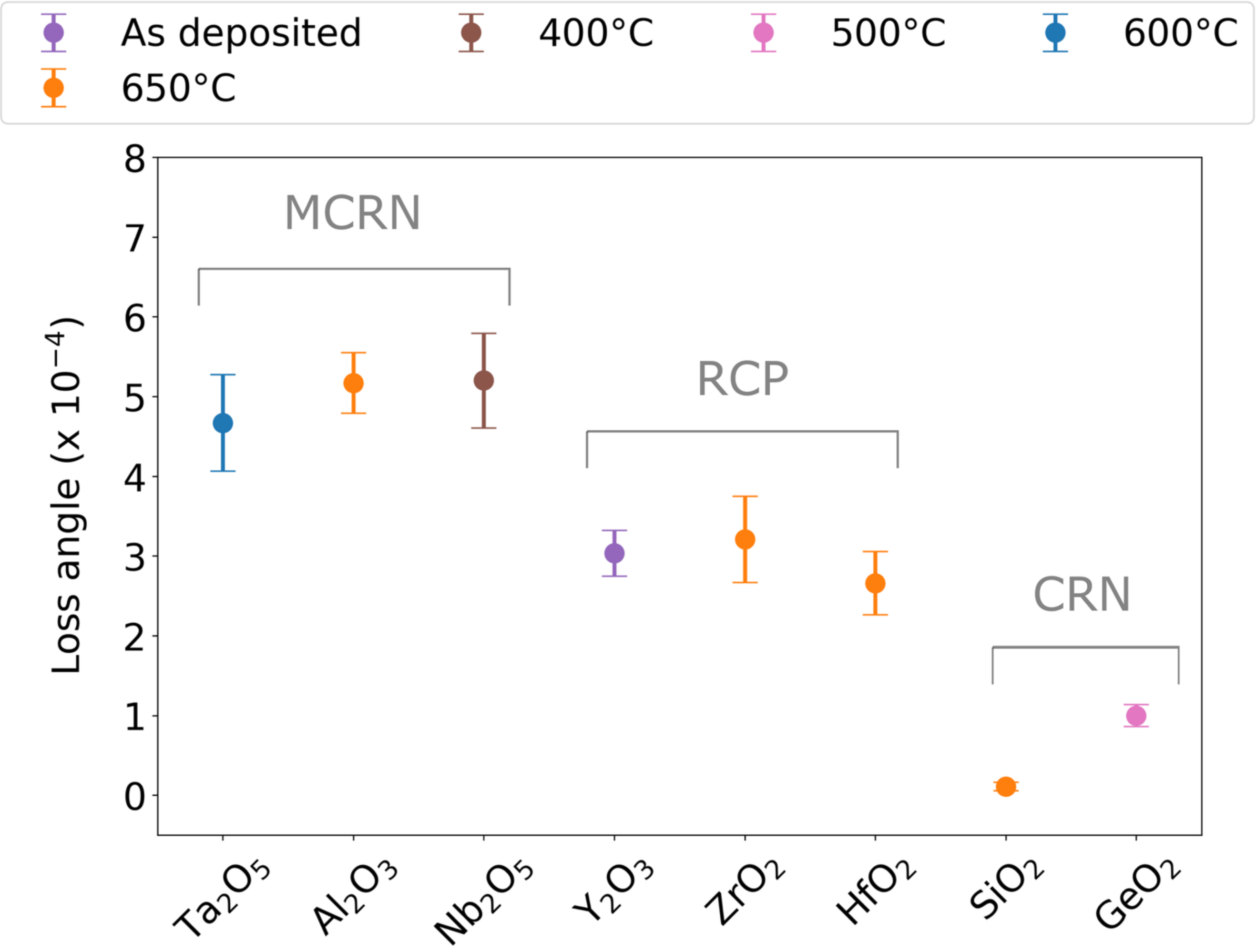
Mechanical loss of binary oxide thin films: amorphous classification



Amorphous solids can be classified as (according to Zallen):

- **Continuous Random Network (CRN)**, have mostly covalent bonds
- **Modified Continuous Random Network (MCRN)**, covalent bonds are modified and disrupted by ionic bonds
- **Random Closed Packed**, consists of a non-periodic closed packing of ions, organic polymers fall in this category

Mechanical loss of binary oxide thin films: amorphous classification



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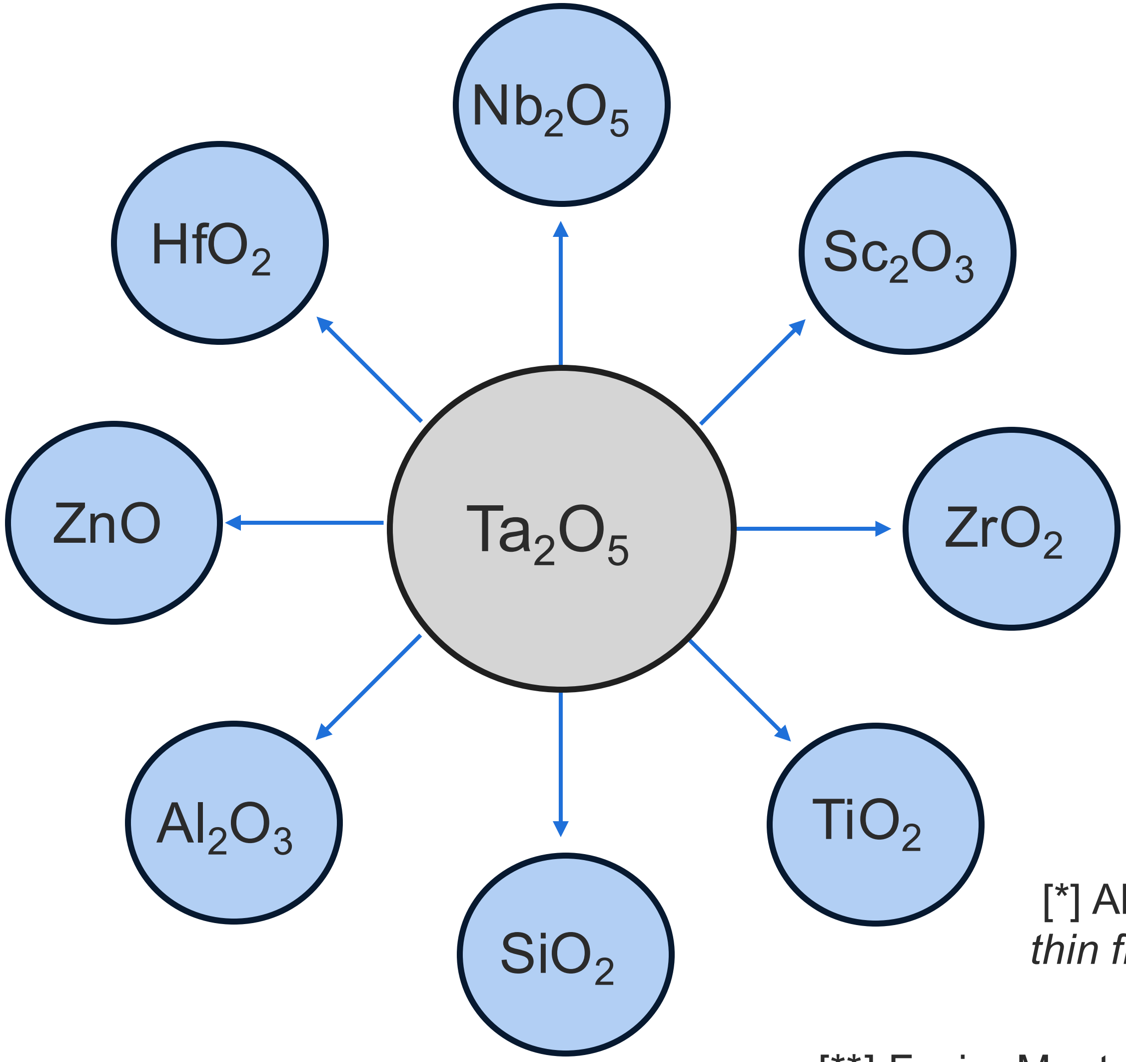
The mechanical loss of binary oxides is correlated to their amorphous morphology.

Exploring different dopants for Ta_2O_5



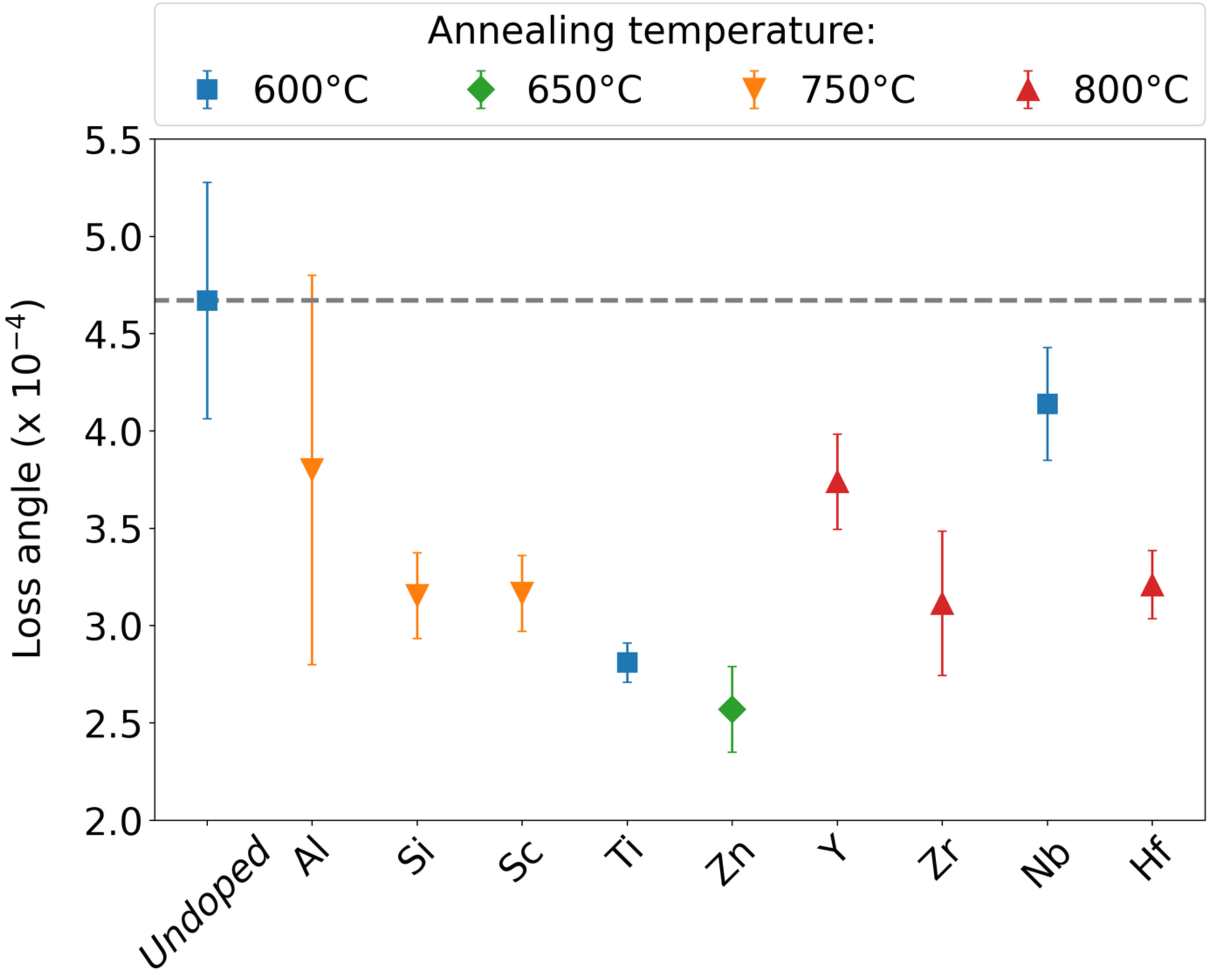
Dopant concentration was kept ~ 20%

Material	Cation ratio	n @ 1064 nm
Ta_2O_5	-	2.12 ± 0.01
TiO_2:Ta_2O_5	0.27 ± 0.04	2.19 ± 0.01
Nb_2O_5:Ta_2O_5	0.12 ± 0.01	2.11 ± 0.01
ZrO_2:Ta_2O_5 [*]	0.23 ± 0.01	2.07 ± 0.01
HfO_2:Ta_2O_5	0.23 ± 0.02	2.05 ± 0.01
Al_2O_3:Ta_2O_5	0.17 ± 0.01	2.01 ± 0.01
SiO_2:Ta_2O_5	0.26 ± 0.01	1.93 ± 0.01
ZnO:Ta_2O_5	0.20 ± 0.01	2.05 ± 0.01
Sc_2O_3:Ta_2O_5 [**]	0.105 ± 0.007	2.08 ± 0.01

[*] Abernathy, M, et. al. "*Exploration of co-sputtered Ta2O5-ZrO2 thin films for gravitational-wave detectors*" Classical and Quantum Gravity, 38(19):195021, 2021.

[**] Fazio, M, et. al., "*Growth and characterization of Sc_2O_3 doped Ta_2O_5 thin films*", Applied Optics, 59(5) : A106–A111, 2020.

Mechanical loss for Ta_2O_5-based mixed oxide coatings



Dopant addition generally reduces mechanical loss.

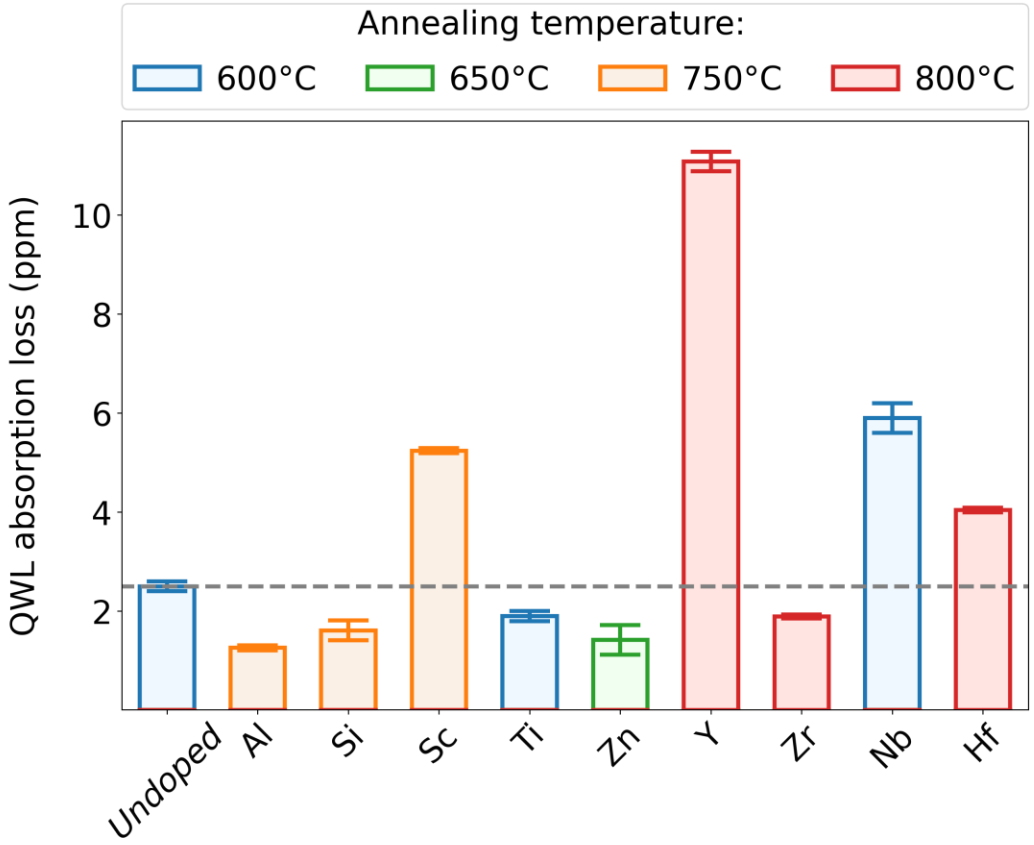
$ZnO:Ta_2O_5$ was found to be partially crystallized so not a good candidate (due to increased scattering loss).

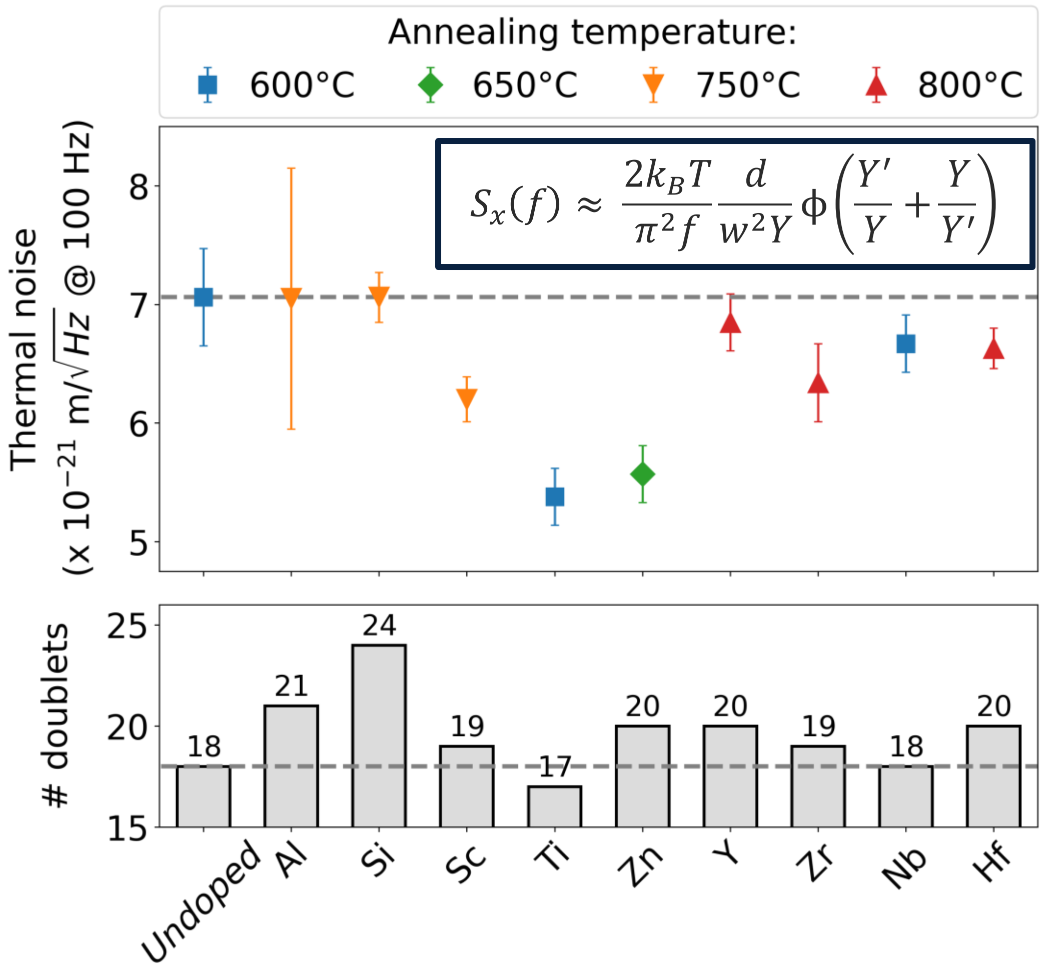
Formation of ternary compounds could be related to reduced mechanical loss.

TiO_2 is still the best dopant (within this limited study).

Fazio, Mariana A., et al. "*Comprehensive study of amorphous metal oxide and Ta2O5-based mixed oxide coatings for gravitational-wave detectors.*", PRD 105 102008 (2022).

Absorption loss and thermal noise estimation for Ta_2O_5-based mixed oxide coatings





<u>Bonus</u>: Fazio, M, et. al., “Prediction of crystallized phases of amorphous Ta2O5-basedmixed oxide thin films using a density functional theory database”, APL Materials, 9(3):031106, 2021.

<u>HR design:</u> QWL of doped Ta_2O_5 + QWL SiO_2 (transmission ~ 5 ppm)

Lessons learned from TiO_2:GeO_2

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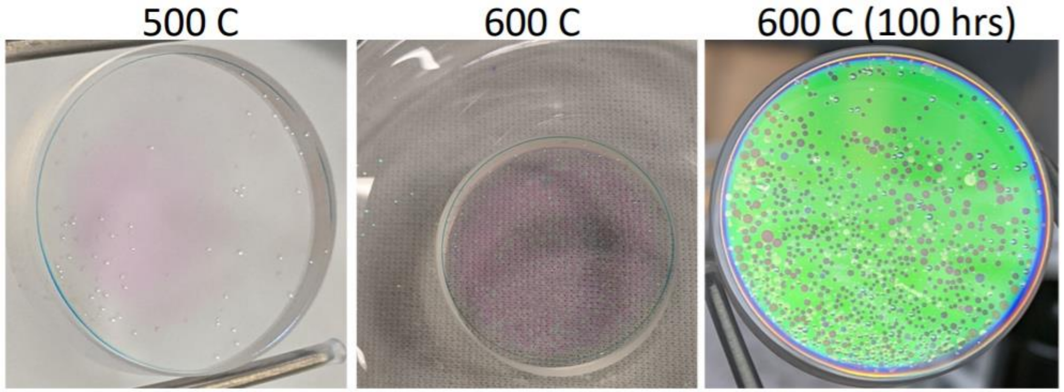
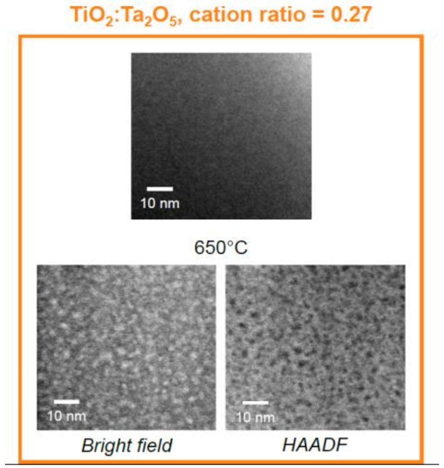
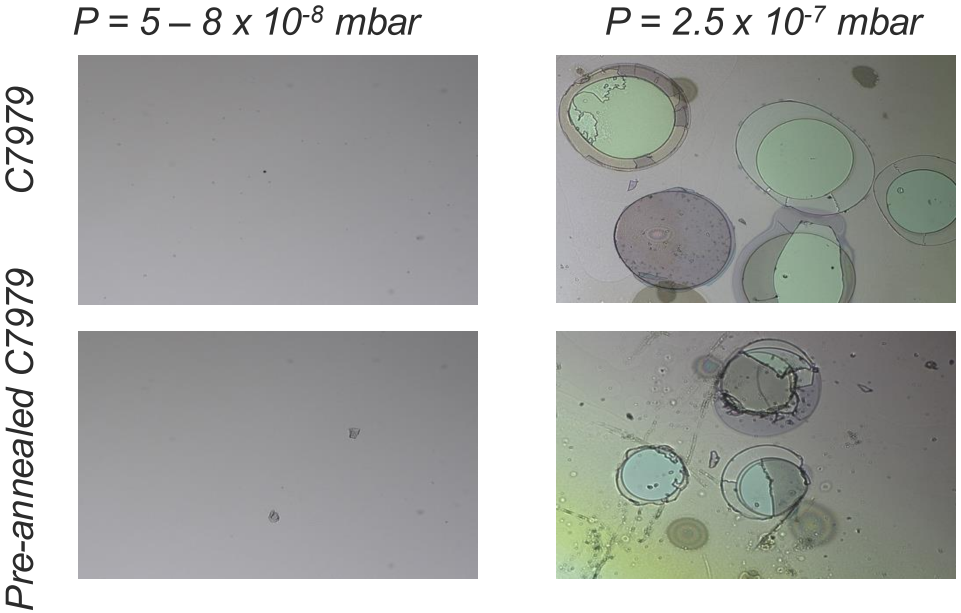
The problem of bubbles – annealing induced defects





- Seem to depend on chamber base pressure (= water partial pressure)
- EPOC suppressed these defects by lowering base pressure to 6 – 8 x 10^{-8} mbar
- Others have solved it by decreasing Ar flow, lowering base pressure and/or growing at high temperatures (150 C)



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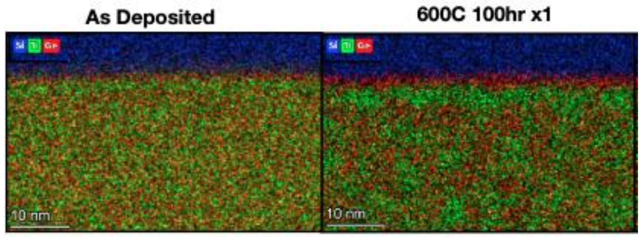
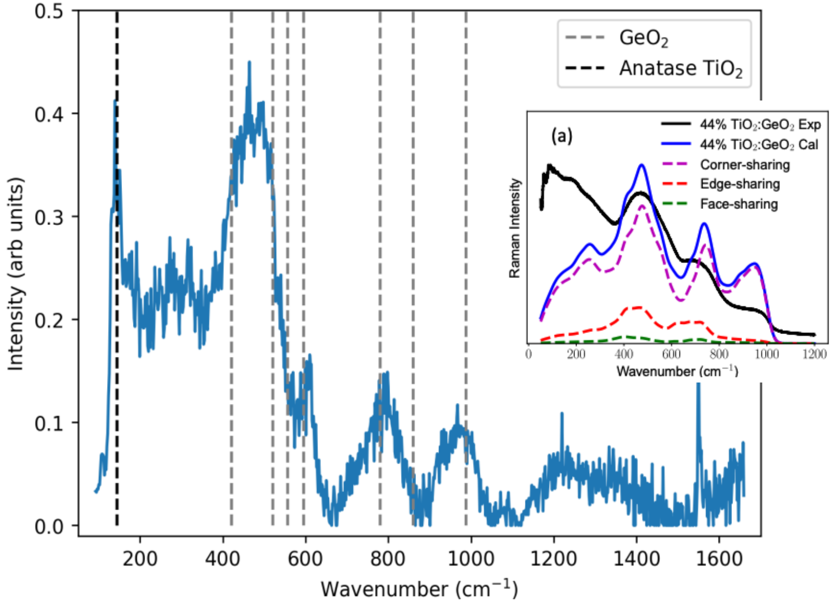
Structure evolution with annealing - Raman

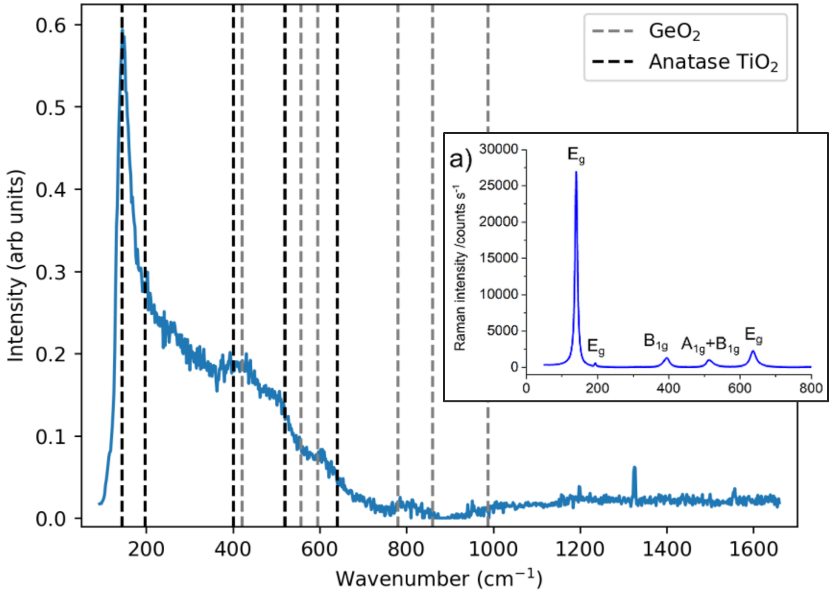
- Huge effort from the TiO2:GeO2 working group to study and optimize these coatings
- We saw that annealing induces clustering
- This was verified by EELS (*Kyung-Ha Lee, et. al., LIGO-G2501921*)
- Crystallization of the titania clusters doesn’t necessarily affect the scattering loss



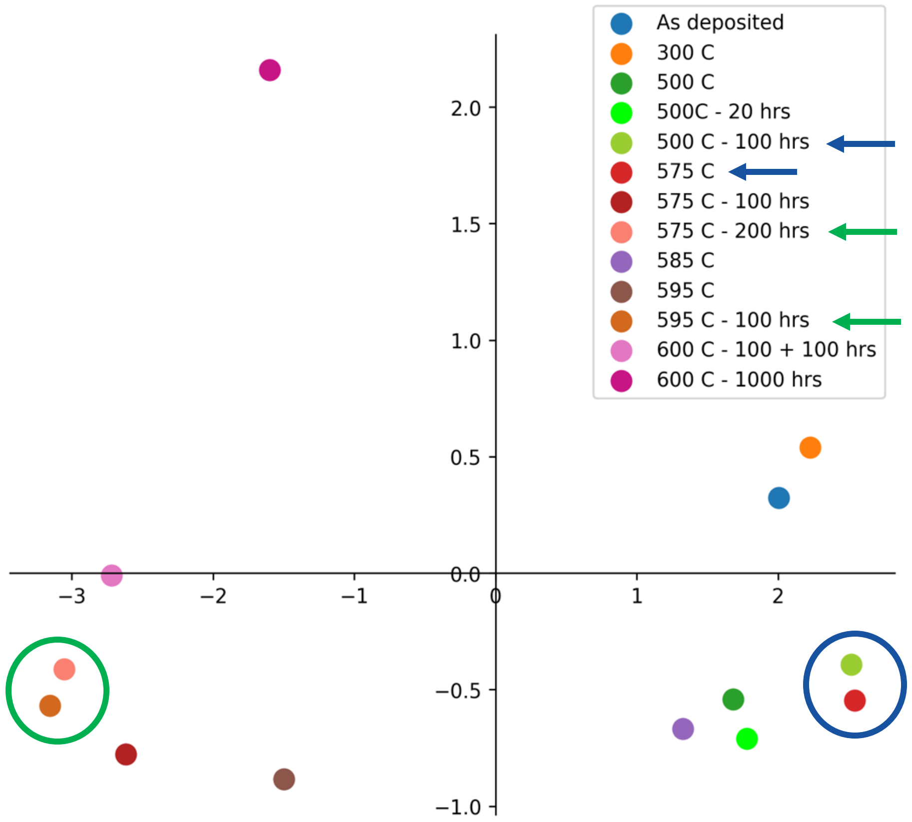
As deposited



Annealed at 595 C for 10 hrs



Structure evolution with annealing - Raman



- Visualization of Raman with PCA.
- Structure of 500 C – 100 hrs similar to that of sample annealed at 575 C for 10 hrs
- Structure of 575 C – 100 hrs close to that of samples annealed at 595 C / 600 C for 100 hrs
- Structure of 595 C – 100 hrs and 575 C – 200 hrs are very similar

- We see similar structure when annealing at lower temperatures for longer soaking time
- This can allow to design annealing protocols that induce the desired structure and avoid crystallization
- Raman combined with ML methods has provided significant insights into the structure

Conclusions

- ✓ For binary oxides, mechanical loss is related to their amorphous morphology.
- ✓ In doped oxides, the resulting structure (especially after annealing) can have unexpected features.
- ✓ The issue of bubbles points towards more research on the IBS process, to determine origin of defects.
- ✓ TiO_2:GeO_2 has been a humbling lesson in expecting the unexpected.
- ✓ ML techniques have been employed to improve characterization techniques with great success.
- ✓ We still have much to learn (and discover) from oxide coatings.

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Thank you for your attention

Work funded by ST/V005634/1, ST/V005642/1, and ST/V005626/1, MF funded on ST/W004844/1 and SR funded on INF\R1\201072.

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