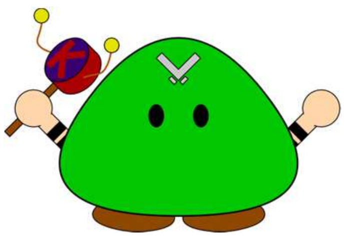
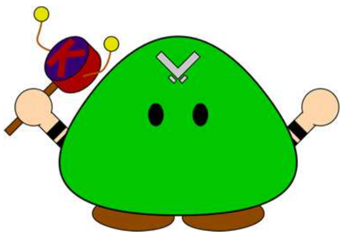


GWADW 2014



The INFN AdCOAT Project

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**University of Sannio, INFN, LVC and KAGRA*

Università degli Studi del Sannio

INFN Istituto Nazionale di Fisica Nucleare

LSC LIGO Scientific Collaboration

VIRGO

KAGRA

Takayama, JP, 25-30 May, 2014

Outline

Historical

Coating Materials for Future Detectors

nm-Layered Composites

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Historical

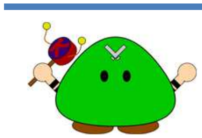
- INFN is the Italian National Institute for Nuclear Physics (established 1951), and the leading funding Agency for fundamental and applied research in nuclear and astroparticle Physics in Italy. Together with French CNRS it is the main funding agency of Virgo. It is organized in Panels (*Commissioni*). Panel V is in charge of *Technological Research.*

  - The study of coating thickness optimization was funded by INFN Panel V. (COAT research program);
  - The study of nm-layered glassy oxide mixtures was also funded by INFN Panel V (MIDI-BRUT research program);
  - Another research proposal, (COACH led by prof Flavio Vetrano) was run in paralel (2007-2010), and was focused on the development and chara-cterization of low-loss materials for optical coatings;

  AdCOAT is the natural follow up of the former, and an attempt to merge several LVC groups working in Italy on the various aspects of coating R&D

The Coating Holy Grail

- Early coating material downselection led first to the choice of Silica (SiO_2) end Tantala (Ta_2O_5) as preferred coating materials for GW detectors [Crooks et al., CQG 23 (2006) 4593]
- It became soon clear that the high-index ingredient (Tantala) was mainly responsible of the coating noise [Penn et al., CQG 20 (2003) 2917].
- Attempts to reduce coating noise led to the development of Titania doped Tantala (SiO_2::Ta_2O_5) [Harry et al., 24, 40 (2006)], and thickness optimized coatings **[Villar et al., PRD 81 (2010) 122001]**.
- Further attempts to find "better" glassy oxide mixture failed sofar, with the possible exception of CSIRO's Silica doped Titania [Netterfeld and Gross, OSA/OIC 07, ThD2] and Tantala [P. Murray, PhD thesis, U. Glasgow, 2008].

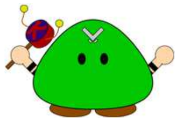


The Coating Holy Grail, contd.

- Mechanical losses in amorphous materials are associated with therm-ally activated local transitions between the minima of asymmetric bi-stable potentials, and can be computed from knowledge of (the distri-butions of) their relevant parameters (barrier height and unbalance) [Gillroy and Phillips, Phil. Mag.B43, 745 (1981)].

- Modeling efforts to deduce these latter from first principles are on-going, with some promising results **[Trinastic et al., LIGO-P130002825]** , [Bassiri et al., LIGO Document G-1400271]. Present knowledge, how-ever is still insufficient to design glassy oxide mixtures with prescribed properties, and the quest for the coating Holy-Grail coating materials still relies on extensive trial-and-error [Flaminio et al., CQC 27 (2010) 084030].

- An EMT based Mickey-Mouse model of glassy mixtures accounts reason-ably well for TNI mesured material loss angles [Villar et al., LIGO- G1100 976], but is largely oversimplified.



The Coating Holy Grail, contd.

- EMT is on the other hand a decently rigorous model for sub-wavelength layered materials, and allows to design composites with prescribed optical (complex refraction index) and viscoelastic (complex Young's modulus) [Pinto et al., LIGO-G1100586 (2011)].

- Subwavelength layered composites based on Titania::Silica and Hafnia:: Silica show promise, and might be suitable for cryo operation (KAGRA, ET).

- The AdCOAT research effort is accordingly focused on the design and characterization of nm-layered composites based on (Silica-Hafnia-Titania), and their comparison with co-sputtered mixtures using the same ingredients.

- It is also aimed at "networking" the experimental facilities and expertises of several Groups in Italy working on different aspects of optical coatings for GW detectors.

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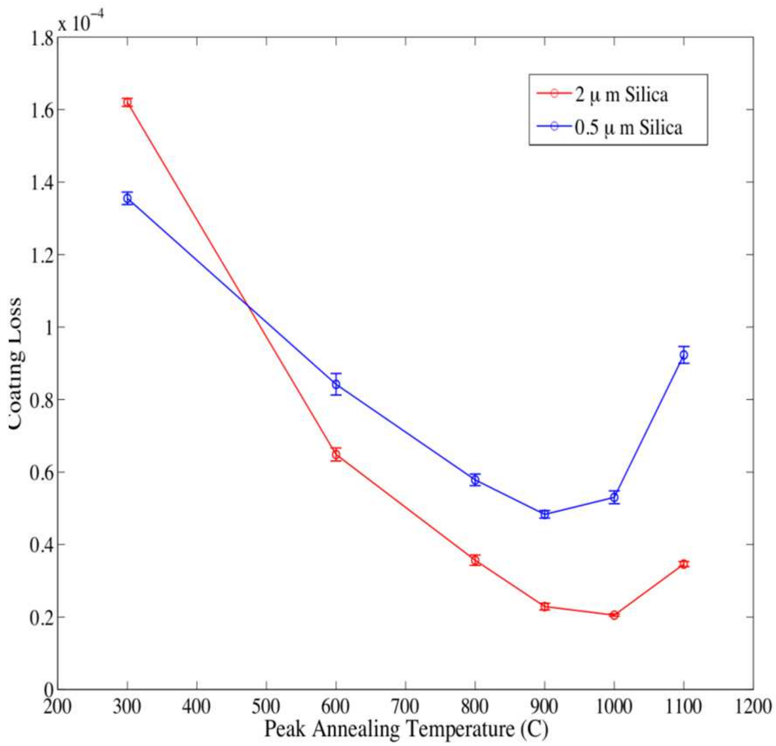
Annealing

- Thermal anneal (TA) is needed to reduce
  optical absorption
  internal stress
  mechanical loss (→ thermal noise).

- The mechanical loss angle vs annealing T curve has a lesser slope for *thinner* films.

- *Most* coating materials (exc. SiO_2, Al_2O_3) tend to form crystals upon annealing. This spoils their quality, causing
  optical scattering loss
  mechanical loss (→ thermal noise).

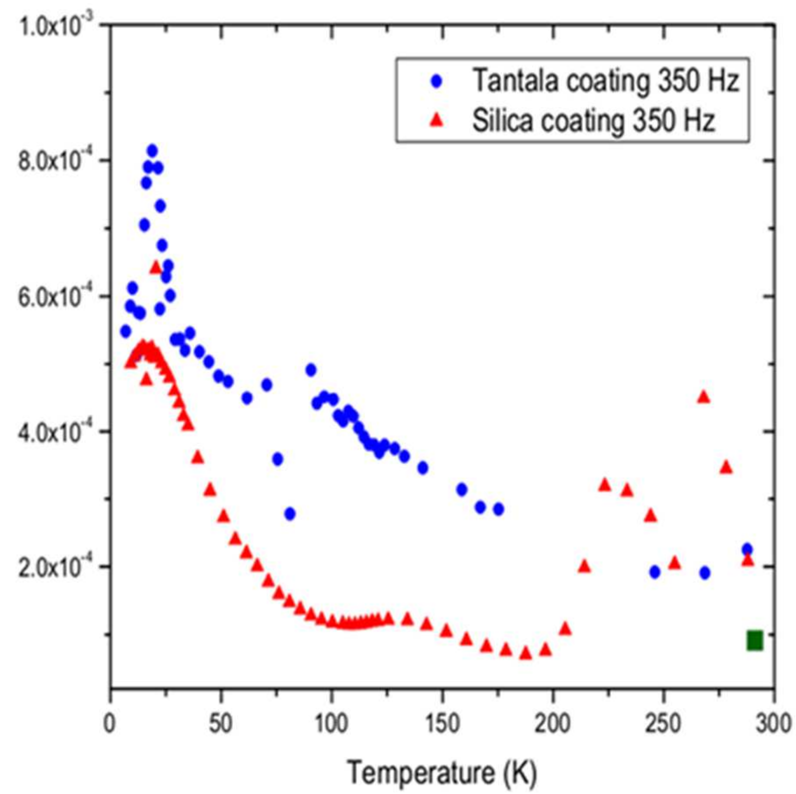


[S. Penn et al., LIGO-G1000932]

Cryopeak

- *Most* coating materials show increasing mechanical losses as the temperature is decreased, up to a peak at a few tens of K.
- For SiO_2 and $(TiO_2::)Ta_2O_5$ the cryopeak occurs around 20K, i.e., where KAGRA and ET will presumably operate.
- The position and level of the cryopeaks are dependent on the details of the heat treat-ment.

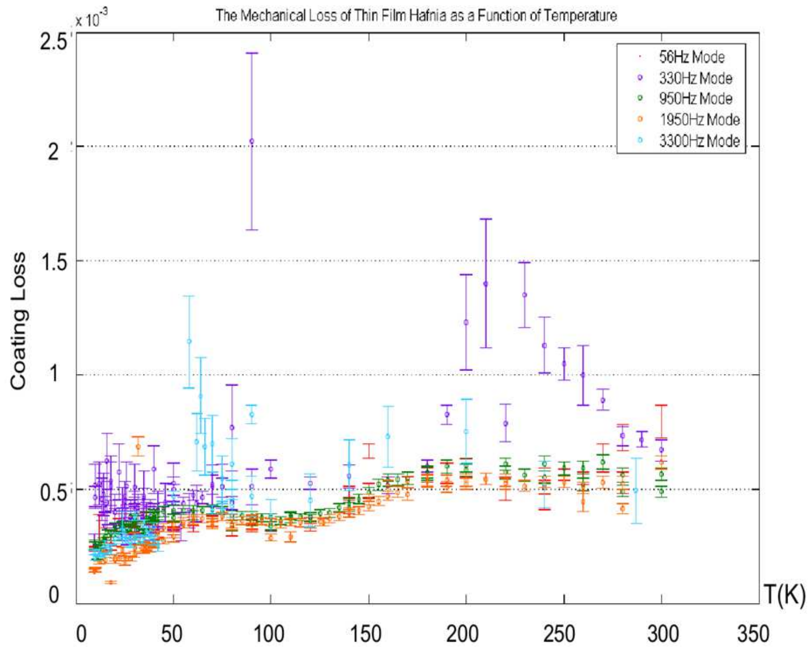
[Martin et al., CQG 27 (2010) 225020]

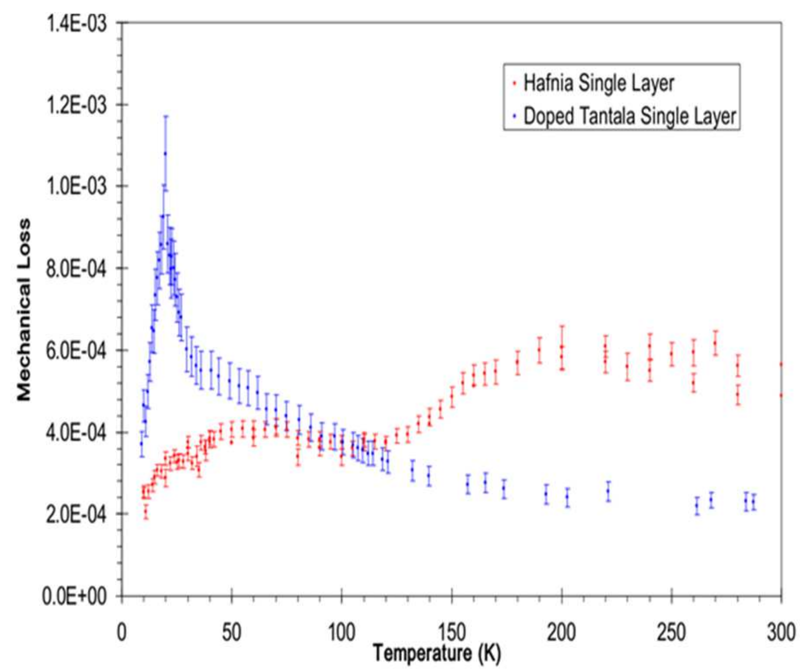


[Martin et al., CQG 25 (2008) 055005]

Cryopeak Free Candidates: Hafnia

"Preliminary results of the mechanical loss of (a-)HfO_2 do not show a large peak in dissipation at $T \sim 20K$ in contrast to Ta_2O_5"

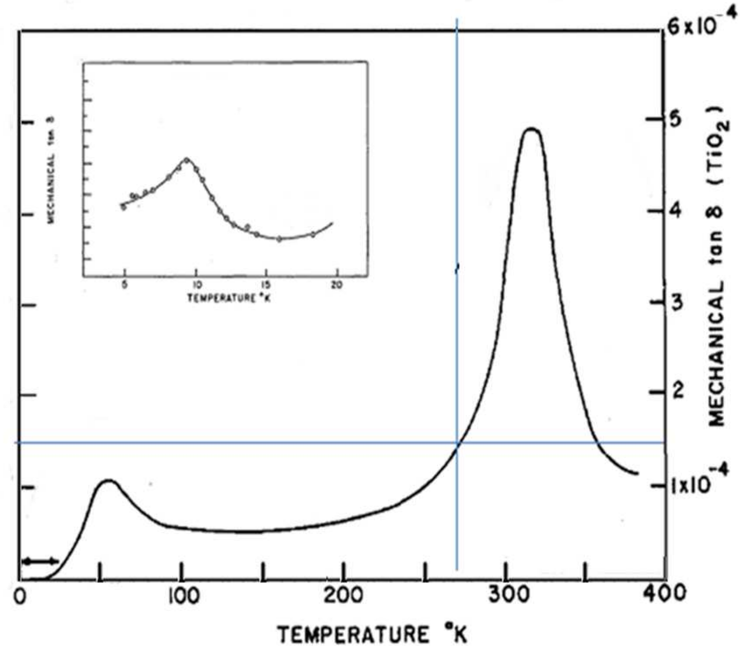




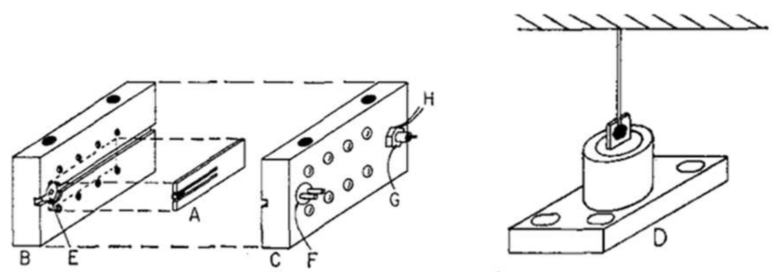
[Chalkley et al., LIGO-GG080314]

Cryopeak Free Candidates: Titania

Results in [Scott and MacCrone, Rev. Sci. Instr. 39 (1968) 821] obtained from a cantilever - like based ring-down measurement setup suggest that a –TiO_2 may also feature a nicely weak cryogenic loss peak.



A—sample; B, C—steel holders; D—steel supporting yoke; E—driver coil; F—capacitive pickup; G—germanium thermometer; H—copper-constantan thermocouple.

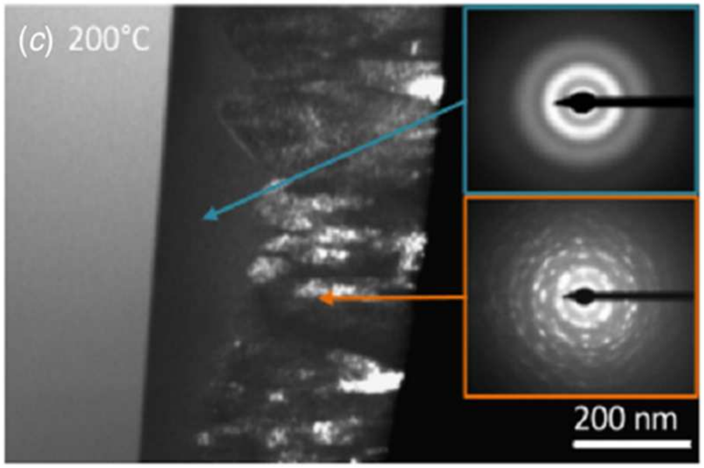


No recent loss angle measurement is available for amorphous TiO_2 mechanical losses in the cryogenic regime .

Fresh measurements needed

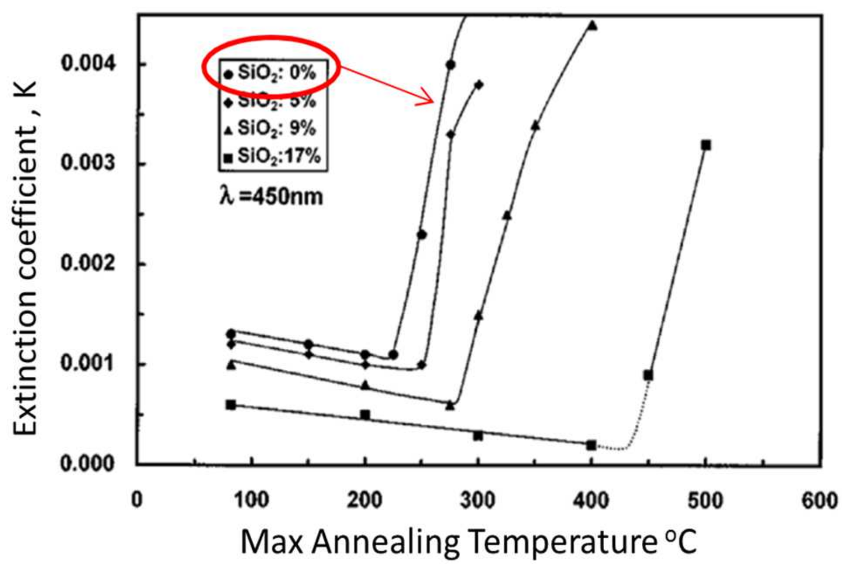
Silica Doping

- **Both Hafnia and Titania are prone to crystalizzation upon annealing.**
- **Silica doping contrasts crystallization** **[S. Pond, Appl. Optics, 28 (1989) 2800]**



[Abernathy, CQG 28(2011) 195017]

SiO_2 *doping stabilizes* HfO_2 (and ZrO_2) against *crystallization* [Ushakov et al., Phys. Stat. Sol. B241 (2004) 2268].



SiO_2 *doping stabilizes* TiO_2 against *crystal-lization* [Wang and Chao, Opt. Lett. 23 (1998) 1417; Chao, JOSA A16 (1999) 1477; Chao et al., Appl. Opt. 40 (2002) 2177]

Silica Doping, contd.

ThD2.pdf © 2007 OSA/OIC 2007

Investigation of Ion-Beam-Sputtered Silica-Titania Mixtures for Use in Gravitational Wave Interferometer Optics

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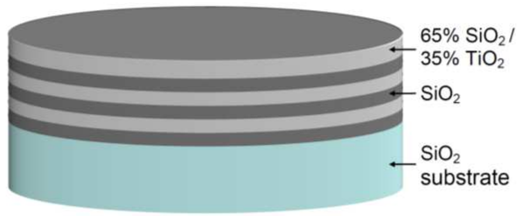
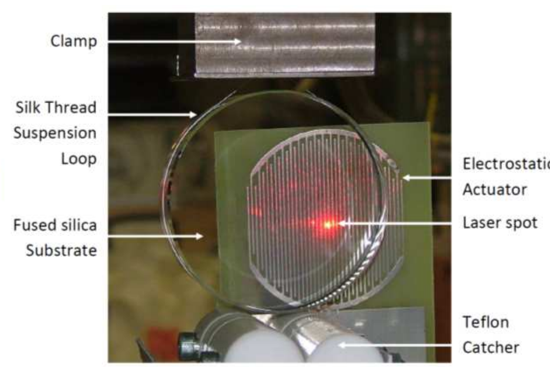
Abstract: Ion-beam-sputtered mixtures of silica and titania are investigated as potential coating materials for use in gravitational wave interferometer optics. Such coatings must have both low optical and mechanical loss to maximize detection sensitivity.

OCIS codes: (230.4170) Multilayers; (160.4670) Optical Materials; (240.0310) Thin Films.

Loss angle measurements made by P. Murray in his PhD thesis.



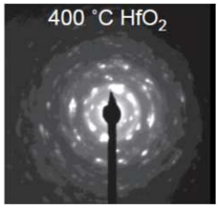
$(\lambda/4, \lambda/4)^{15}$	$10^4 \times$ (residual loss angle)	n_H
Ta_2O_5 / SiO_2	4.4 ± 0.2	2.02
TiO_2:: Ta_2O_5 / SiO_2 (15::85)	2.4 ± 0.2	2.07
SiO_2:: Ta_2O_5 / SiO_2 (35::65)	2.5 ± 0.4	1.83
TiO_2:: Si_2O_2 / SiO_2 (35::65)	1.7 ± 0.4	1.77

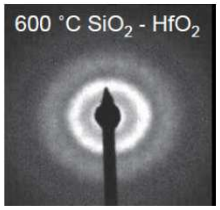


Silica Doped Hafnia – No Cryopeak

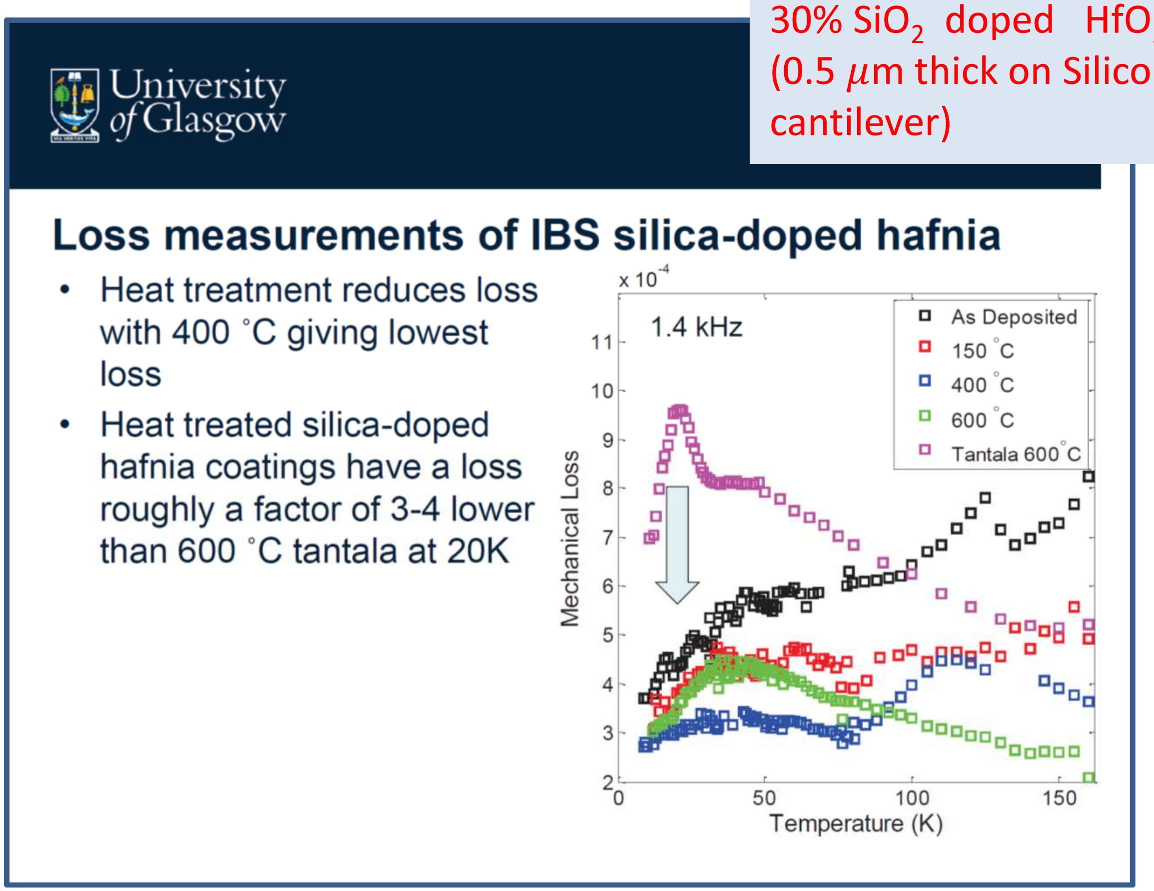
Silica doping, besides halting crystallization upon annealing, does not spoil nice (no cryo-peak) behaviour of undoped Hafnia.

Mechanical losses 4 times lower @ 20 K compared to Titania doped Tantala





30% SiO_2 doped HfO_2 (0.5 μm thick on Silicon cantilever)



[P. Murray, LIGO G-1400275]

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Thickness

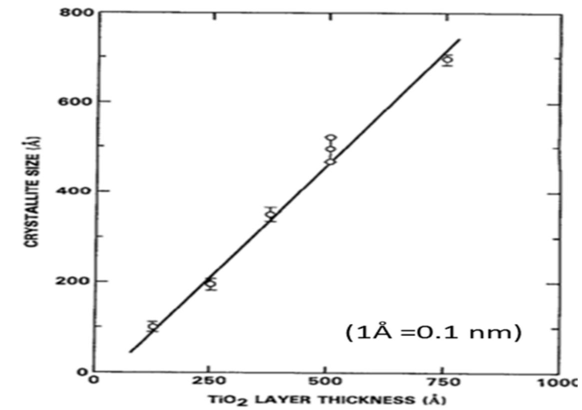
Seminal work on *co-sputtered* TiO_2/SiO_2 films by Sankur & Gunning [J. Appl. Phys. 66 (1989) 4747]

"Thinner layers (< 250 Å) required higher temperatures [to crystallize]. 65 Å layer films exhibit diffraction only after annealing at 600°C."

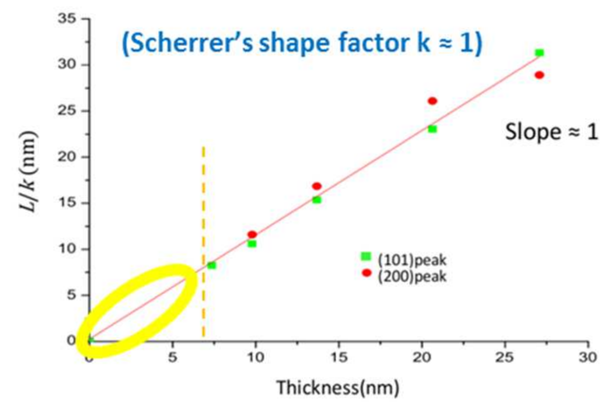
"Grain size, as deduced from diffraction line broadening, was comparable to the layer thickness"

"Thicker layers remain in the Anatase phase and never transform into Rutile, even for prolonged (72 h) annealing at the highest temperature s (1100°C). Thinner layers (65 Å) convert into Rutile starting at 900°C"

"Below a certain critical thickness crystallization in pure TiO_2 films is inhibited"



[Gluck et al., J. Appl. Phys. 69 (1991) 3037]



[S. Chao et al., LIGO-G1300921]

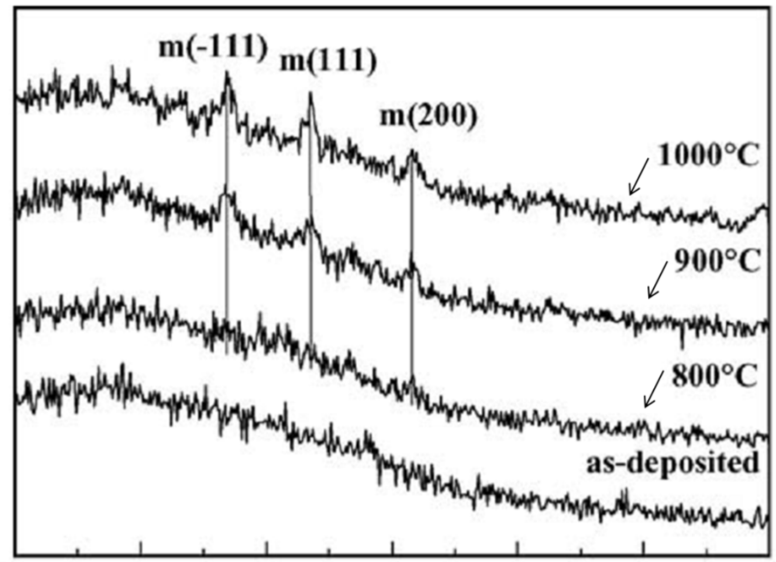
Thickness, contd.

***Nanometer-layered* Hafnia (12nm)/Alumina (3nm) composites *do not* crystallize upon annealing, up to temperatures of 800 ^oC**

[M. Liu et al., Appl. Surf. Sci. 252 (2006) 6206].

"XRD analysis shows that the films remain amorphous up to an anneal-ing temperature of 800 ^oC"

"FTIR indicates that no interface layer forms during annealing up to 800 ^oC"



Thickness, contd.

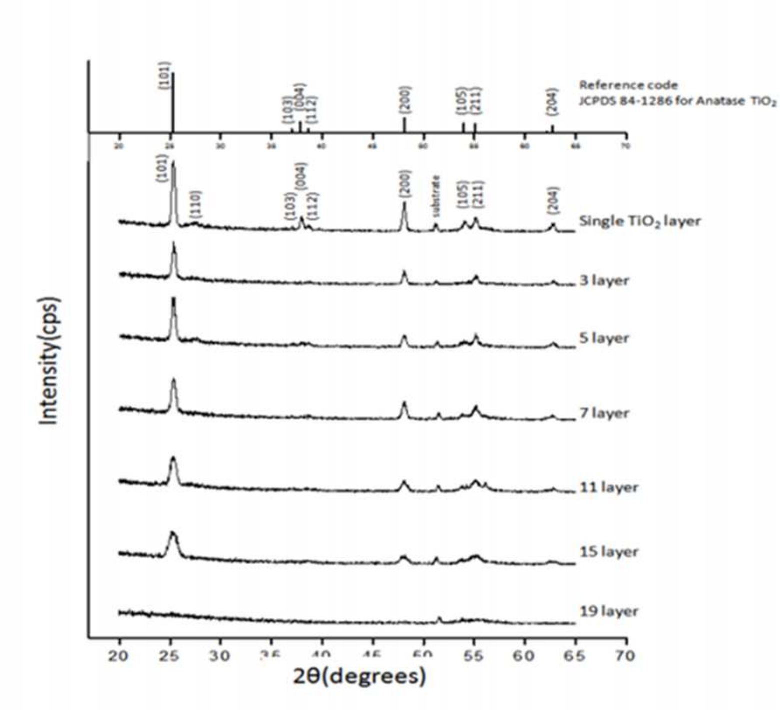
nm-layered **Titania/Silica composites** ***do not*** **crystallize upon annealing**

[S. Chao et al., LIGO-G1300921].

“quarter-wavelength thick nm-layered Titania / Silica composites differing in the number and thickness of the nm-layers show increasing frustration of grain crystal formation as the layers’ thickness is decreased”

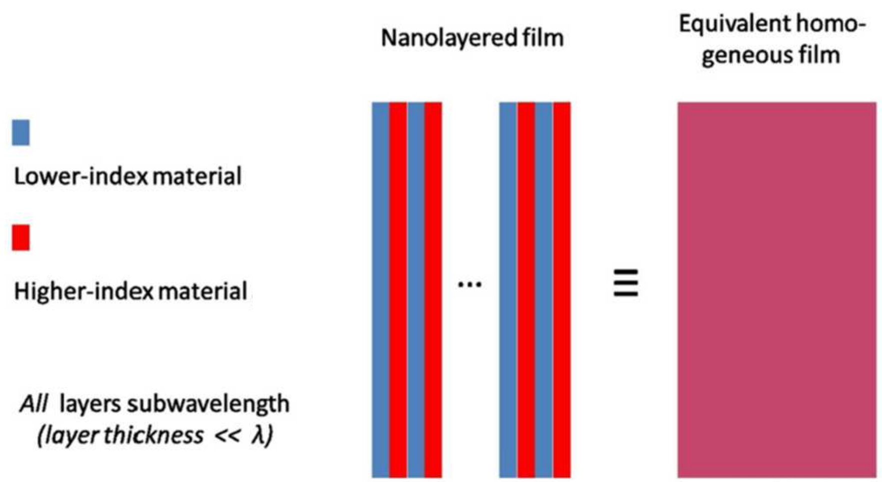
“TEM indicates that no interface layer forms during annealing”

After anneal at 300°C 24hours



Nanolayer Composite Modeling

SWL composite properties depend *only* on the constituents' properties and the thickness ratio $d_{H,tot}/d_{L,tot}$



$$n_{eff} = \left[r_H {n_H}^2 + (1 - r_H){n_L}^2\right]^{1/2}$$

Drude's formula

$${Y_f}^{\parallel} = r_H Y_H + (1 - r_H) Y_L$$

Voigt formula

$${Y_f}^{\perp} = [r_H/Y_H + (1 - r_H)/Y_L]^{-1}$$

Reuss formula

$$\phi_f = \frac{\left(\frac{Y_s}{Y_H} + \frac{Y_H}{Y_s}\right) r_H \phi_H + \left(\frac{Y_s}{Y_H} + \frac{Y_H}{Y_s}\right)(1 - r_H)\phi_L}{Y_s[r_H/Y_H + (1 - r_H)/Y_L] + {Y_s}^{-1}[r_H/Y_H + (1 - r_H)/Y_L]^{-1}} \quad , \quad r_H = \frac{d_{H,tot}}{d_{L,tot} + d_{H,tot}}$$

Harry's formula

Nanolayer Composite Design, contd.

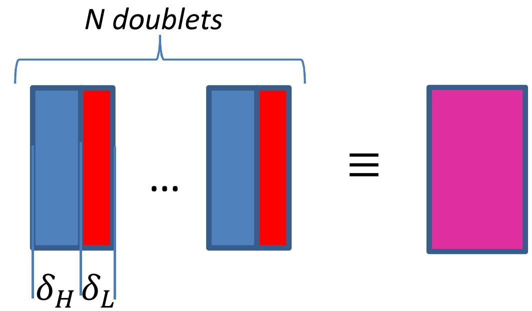
For given $n_{L,H}$, prescribing the slab index n_{eff} determines uniquely the thickness ratio of the low/high index materials in it (from Drude's equation),

$$\frac{\delta_L}{\delta_H} = \left(\frac{{n_H}^2 - {n_{eff}}^2}{{n_{eff}}^2 - {n_L}^2}\right)$$

Further prescribing the optical thickness *z* of the composite slab (in units of the local wavelength), and the minimum thickness of the nanolayers) yields *all equivalent* slab designs

$$(N, \delta_H, \delta_L)$$

(from $N(\delta_H + \delta_L) = z\lambda_0 {n_{eff}}^{-1}$).



Equivalent TiO_2/SiO_2 Subwavelength Doublet Based QWL etalons with n_{eff}=2.09

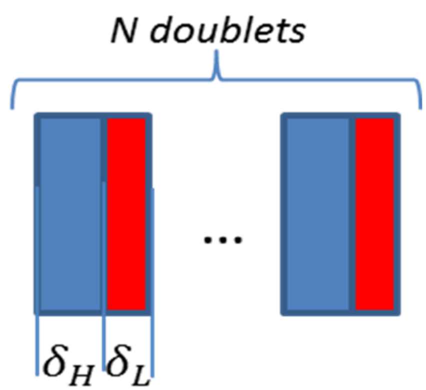
N	$\delta_{TiO_2}[nm]$	$\delta_{SiO_2}[nm]$
1	78.0559	49.2168
2	39.0279	24.6084
3	26.0186	16.4056
4	19.514	12.3042
5	15.6112	9.84337
6	13.0093	8.20281
7	11.1508	7.03098
8	9.75699	6.1521
9	8.67288	5.46854
10	7.80559	4.92168
11	7.09599	4.47426
12	6.50466	4.1014
13	6.0043	3.78591
14	5.57542	3.51549
15	5.20373	3.28112
16	4.87849	3.07605
17	4.59152	2.89511
18	4.33644	2.73427
19	4.1082	2.59036
20	3.90279	2.46084
21	3.71695	2.34366
22	3.548	2.23713
23	3.39373	2.13986
24	3.25233	2.0507
25	3.12224	1.96867

Nanolayer Composite Design, contd.

Composite properties depend *only* on the *constituents' properties* and thickness ratio $d_{H,tot}/d_{L,tot}$.

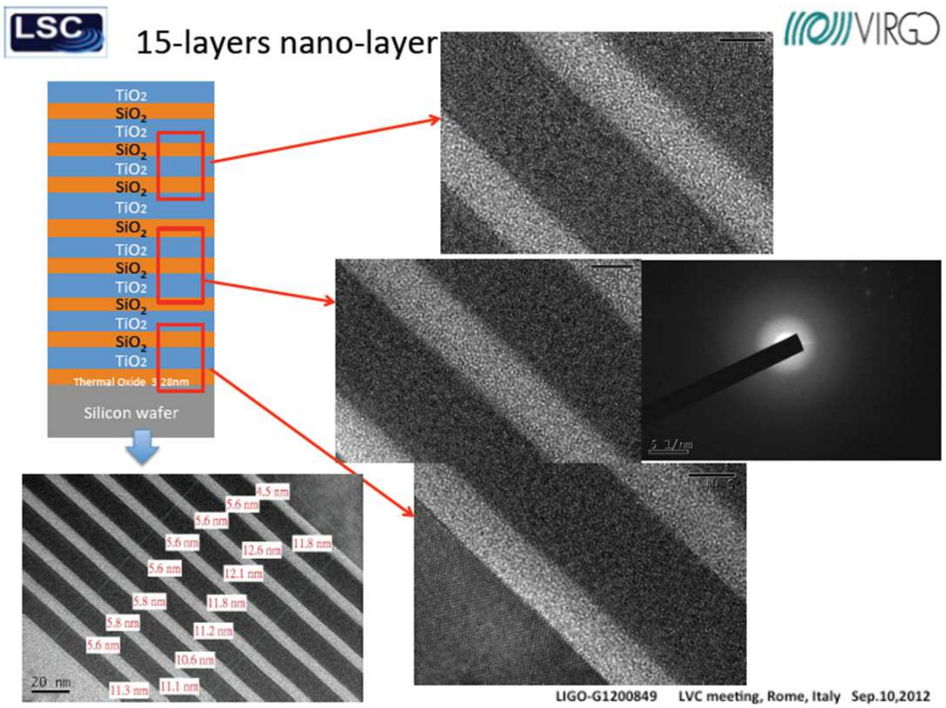
Composites using the same materials, and *differing in the thicknesses of the individual layers, but having the same thickness ratio* $d_{H,tot}/d_{L,tot}$ are *equivalent*.

The possibly simplest way to produce them is by laying down identical *subwavelength doublets (SWD)* with thicknesses $\delta_{(L,H)} = d_{(L,H),tot}/N$



Luckily, even *relatively large thickness errors* in the *individual* low/high index layer *thicknesses* are *irrelevant*, provided *each layer is sub-wavelength* and *the total thickness ratio has the design value*. *Mild* technological challenges…

Nanolayer Composite Prototypes



Deposition rate: TiO_2 3.40 nm/min SiO_2 7.79 nm/min						
layer	Designed thickness	Deposition time	Measured thickness (TEM)	Thickness error	Measured thickness (Ellipsometry)	Thickness error
15	10.64nm	3'8"	11.30 nm	6.2%	Not available	
14	6.02nm	46"	5.60 nm	7.0%		
13	10.64nm	3'8"	11.10 nm	4.3%		
12	6.02nm	46"	5.80 nm	3.8%		
11	10.64nm	3'8"	10.60 nm	0.4%		
10	6.02nm	46"	5.80 nm	3.7%		
9	10.64nm	3'8"	11.20nm	5.3%		
8	6.02nm	46"	5.60nm	7.0%		
7	10.64nm	3'8"	11.80nm	11%		
6	6.02nm	46"	5.60 nm	7.0%		
5	10.64nm	3'8"	12.10 nm	13.7%		
4	6.02nm	46"	5.60nm	7.0%		
3	10.64nm	3'8"	12.60 nm	18.4%		
2	6.02nm	46"	5.60 nm	7.0%		
1	10.64nm	3'8"	11.80 nm	10.9%		
TiO_2 average thickness for layer1 : 12.06 nm Standard deviation : 0.14 nm SiO_2 average thickness for layer 2 : 5.24 nm Standard deviation : 0.20 nm						

Full details in S. Chao et al., LIGO-G1200849

Nanolayered vs Cosputtered

Optical Properties

Co-sputtered (Bruggemann formula)

$$r_H \frac{{n_H}^2 - {n_{eff}}^2}{\gamma {n_H}^2 + (1-\gamma){n_{eff}}^2} =$$

$$= (1-r_H)\ \frac{{n_{eff}}^2 - {n_L}^2}{\gamma {n_L}^2 + (1-\gamma){n_{eff}}^2}$$

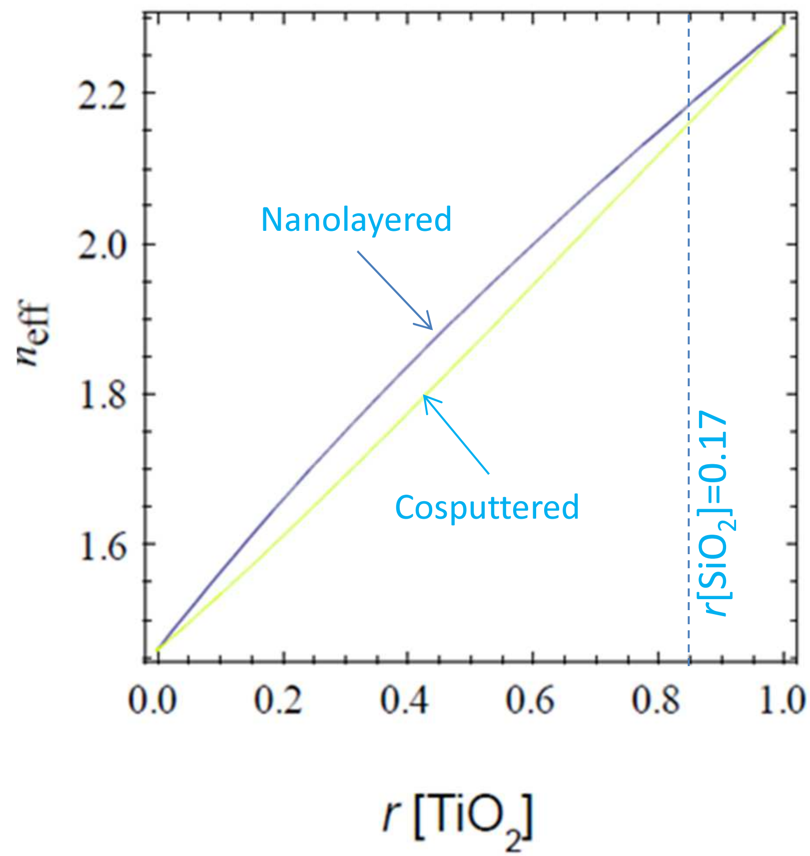
γ is Bruggemann shape-factor (1/3 for sph. incl.)

Nanolayered (Drude's formula)

$$n_{eff} = [r_H {n_H}^2 + $$

$$+(1-r_H){n_L}^2]^{1/2}$$

SiO_2/TiO_2 Composites



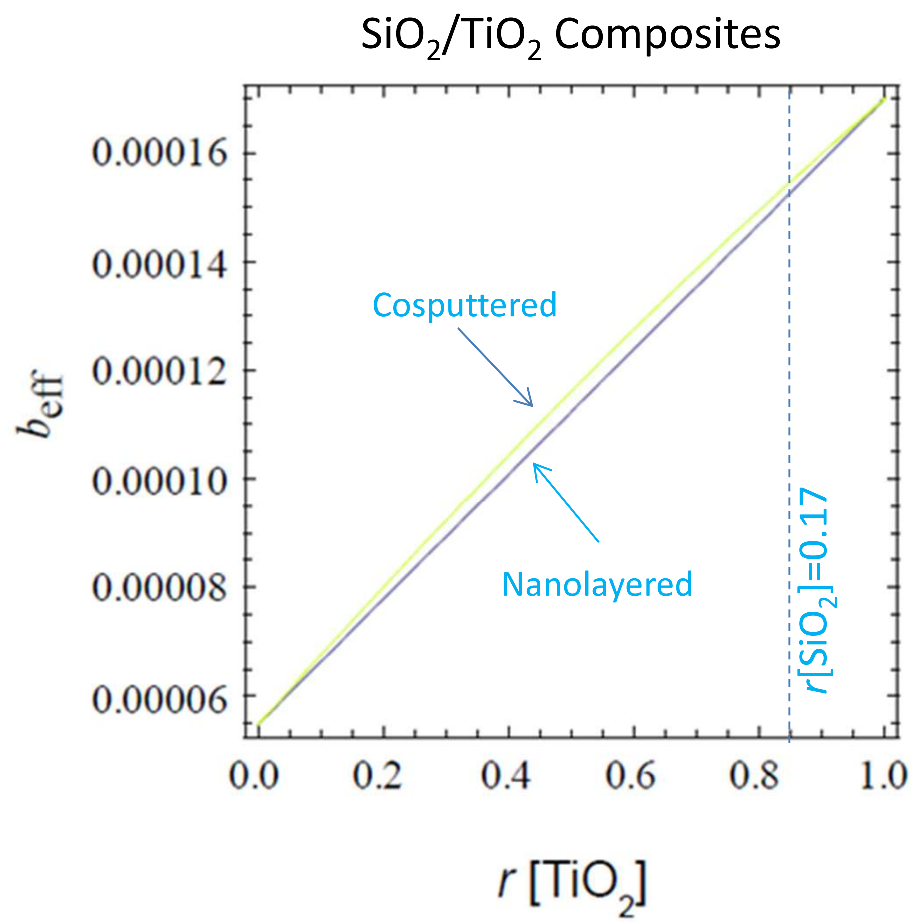
Nanolayered vs Cosputtered, contd.

Viscoelastic Properties
(noise coefficients)

Material noisyness per unit thick-ness is represented by the coeff-icients

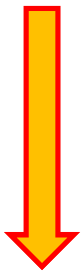
$$b_{eff} = \frac{\phi_{eff}}{n_{eff}} \left(\frac{Y_{eff}}{Y_s} + \frac{Y_s}{Y_{eff}} \right)$$

(Silica substrate in simulation)



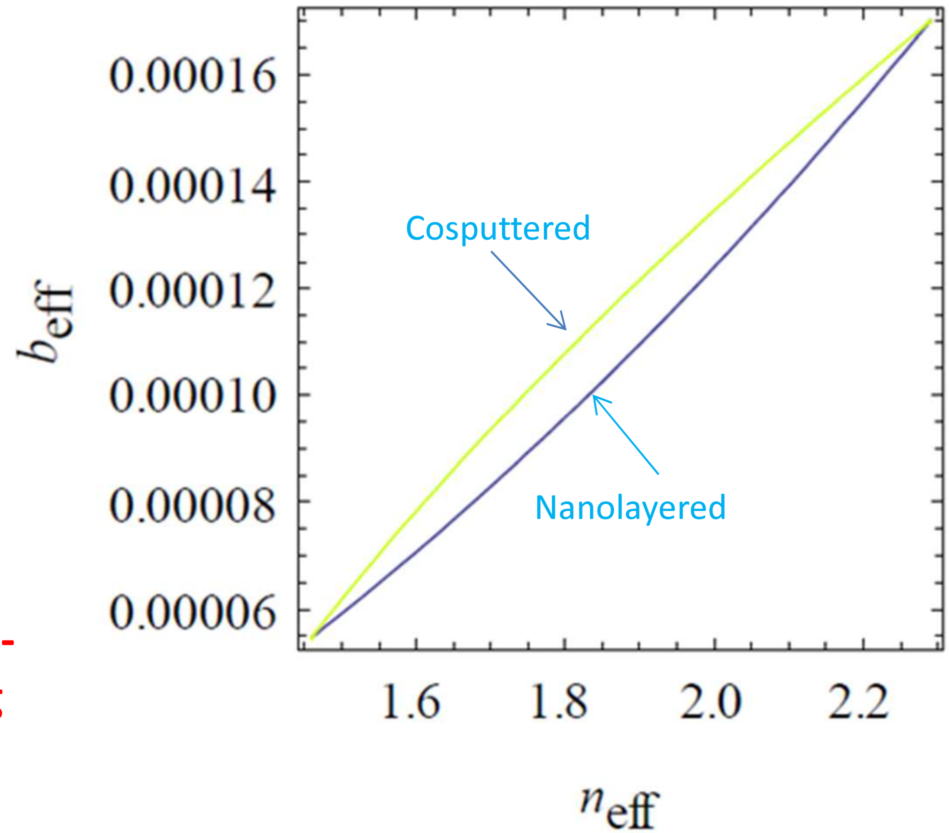
Nanolayered vs Cosputtered, contd.

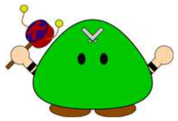
Combine previous results, so as to display b_{eff} vs n_{eff}



Nanolayered SiO2/TiO2 composites are *less noisy* , compared to co-sputtered SiO2/TiO2 composites having *the same refraction index*

SiO_2/TiO_2 Composites





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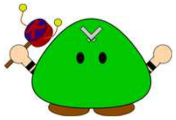
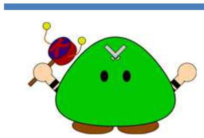


AdCOAT Mission

"Investigating, characterizing and comparing the properties (morpho-logical, structural, optical and viscoelastic) of Silica::Titania and Si-lica::Hafnia mixtures, both nm-layered and co-sputtered, both at ambient and cryogenic temperatures."

"Setting up a *coherent interaction* between different Italian Groups working on diverse aspects of coating science and technology, with Specific reference to Interferometric Detectors of gravitational waves."



AdCOAT – Working Groups

Innocenzo M. Pinto (PI, AdCOAT Coordinator)
Vincenzo Galdi, Vincenzo Pierro, Maria Principe, Dario Castellano, Silvio Savoia

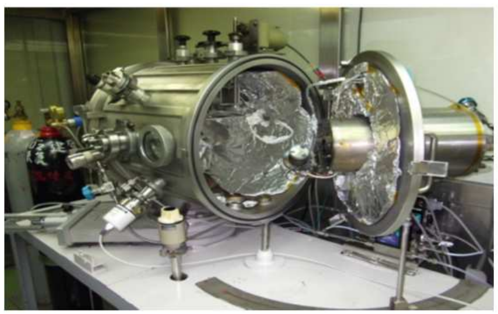
Maurizio Canepa (PI), Corrado Boragno, Francesco Buatier de Mongeot, Mauro Giovannini, Lorenzo Mattera

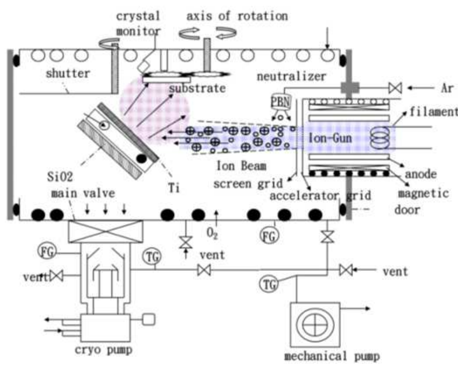
Helios Vocca (PI), Marzia Colombini , Luca Gammaitoni, Fabio Marchesoni, Maurizio Mattarelli, Igor Neri

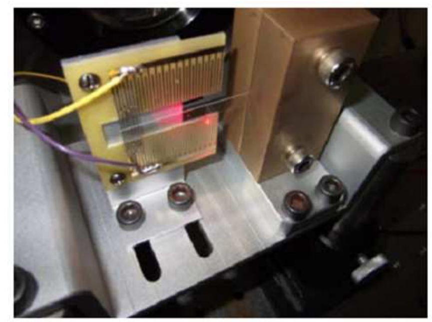
Alessio Rocchi (PI), Elisabetta Cesarini, Eugenio Coccia, Viviana Fafone, Yuri Minenkov

AdCOAT – Partners

Prof. Shiuh Chao and Co-workers, National Tsing Hua University, Taiwan, ROC. Author of seminal papers on SiO_2 doped TiO_2 [Wang and Chao, Opt. Lett. 23 (1998) 1417; Chao, JOSA A16 (1999) 1477; Chao et al., Appl. Opt. 40 (2002) 2177] .
In collaboration with USannio they produced and characterized the first prototypes of nm-layered SiO_2::TiO_2 composite films.







Statements of interest from, and plans of collaboration with the “big” coaters [*Laboratoire Materiaux Avances*, Lyon, FR (LMA), and *Commonwealth Scientific and Industrial Research Organisation*, Clayton, AU (CSIRO)].

AdCOAT – Partners, contd.

Statements of interest for the scientific case of AdCOAT, and letters of support to the related funding application to INFN, were provided by :

- Prof. Shiuh Chao, NTHU, National Tsing Hua University, Taiwan, ROC;
- Prof. Hai-Ping Chen, University of Florida, USA;
- Prof. Martin Fejer, Stanford University, Stanford University, Stanford CA, USA;
- Dr. Roberto Felici, European Synchrotron Radiation Facility (ESRF), Grenoble FR;
- Prof. Federico Ferrini, European Gravitational Observatory, Cascina (PI), IT
- Prof. Gregg Harry, American University, Washington DC, USA;
- Prof. Kazuaki Kuroda, ICRR-University of Tokyo, JP;
- Prof. Iain Martin, SUPA, University of Glasgow, UK;
- Prof. Ronny Nawrodt, Friedrich Schiller Universitat, Jena, GER;
- Prof. Steve Penn, Hobarth and Williams College, Geneva, NY, USA;
- Prof. Fulvio Ricci, Virgo Collaboration, Cascina (PI), IT
- Prof. Sheila Rowan, SUPA, University of Glasgow, UK;
- Prof. Sergey Vyatchanin, Moscow State University, Moscow, RU;

…many of them are here. Thanks to all of them, on behalf of the AdCOAT team.

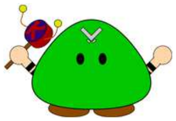
AdCOAT – WG Background/Tasks

Genoa WG - Coating morphology analysis [Prato et al., J. Phys. Conf. Ser., 228 (2010) 012020]; optical properties characterization [Prato et Al., Thin Solid Films 519 (2011) 2877]

Perugia WG – Dissipation mechanism modeling in glasses **[Travasso et al., Materials Science Eng. A521 (2009) 268;** Euro Physics Lett. 80 (2007) 50008**]** ; viscoelastic parameters measurement techniques [P. Amico et al., J. Phys. Conf. Ser., 32 (2006) 413]

Rome "Tor Vergata" WG - Cryogenic subsystems [Coccia, Physica B 280 (2000) 52]; development of gentle nodal suspension setup [Cesarini et al., Rev. Sci. Instr. 80 (2009) 1.3124800; CQG 27 (2010) 084031].

Sannio WG - Coating design [Villar et al., Phys. Rev.D81 (2010) 122001], EMT modeling of glassy oxide mixtures [Pinto et al., LIGO-G1100372], nm-layered composite modeling and design [Pinto et al., LIGO-G 1100586].



AdCOAT – Experimental Facilities

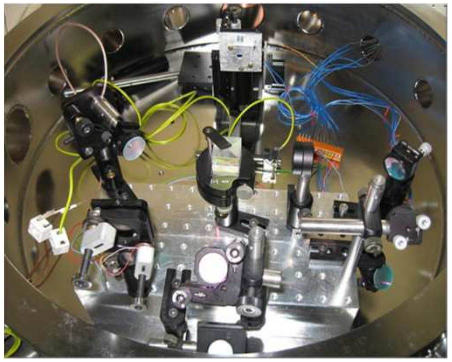
Genoa WG - HR-TEM (JEOL JEM 2010 + accessories); SEM (Zeiss EVO 40 HV + accessories); FE-SEM (Zeiss SUPRA 40 VP + accessories); SPM (Veeco Multimode Picoforce + accessories); AFM (Dimension 3000 +accessories); XRD (Philips XPERT MPD PRO); 2 x OSEs (Woollam M-2000 and VASE); some cryogenic facilities.

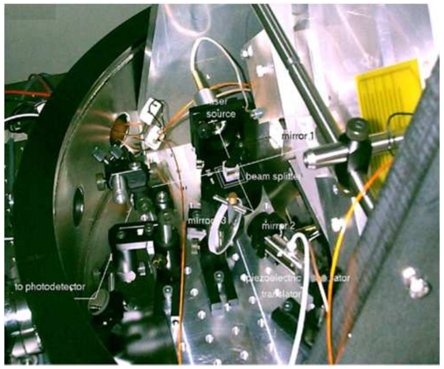
(courtesy M. Canepa, INFN Ge)

AdCOAT - Experimental Facilities, contd.

Perugia WG - Two cryostats (resp. nitrogen/helium, and pulse-tube). Three different setups for mechanical Q measurement at ambient temperature; optical lever based setup; frequency stabilized Michelson interferometer. FE - SEM for film surface quality analysis down to 2 nm.

3 optical systems (Michelson IFO, FP cavity and shadow-meter) with stabilized Laser for measuring mechanical modes of membranes and mirrors, at ambient and cryogenic temperatures.

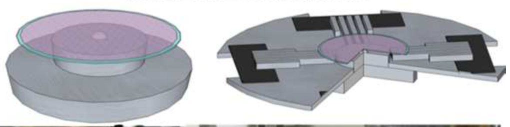


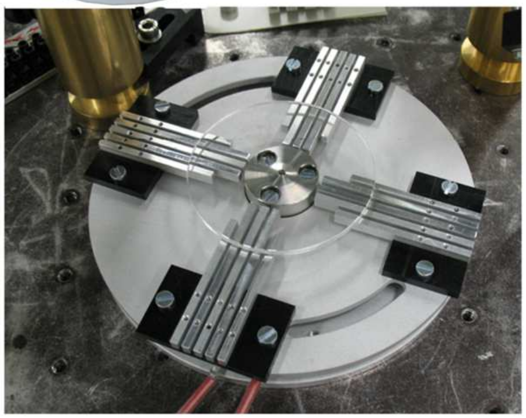


(courtesy H. Vocca, INFN Pg)

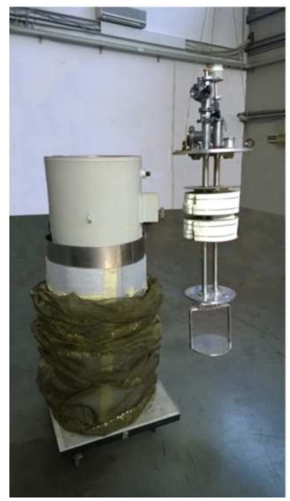
AdCOAT - Experimental Facilities, contd.

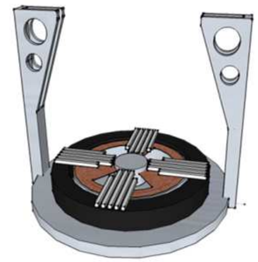
GeNS (Gentle Nodal Suspension)





Rome WG - Gentle nodal suspension (GeNS) based setups for Q measurement at ambient and (soon) cryogenic temperatures.
Some systems for thermal and laser annealing (CO_2 laser)





mechanical drawings by Matteo Lorenzini

- Laser assisted centering
- Q independent of suspension point
- coating preserved
- butterfly-mode matched exciters
- lowest loss angle measured so far (fused Silica) : ϕ=4.8 10^{-8}

- l-He cooling and radiation shield
- optical lever readout
- alternative (capacitive) readout
- electrostatic (comb) actuators
- cryogenic positioners

(courtesy E. Cesarini, INFN Rm-TV)

AdCOAT - Experimental Facilities, contd.

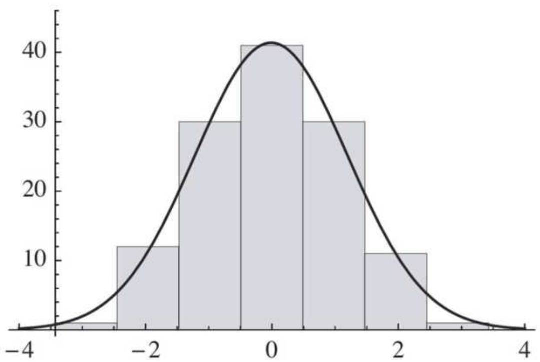
Sannio WG – Dual head pulse tube cryocooler (Sumitomo SRP - 052A - W71 D) . Two NVIDIA "Fermi" C2070 GPU based WS.

In house developed SW for coating simulation (thickness optimization, mixture analysis and design, nm-layered composites analysis and de-sign, statistical treatment of measu-rement residuals).

(courtesy M. Principe, UniSannio and INFN)

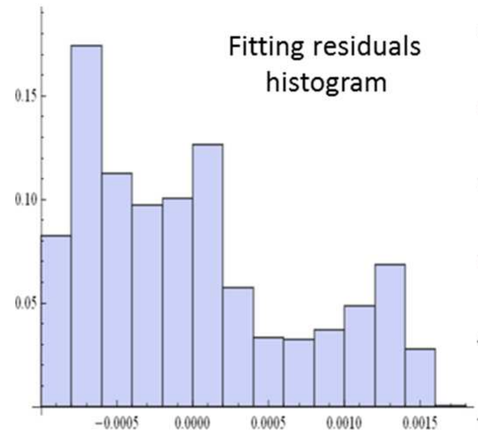
Digression : Fitting Residuals

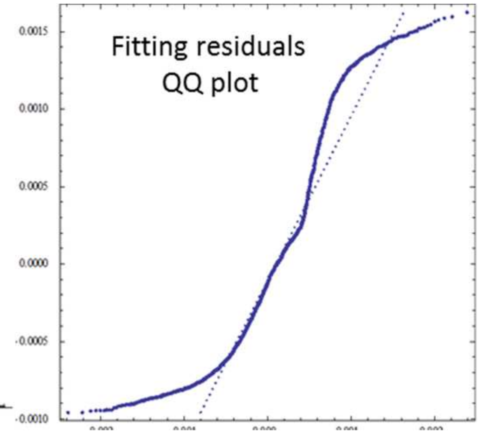
Typical loss angle fitting residuals, TNI measure-ments



[Villar et al., PRD 81 (2010)]

Typical loss angle fitting residual, clamped cantilever based ring-down measurement





[data courtesy N. Morgado (2008)]

Confidence intervals must be properly estimated in the non Gaussiann case.
May help reconciling discrepancies between TNI and ringdown measurements ?

Conclusions

… please, wish us good luck !