ICRR Research Results Presentation Meeting of Inter-University Research Program

Institute for Cosmic Ray Research
University of Tokyo

Nanolayered Coatings for Cryogenic Operation

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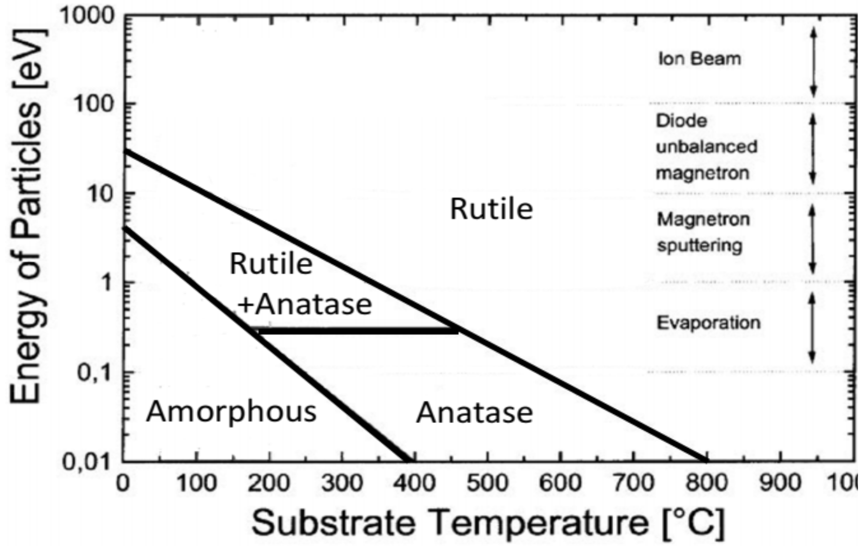
Tokyo, JP – December 13-14 2019

Outlook

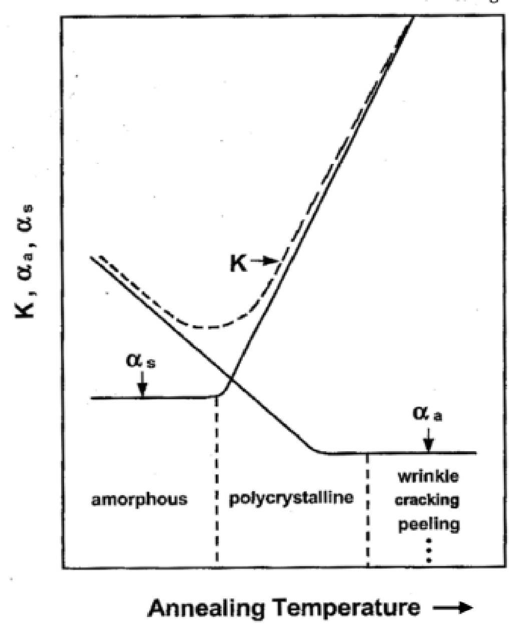
- Nanolayers: the Titania Paradigm
- Nanolayered Silica/Titania Composites
  - Suppression of Annealing-Induced Crystalization
  - Suppression of Cryogenic Loss-Peak
- Nanolayered-composite Based HR Coatings
- Feasibility
- The USannio/UniSA Labs & their VCR&D Research Program
- Candidate Materials
- Questions and Simulations
  - Nanocomposite Modeling
  - HR Coating Modeling (QWL)
- Best Nanolayered-composite Based HR Designs
- Conclusions

The TiO_2 Paradigm



J. Szczyrbowski, Surf. Coat. Technol. 112 (1999) 261–266.

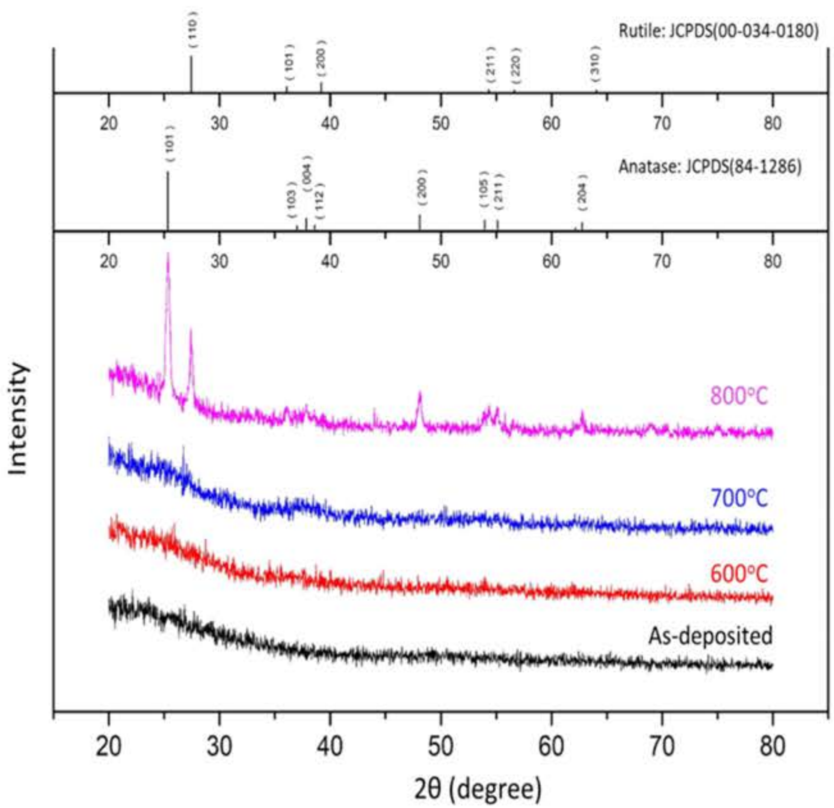
Extinction coefficient, and its absorption and scattering components, vs $T_{annealing}$.

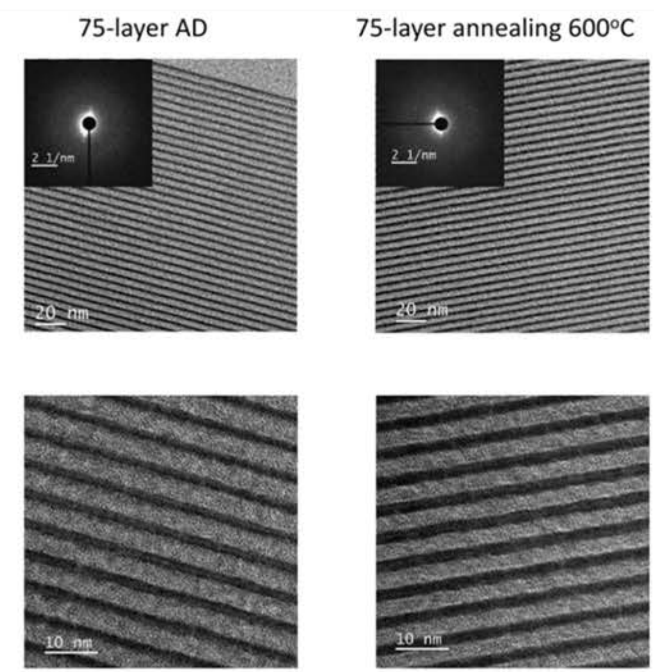


W.H. Wang & S. Chao, Opt. Lett., 23 (1998) 1417

SiO_2/TiO_2 Nanolayered Composites

[S. Chao et al., LIGO-G1900356]

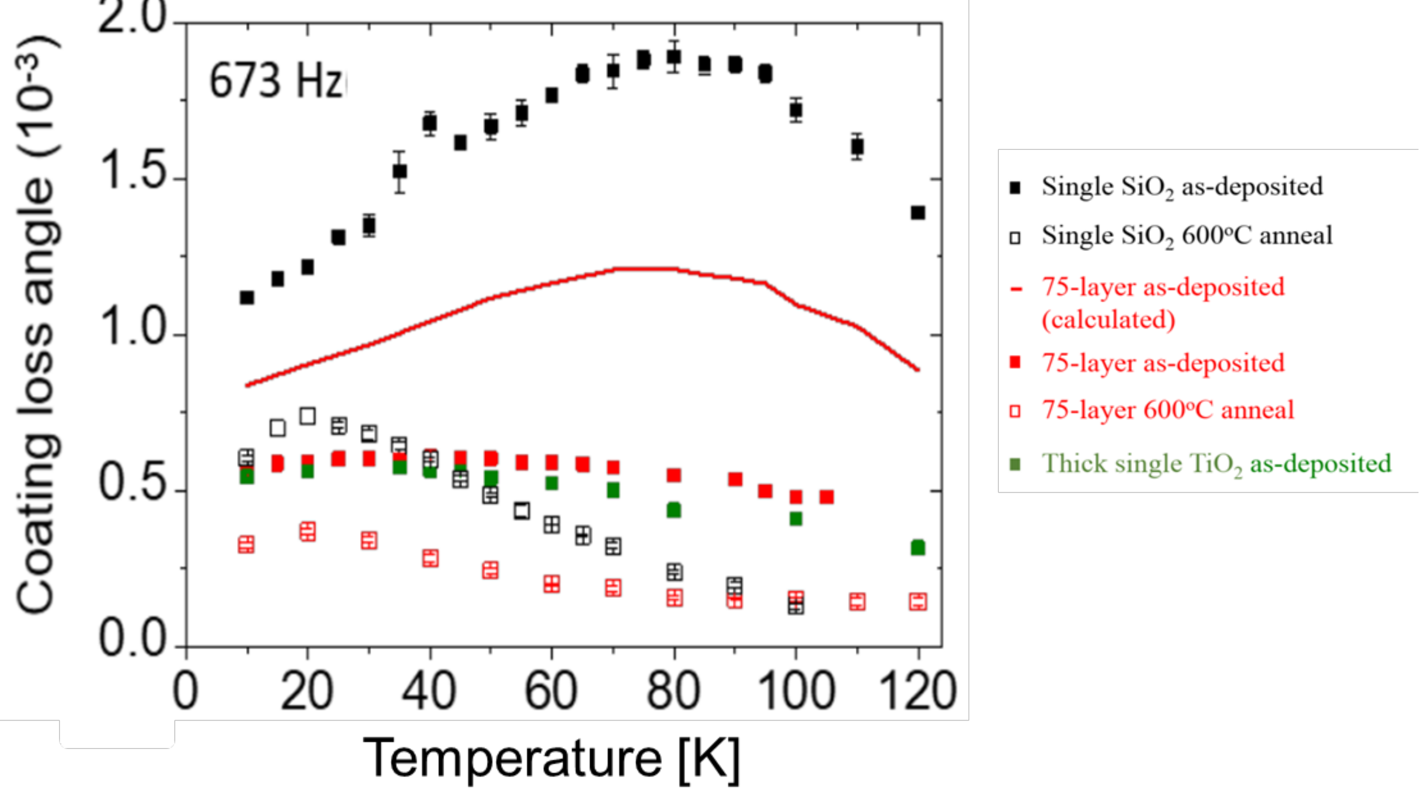




A 75 layer nanocomposite film [38 x (TiO_2=1.8nm), 37 x (SiO_2=3.6nm)] remains amorphous after annealing at 600C, and its morphology is preserved

Mechanical Losses of SiO_2/TiO_2 Nanolayers

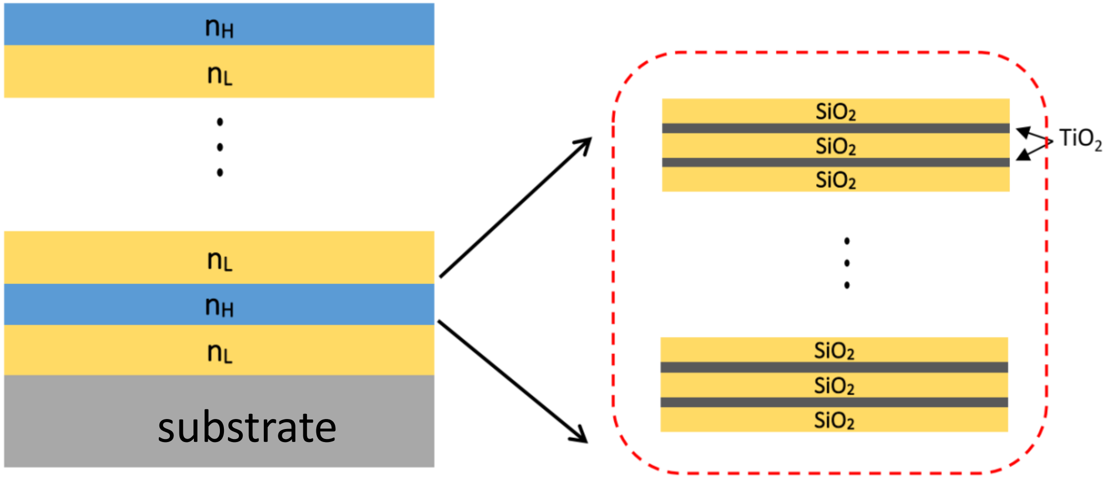
[S. Chao et al., LIGO-G1900356]



- The mechanical loss-angle of the [38 x (TiO_2=1.8nm), 37 x (SiO_2=3.6nm)] nanolayered film was reduced to $\approx 1 \cdot 10^{-4}$ after 600C annealing (a factor ≈ 9 lower than as deposited)
- **No cryopeak is inherited from Silica.**

Nanocomposite Based HR Coatings

Nanocomposite replacement for high-index coating layers



Feasibility

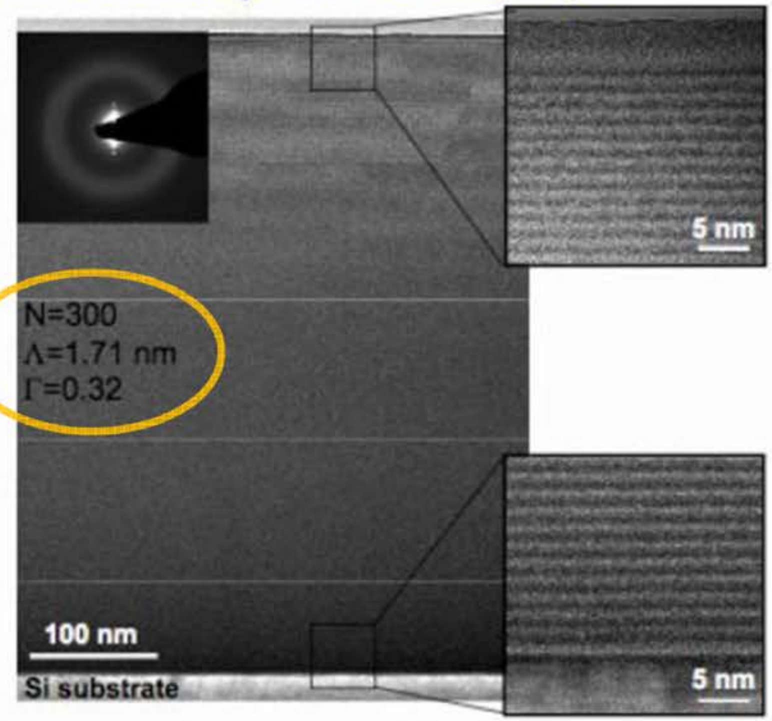
Nanolayers are *routinely deposited* to produce X-Ray Mirrors

Interference mirrors consisting of hundreds/thousands of nm scale layers, with sub - nm precision [see, e.g., Proc . 10th PXRMS Conf. (2008)] **, using**

- **Interleaved nm-scale "buffering" layers to prevent crystallization & maintain flatness** [E. Gullikson, Proc. 8th PXRMS (2006)]
- **Ion assisted (modulated) magnetron sputtering** [N. Ghafoor et al., Thin Sol. Films 516 (2008) 982]

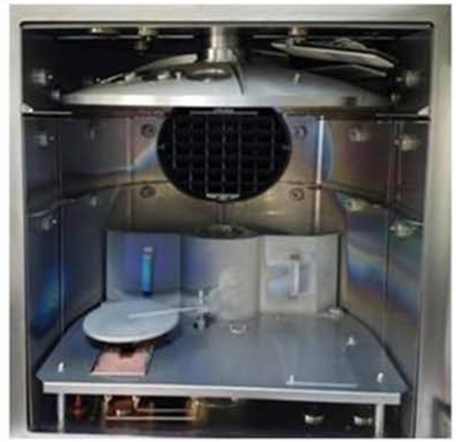
Control of stress, crystallite size, and roughness [D.L. Windt, Proc. SPIE (2007) vol 6688]

Interleaved B_4C- Cr/Sc multilayer



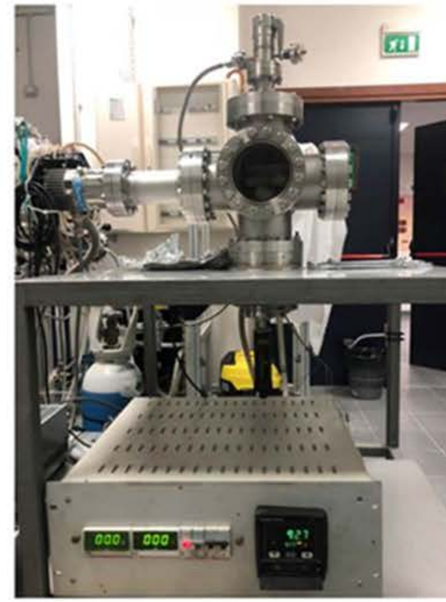
The USannio/UniSA Labs

University of Sannio Optical Film Deposition Lab



- Up to 6 co-deposited materials;
- Dual e-beam (one installed);
- Plasma IAD;
- Fully programmable deposition with GUI;
- Accuracy/repeatability at the Å level;

UniSA-SpNM Lab



- Annealing oven
- May accommodate up to 3-inch Ø dyes;
- Max annealing temperature : 900 ° C;
- Fully programmable annealing schedule;
- PID feedback-controlled heater power supply with 0.05 °C accuracy/stability;
- High-vacuum or controlled atmosphere (air, O_2, N_2 ,etc.)

The USannio/UniSA Coating Labs

Dept. of Physics "E.R. Caianiello", SpNM and CNR-SPIN Labs, University of Salerno

Left to right, top to bottom:
- FEI Tecnai 20 (TEM);
- Zeiss LEO-EVO 50 (EDS-SEM);
- Zeiss Sigma Gemini (FE-SEM);
- Renishaw Invia (Raman);
- JPK Nanowizard 3 (AFM);
- Philips 'Xpert-Pro (XRD);

Candidate Materials

	n @ 1064nm	k @ 1064nm	Y [Gpa]	ϕ	ν
SiO_2	1.4496 (1)	uncertain	72 (2)	$5.0 \cdot 10^{-5}$ (2,3)	0.17 (2)
Al_2O_3	1.7545 (1)	uncertain	210 (2)	$2.4 \cdot 10^{-4}$ (2)	0.22 (2)
Ta_2O_5	2.0760 (1)	uncertain	140 (2)	$4.72 \cdot 10^{-4}$ (3)	0.23 (2)
HfO_2	2.0813 (1)	uncertain	380 (2)	$5.9 \cdot 10^{-4}$ (2)	0.2 (2)
ZrO_2	2.1224 (1)	uncertain	200 (2)	$2.3 \cdot 10^{-4}$ (2)	0.27 (2)
Nb_2O_5	2.2537 (1)	uncertain	68 (2)	$4.6 \cdot 10^{-4}$ (2)	0.20 (2)
TiO_2	2.4789 (1)	uncertain	165 (3)	$1.4 \cdot 10^{-4}$ (3)	0.28 (2)

(1) https://refractiveindex.info/

(2) J. Franc et al., ET-021-09 (2009), arxiv/papers/0912/0912.0107.pdf; Flaminio et al., CQG 27 (2010) 083030

(3) M. Principe et al., Phys. Rev. D81 (2015) 022005, also Scott and MacKrone, Rev. Sci. Instr. 39 (1968) 821

More recent reviews of coating materials :

M. Granata et al., Phys. Rev. D93 (2016) 012007

G. Vajente, LIGO-G1900400

Questions and New Results

- *Blind* trial-and-error would be *impractically time (and money) consuming.*
  We implemented estensive simulations to provide guidelines to our experiments.

- Which materials are *more promising* to produce (binary) nanolayered films ?
  (Goals: noise reduction, cryo-compatibility)

- Which nanolayer *designs* are *more feasible* for a given material pair ?

Noise PSD (and Coating Loss Angle) Reduction Factor compared to Reference (Si/Ti::Ta, 5.32ppm@1064nm)							
	SiO_2	Al_2O_3	HfO_2	Ta_2O_5	ZrO_2	Nb_2O_5	TiO_2
SiO_2	x	>1	>1	>1	>0.794	>0792	>0.309
Al_2O_3		x	>1	>1	>0.833	>0.796	>0.352
HfO_2			x	>1	>1	>0.849	>0.663
Ta_2O_5				x	>1	>0.802	>0.415
ZrO_2					x	>0.791	>0.369
Nb_2O_5						x	>0.433
TiO_2							X

[I. Pinto, LIGO-G1902307]

HR Coating Modeling (QWL)

Transmittance

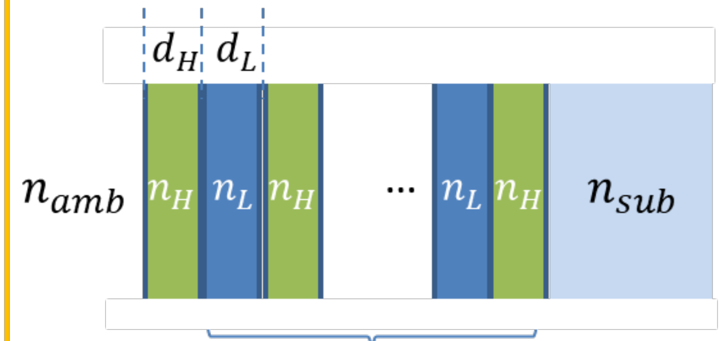
number of doublets

$$\Gamma = \frac{1 - \left(\frac{n_H}{n_L}\right)^{2N_D+2} \frac{{n_L}^2}{n_{amb} n_{sub}}}{1 + \left(\frac{n_H}{n_L}\right)^{2N_D+2} \frac{{n_L}^2}{n_{amb} n_{sub}}}, \qquad \tau_P = 1 - |\Gamma|^2$$

index ratio (*contrast*)

$\cong n_L$, typically

$(A)H(LH)^{N_D}(S)$



N_D doublets

(a further HWL low-index protective-cap may be added)

➡ yields N_D from prescribed τ_P and given contrast

Coating Loss Angle (Thermal (Brownian) Noise PSD) can be written

$$\phi_c = \frac{1}{4\pi^{1/2}} \frac{\lambda_0}{w} \frac{\phi_L}{n_L} \left(\frac{Y_L}{Y_s} + \frac{Y_s}{Y_L}\right) \left[N_d + \frac{n_L}{n_H} \frac{\phi_H \left(\frac{Y_H}{Y_s} + \frac{Y_s}{Y_H}\right)}{\phi_L \left(\frac{Y_L}{Y_s} + \frac{Y_s}{Y_L}\right)} (1 + N_d) \right]$$

blows up linearly with N_d

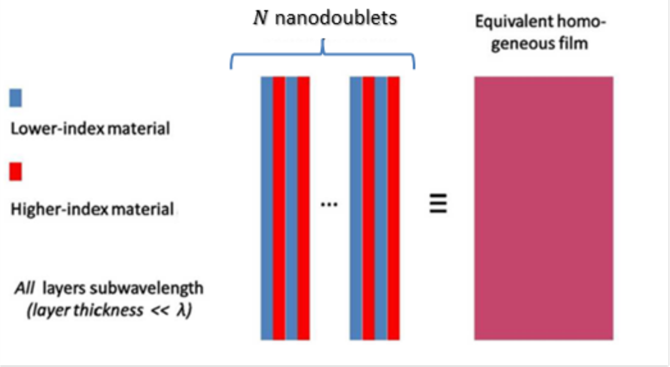
depends on choice of L-material only (usually the less noisy)

key quantity, depends on choice of H-material

Nanocomposite Modeling

$$n_{eff} = \left[r_H \tilde{n}_H^{\,2} + (1 - r_H)\tilde{n}_L^{\,2}\right]^{1/2}$$

Drude's formula



(normal incidence, $\delta_{L,H} \ll \lambda$)

$$\phi_{eff}\left(\frac{Y_{eff}}{Y_s} + \frac{Y_s}{Y_{eff}}\right) = \left(\frac{\tilde{Y}_H}{Y_s} + \frac{Y_s}{\tilde{Y}_H}\right) r_H \tilde{\phi}_H + \left(\frac{\tilde{Y}_L}{Y_s} + \frac{Y_s}{\tilde{Y}_L}\right)(1 - r_H)\tilde{\phi}_L$$

Let : $r_H = \dfrac{\delta_H}{\delta_L + \delta_H}$

thickness fraction

total metric thicknesses of high (δ_H) and low (δ_L) index nanocomposite contituents

- Assigning the effective index $n_{eff} \in (\tilde{n}_L, \tilde{n}_H)$ yields the ratio : $\dfrac{\delta_L}{\delta_H} = \left(\dfrac{\tilde{n}_H^{\,2} - n_{eff}^{\,2}}{n_{eff}^{\,2} - \tilde{n}_L^{\,2}}\right)$ (1)
- Prescribing the optical thickness z enforces the condition : $N(\delta_H + \delta_L) = z\dfrac{\lambda_0}{n_{eff}}$ (2)
  N being the total number of nanodoublets

Eqs (1) and (2) yield *an infinity of alternative and equivalent* designs (δ_L, δ_H, N). *Larger* N corresponding to *thinner* nano layers. Minimum and maximum allowed thicknesses are set by deposition machine and by chosen material (crystallization).

Simulation Algorithm

Determine PSD of reference ($\tau_P \leq 6\, ppm$) QWL design (Si/Ti::Ta_2O_5) [⇒ 17 doublets, $\tau_P = \tau_{P(ref)} = 5.25386\, ppm$]

↓

For each pair of ingredients in {SiO_2, Al_2O_3, HfO_2, Ta_2O_5, ZrO_2, Nb_2O_5, TiO_2}

↓

Determine $N_d = {N_d}^{(0)}$ yielding $\tau_P \leq \tau_{P(ref)}$ for $r_H = 1$

↓

Decrease r_H until $\tau_P = \tau_{P(ref)} \Rightarrow r_H(\tau_{P(ref)}, N_d)$

↓

Compute coating loss angle reduction factor R_ϕ for the QWL coating of N_d doublets using silica and a nanocomposite with $r_H = r_H(\tau_{P(ref)}, N_d)$

↓

While R_ϕ<1 , N_d=N_d+1 (loop back to "Decrease r_H")

↓

Move to next pair of ingredients (loop back to "Determine N_d")

SiO_2/TiO_2 Nanocomposite HR Coatings

Sample output of simulation code : Silica-Titania

QWL HR Coating – Silica (L) / Silica-Titania nanolayered composite (H)				
#of doublets in Coating	Silica Fraction in nanocomposite	Effective index of nanocomposite	Coating power transmittance	Noise PSD Reduction
12	0.0882959	2.40581	5.25386	0.308983
13	0.188226	2.32031	5.25386	0.322875
14	0.269172	2.24867	5.25386	0.337020
15	0.335952	2.1878	5.25386	0.351363
16	0.391907	2.13547	5.25386	0.365862
17	0.439421	2.09	5.25386	0.380487

Code yields alternative nanocomposites, featuring

- different Silica/Titania (thickness) fractions
- different refractive index

to produce alternative HR coatings having the same reference transmittance, but

- different number of QWL doublets
- different noise PSD reduction factors

Selecting a Nanocomposite Design

$r_L = 0.0882959$ ($R_{PSD} = 0.30983$)

N	$\delta_L[nm]$	$\delta_H[nm]$
7	1.39464	14.4005
8	1.22031	12.6004
9	1.08472	11.2004
10	0.97625	10.0803

$r_L = 0.188226$ ($R_{PSD} = 0.322875$)

N	$\delta_L[nm]$	$\delta_H[nm]$
18	1.19879	5.17009
19	1.13570	4.89798
20	1.07891	4.65308
21	1.02754	4.43151
22	0.98083	4.23008

$r_L = 0.269172$ ($R_{PSD} = 0.33702$)

N	$\delta_L[nm]$	$\delta_H[nm]$
28	1.13717	3.08754
29	1.09796	2.98108
30	1.06136	2.88171
31	1.02713	2.78875
32	0.99503	2.70160

Expected to crystallize before reaching $T_{ann} = 650C$

$r_L = 0.335952$ ($R_{PSD} = 0.351363$)

N	$\delta_L[nm]$	$\delta_H[nm]$
27	1.51282	2.99026
28	1.45879	2.88347
29	1.40848	2.78404
30	1.36153	2.69123
31	1.31761	2.60442
32	1.27644	2.52303
33	1.23776	2.44658
34	1.20135	2.37462
35	1.16703	2.30677
36	1.13461	2.24270
37	1.10395	2.18208
38	1.07490	2.12466
39	1.04733	2.07018
40	1.02115	2.01843
41	0.996245	1.96920

$r_L = 0.391907$ ($R_{PSD} = 0.365862$)

N	$\delta_L[nm]$	$\delta_H[nm]$
25	1.95268	3.02983
26	1.87758	2.91330
27	1.80804	2.80540
28	1.74347	2.70521
29	1.68335	2.61192
30	1.62724	2.52486
31	1.57474	2.44341
32	1.52553	2.36705
33	1.47930	2.29533
34	1.43580	2.22782
35	1.39477	2.16416
36	1.35603	2.10405
37	1.31938	2.04718
38	1.28466	1.99331
39	1.25172	1.94220
40	1.22043	1.89364
41	1.19066	1.84746
42	1.16231	1.80347
43	1.13528	1.76153
44	1.10948	1.72149
45	1.08482	1.68324
46	1.06124	1.64665
47	1.03866	1.61161
48	1.01702	1.57804
49	0.996267	1.54583

$r_L = 0.439421$ ($R_{PSD} = 0.3808437$)

N	$\delta_L[nm]$	$\delta_H[nm]$
23	2.43158	3.10202
24	2.33026	2.97277
25	2.23705	2.85386
26	2.15101	2.74409
27	2.07135	2.64246
28	1.99737	2.54809
29	1.92849	2.46022
30	1.86421	2.37821
31	1.80407	2.30150
32	1.74770	2.22958
33	1.69474	2.16201
34	1.64489	2.09842
35	1.59789	2.03847
36	1.55351	1.98184
37	1.51152	1.92828
38	1.47175	1.87754
39	1.43401	1.82940
40	1.39816	1.78366
41	1.36406	1.74016
42	1.33158	1.69872
43	1.30061	1.65922
44	1.27105	1.62151
45	1.24281	1.58548
46	1.21579	1.55101
47	1.18992	1.51801
48	1.16513	1.48638
49	1.14135	1.45605
50	1.11853	1.42693
51	1.09659	1.39895
52	1.07551	1.37205
53	1.05521	1.34616
54	1.03567	1.32123
55	1.01684	1.29721
56	0.99868	1.27404

Expected to remain amorphous at $T_{ann} = 650C$

Select a design featuring smallest R_{PSD} and minimum N, subject to $\delta_L \geq 0.9nm$, (feasible) and $\delta_H \leq 2nm$ (remaining amorphous after 650C annealing)

Best Nanocomposite Based HR Designs

$$(\delta_L, \delta_H)^{N_N}[\delta_{SiO_2}, (\delta_L, \delta_H)^{N_N}]^{N_D}$$

[I. Pinto, LIGO-G1902307]

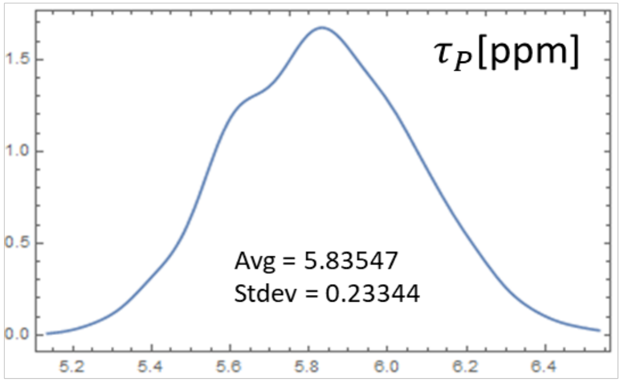
	Silica-Titania	Niobia-Titania	Zirconia-Titania	Alumina-Titania	Tantala-Titania
τ_P [ppm]	5.25386	5.25386	5.25386	5.25386	5.25386
R_{PSD}	0.36586	0.43352	0.464358	0.49998	0.57711
N_D	16	12	13	14	13
N_N	38	37	31	39	34
δ_L [nm]	1.28466	1.00106	1.71587	1.07652	1.39840
δ_H [nm]	1.99331	1.98720	1.98220	1.95661	1.97337
δ_{SiO2} [nm]	183.499	183.499	183.499	183.499	183.499

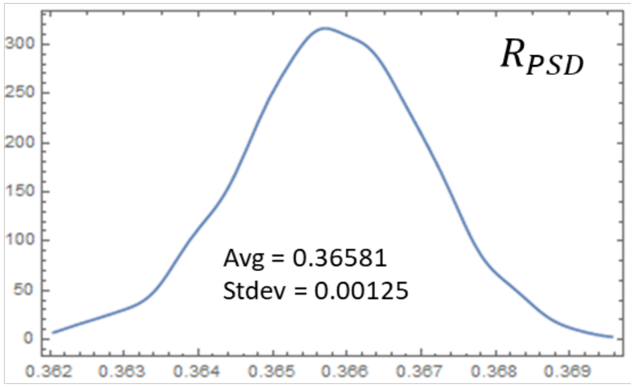
→ Reducing the noise PSD by a factor R_{PSD} yields an event rate boost by $(R_{PSD})^{-3/2}$

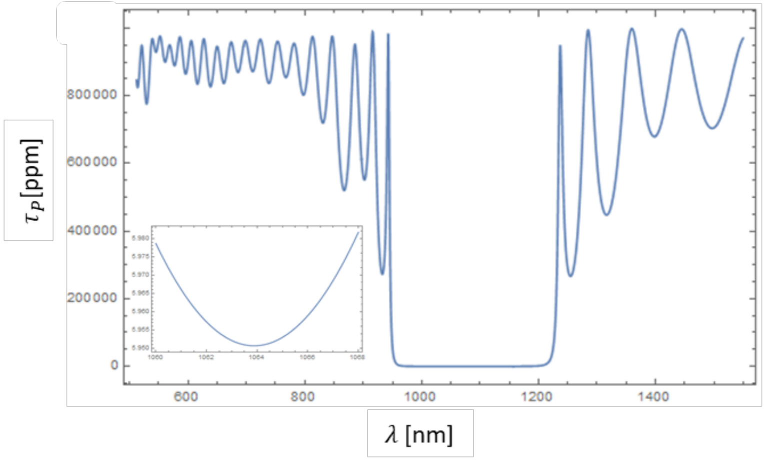
Robustness

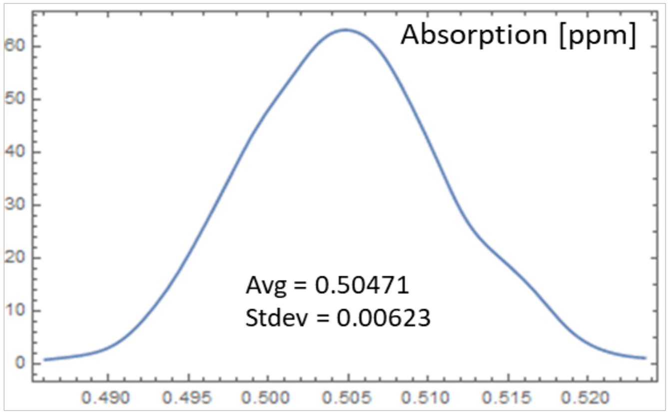
Assumption: i.i.d. Gaussian thickness errors with 0 avg. and .3nm std. deviation on each layer. 1000 trial-depositions.

	Silica-Titania
τ_P [ppm]	5.25386
R_{PSD}	0.36586
N_D	16
N_N	38
δ_L [nm]	1.28466
δ_H [nm]	1.99331
δ_{SiO2} [nm]	183.499



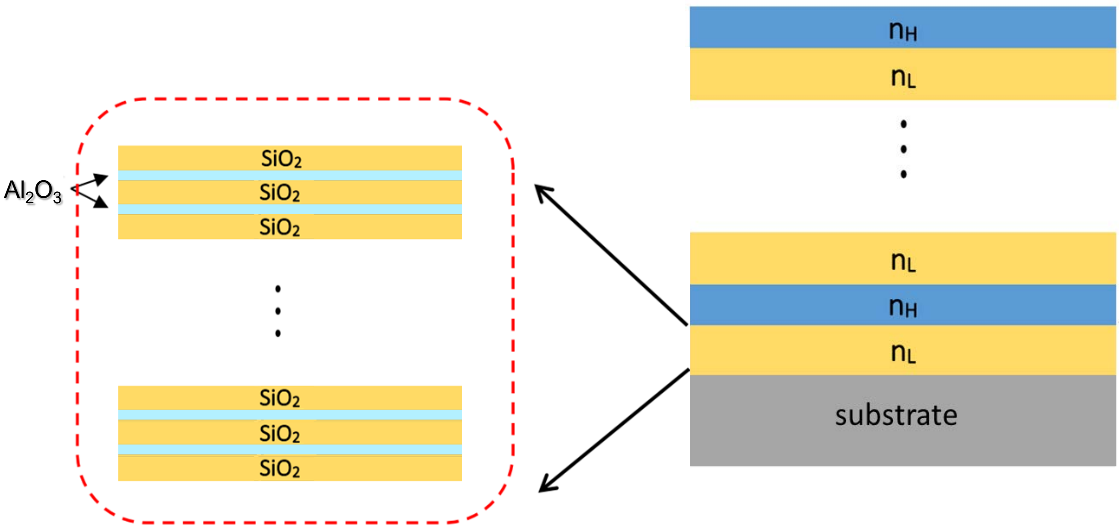






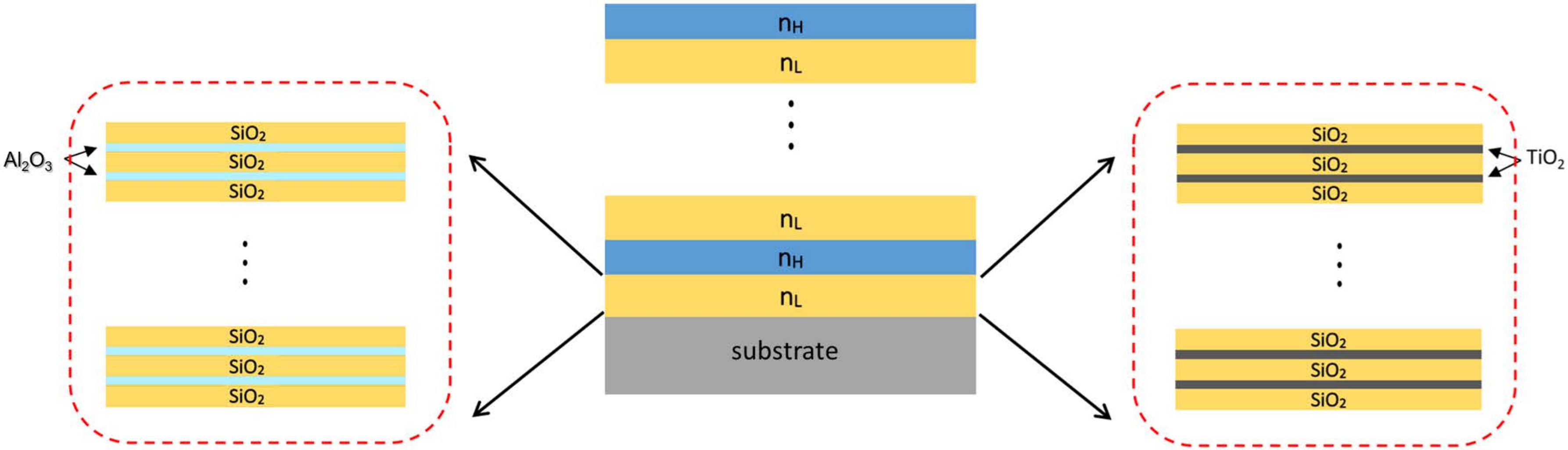
Nanocomposite Based HR Coatings, contd.

Nanocomposite replacement for low-index coating layers



Nanocomposite Based HR Coatings, contd.

Nanocomposite replacement for both low-index and high-index coating layers



Conclusions

- The nanolayered (binary) composites that may be used in place of the high index material in a QWL coating with the same τ_P as the reference Si/Ti::Ta2O5 design, to obtain $PSD \leq 0.5\, PSD_{REF}$ are (best to worst)

  {SiO_2, TiO_2}, {Nb_2O_5,TiO_2}, {ZrO_2, TiO_2}, {Al_2O_3, TiO_2}, {Ta_2O_5, TiO_2}

- The best (lowest refractive index, lowest mechanical loss) nanolayered (binary) composite that may be used in place of the low index material is {SiO_2, Al_2O_3}.
- For each material-pair it is possible to select a nanolayer design that should be free from crystallization upon annealing up to 650C, and feasible/easy to deposit (not too many nanolayers, not too thin).
- Extensive numerical simulations have been implemented to identify the above optimal designs. These are now pipelined for deposition and characterization in our Labs, in the frame of the VCR&D.
- Thickness optimization of the above coating designs may be added, and is expected to provide some extra noise reduction.

Thank you so much for your attention

ご 清 聴 ありがとうございました

直接会えないことをお びします