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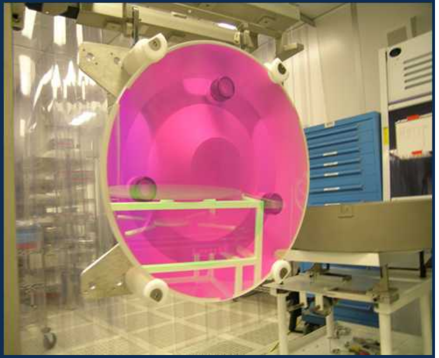

SAND2016-2771C

Titania thin films for broad bandwidth high reflection optical coatings to meet femtosecond-pulse, high-energy laser requirements

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Energy, Materials and Nanotechnology Meeting on Titanium Oxides, Honolulu, Hawaii, March 28, 2016

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Outline

- Production of optical coatings on meter class optics for high-energy, terawatt (TW) and petawatt (PW) lasers
- Multi-layer dielectric coatings for broad bandwidth high reflection (BBHR) of femtosecond (fs) class PW laser pulses
- Choice, properties and deposition of TiO_2 and SiO_2 thin films for high laser-induced damage threshold (LIDT) BBHR optical coatings
- Coating design based on TiO_2/SiO_2 layer pairs for low group delay dispersion (GDD) BBHR of fs class PW laser pulses
- LIDT results for the BBHR coatings, and morphology of laser-induced damage in the case of 675 fs high energy laser pulses

Sandia's Z-Backlighter Lasers

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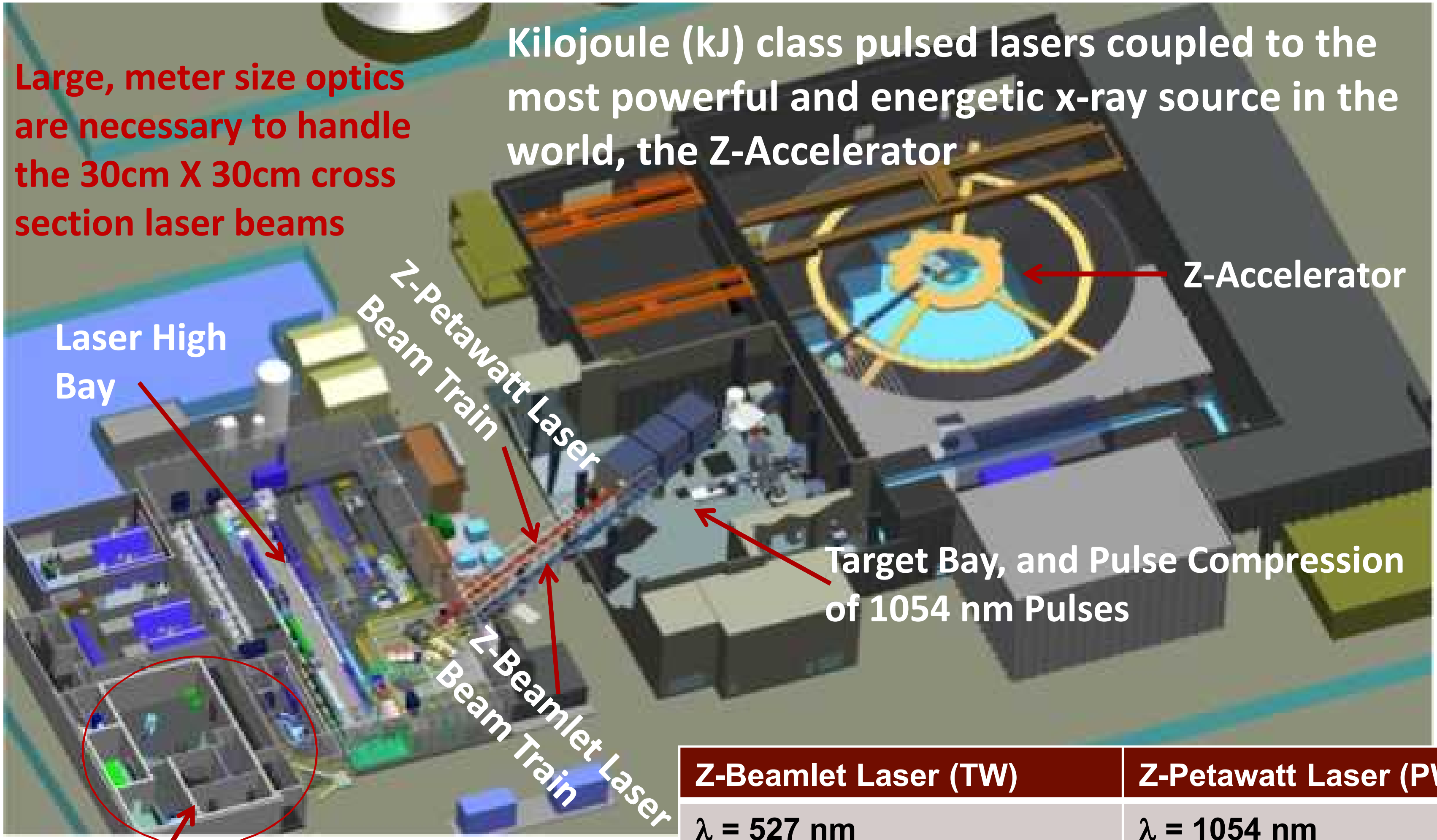

Kilojoule (kJ) class pulsed lasers coupled to the most powerful and energetic x-ray source in the world, the Z-Accelerator

Large, meter size optics are necessary to handle the 30cm X 30cm cross section laser beams

Optics Support Facility and Large Optics Coater
(Class 100 Clean Area)

Z-Beamlet Laser (TW)	Z-Petawatt Laser (PW)
λ = 527 nm	λ = 1054 nm
τ = 0.3 – 6 nanosecond (ns)	τ = 500 fs
I = 10^{16} W/cm^2	I = 10^{20} W/cm^2
E = 4 kJ	E = 0.5 kJ

Sandia's Large Optics Coating Facility:
Producing nano-scale thin films of uniform thickness and high LIDT on meter-scale optics for the Z-Backlighter Lasers

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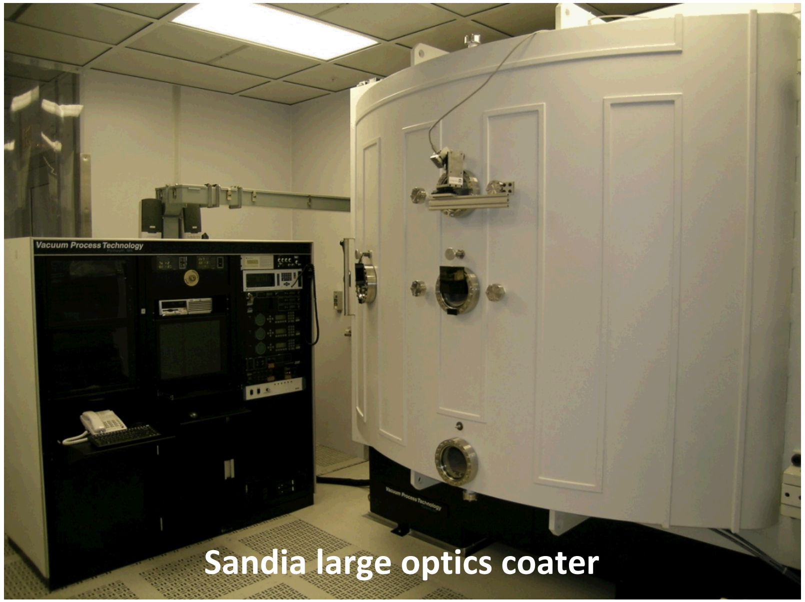
The large optics coater:

2.3 m X 2.3 m X 1.8 m vacuum chamber
- Installed in the Class 100 Optics Support Facility
- Equipped with three 22-inch cryo pumps

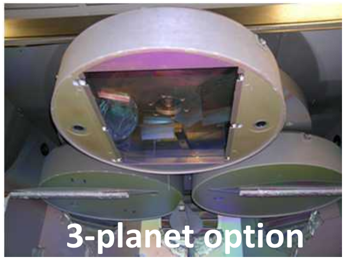
Thin film deposition by e-beam evaporation of materials
- e-beam evaporation – arguably the only way to uniformly deposit low-stress thin films on meter-scale optics
- e-beam sources (one 6-crucible and two dish sources) accommodate several thin film source materials
- Ion source – provides options for ion-assisted deposition (IAD) to densify coatings and control coating stress
- Oxygen back-pressure control for reactive deposition
- Crystal sensors for layer thickness monitoring

Masking & planetary motion ensure good coating uniformity
- 3-planet option – optic dimension up to 94 cm
- 2-planet counter-rotation option – optic dimension up to 1.2 m


Sandia large optics coater


3 e-beam sources


Ion source


3-planet option


2-planet option with counter rotation

Measurement of coating uniformity over a 94cm X 44cm optical substrate

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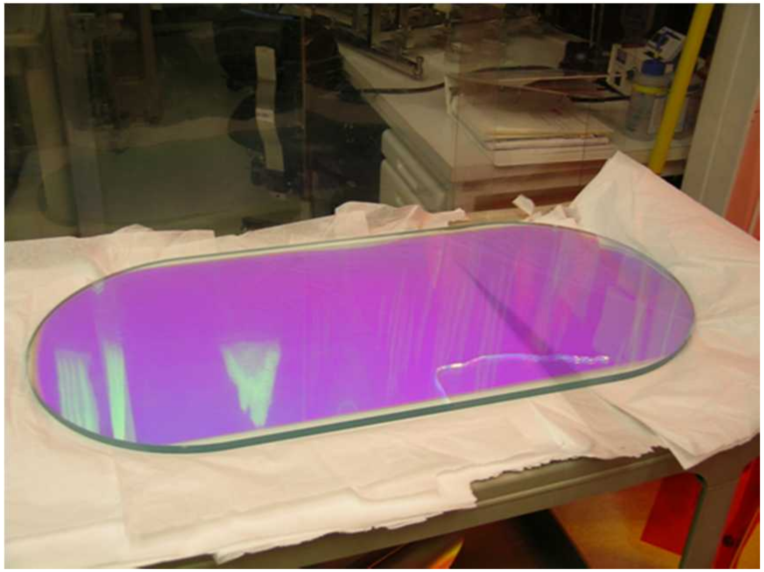
Coating: 17 HfO_2/SiO_2 layer pairs (~ 5 μm thickness) for HR at 45° AOI, deposited in the Sandia large optics coater

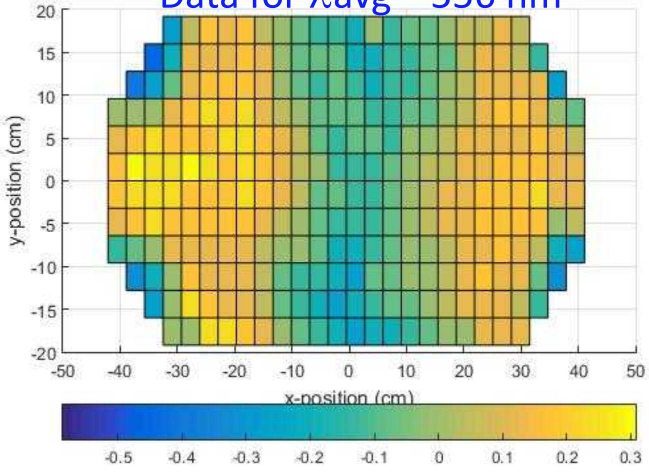


Spectrometer measurements tracked the wavelengths, λ, of 3 spectral reflection peaks near average wavelengths, λavg , as indicated.

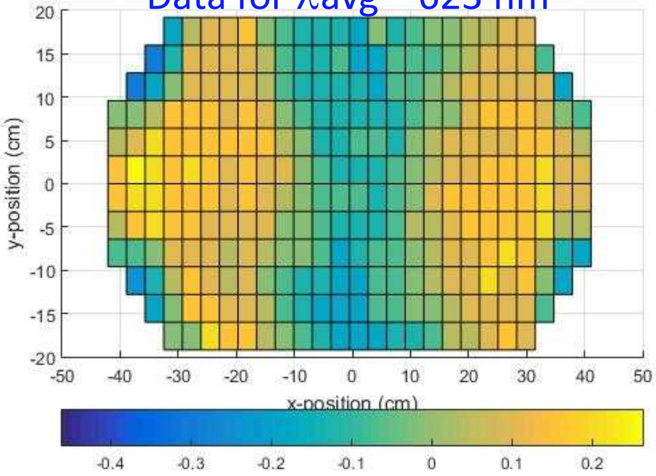
Data was taken over a grid with 3.2 cm between data points

The color scales show % Deviation, [(λ - λavg)/λavg] X 100, of the spectral peak wavelengths, λ, relative to their λavg.

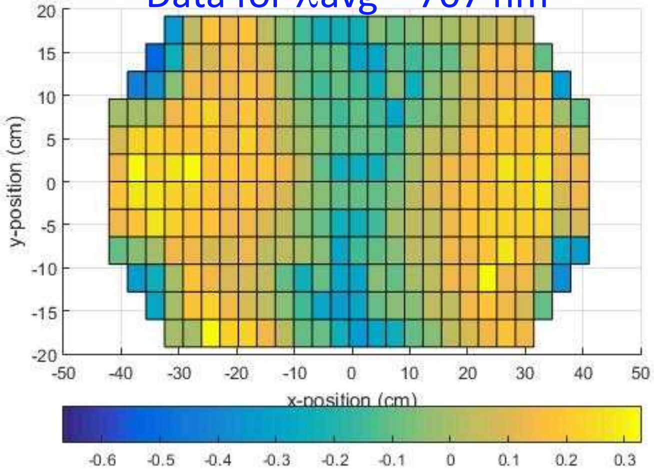
Data for λavg ~ 556 nm



Data for λavg ~ 623 nm



Data for λavg ~ 707 nm



Uniformity within +/- 0.5% over 94 cm X 44 cm coated area

Nanosecond (ns) to sub-picosecond (ps) pulse regimes and their BBHR requirements for PW class lasers

pulse duration ($\Delta\tau$) versus frequency range ($\Delta\nu$) and bandwidth ($\Delta\lambda$)

$$\Delta\tau = 1/\Delta\nu \qquad \Delta\lambda = (\lambda_o^2/c)\Delta\nu$$

(c = speed of light; λ_o = pulse center wavelength)

Example: 900 nm

line center pulses:

$\Delta\tau$	$\Delta\nu$ (10^9Hz)	$\Delta\lambda$ (nm)
1 ns	1	0.0027 nm
100 ps	10	0.027 nm
10 ps	100	0.27 nm
1 ps	1000	2.7 nm
100 fs	10000	27 nm
10 fs	100000	270 nm

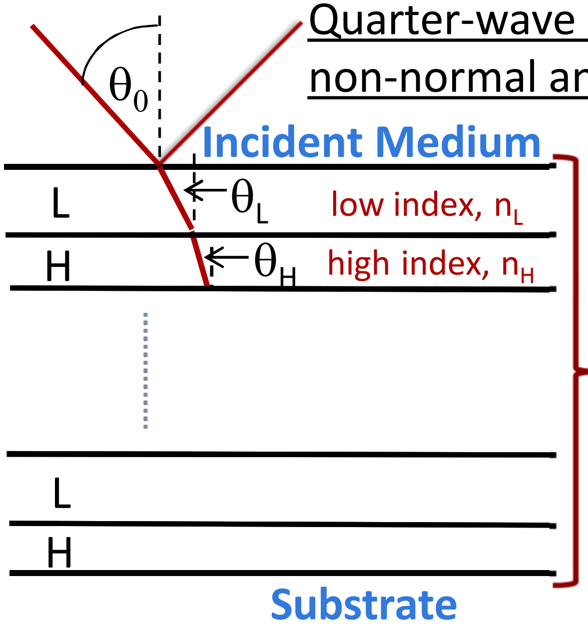
Range of pulse durations for high-energy PW lasers

BBHR regime

The temporal duration and profile of fs class pulses depend critically on the phase correlations between the broad range of the frequency components that combine to form the pulses

Illustration of quarter-wave HR coatings and dispersion

Quarter-wave HR layer structure showing non-normal angle of incidence (AOI)

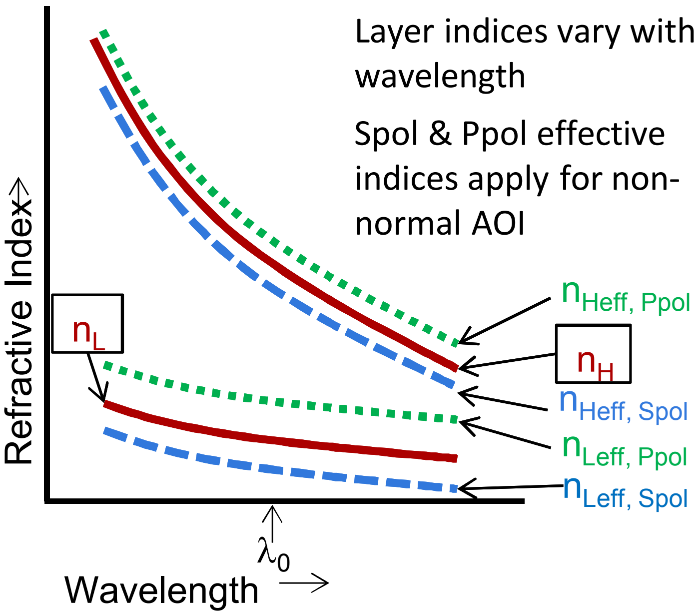


Alternating low (L) and high (H) index quarter-wave layers

The higher the layer index, the smaller the layer propagation angle (θ)

S & P polarization (Spol & Ppol) optical E-fields are normal to (Spol) and in (Ppol) the plane of incidence

Dispersion



Layer indices vary with wavelength

Spol & Ppol effective indices apply for non-normal AOI

BBHR depends on high layer index contrast ratio, n_H/n_L and large number of H/L layer pairs

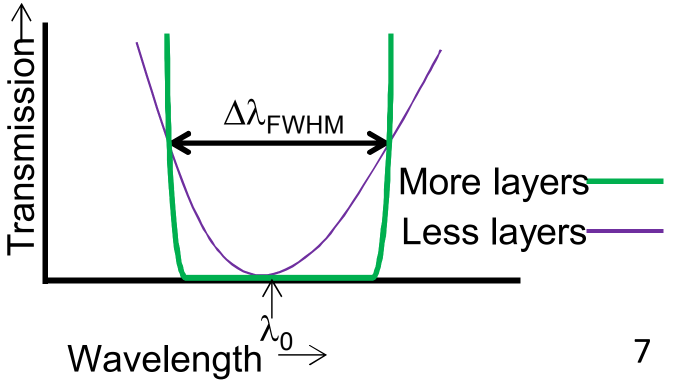
- bandwidth for HR increases with number of layers
- but is limited by $\Delta\lambda_{FWHM}$, which increases as n_H/n_L

Index and resulting phase changes due to dispersion over a BBHR band are large due to the large span of wavelengths

- optical path – (index) X (physical path) – and corresponding phase differ appreciably over BBHR bands
- propagation angles within layers, and corresponding optical paths/phases, also differ over BBHR bands

Coating transmission spectrum

(HR means low transmission & absorption)



BBHR coating dispersion issues for fs pulses

- fs pulses consist of a broad band of phase correlated frequency components

- Coating layers, due to dispersion, provide different optical thicknesses and phases for wavelengths across the BBHR band

- Optical propagation velocity in coating layers varies across frequencies of the fs pulse

- Non-linear group delay (GD) occurs for fs pulse frequency components
  - Leads to non-constant group delay dispersion (GDD) across BBHR band
  - disrupts relative phases between fs pulse frequency components
  - results in temporal broadening and distortion of the pulse.

Some common amorphous oxide thin films deposited by e-beam evaporation in Sandia's large optics coater

Thin film oxide	Band gap (eV)	Index at 500 nm	Source material and deposition process
TiO_2	3.3*	2.42	Ti metal, reactive with IAD
TiO_2	3.3*	2.41	Ti_3O_5, reactive with IAD
Nb_2O_5	3.4#	2.37	Nb_2O_5, reactive with IAD
Ta_2O_5	3.8*	2.19	Ta_2O_5, reactive with IAD
HfO_2	5.1*	2.02	Hf metal, reactive with IAD
Al_2O_3	6.5*	1.61	Al_2O_3, reactive with IAD
SiO_2	8.3*	1.47	SiO_2, IAD

*Phys. Rev. B **71**, 115109 (2005); #Appl. Opt. **53**, A186 (2014)

Dilemma: A BBHR coating for fs high energy laser pulses of 900 nm center wavelength and 200 nm HR band requires

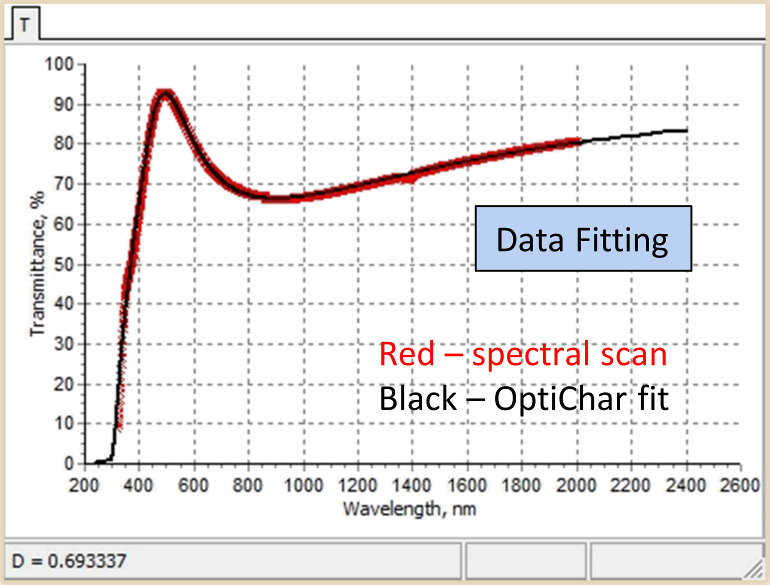
1. the highest index contrast afforded by TiO_2/SiO_2 layer pairs, and (in addition to low GDD)
2. must also afford high LIDT that is NOT favored by TiO_2 because of its low band gap

Challenge: Design such a BBHR coating based on TiO_2/SiO_2 layer pairs in a way that achieves the 200 nm bandwidth while maximizing LIDT and minimizing GDD

Development of high index TiO_2 layers at Sandia

- Reactive evaporation of Ti metal or Ti_2O_5 source materials in O_2 back pressure using IAD to optimize index of refraction
- Tested different temperatures, deposition rates, O_2 back pressures, and IAD settings for ~ 100 nm thick layers
- Measured the spectra of TiO_2 single layers using Lambda 950 spectrophotometer
- Analyzed the single layer spectra with OptiChar software in terms of index of refraction (n) and extinction coefficient (k)
  - We selected the normal dispersion model (Cauchy) for index and UV/Vis model (exponential) for absorption

Example: TiO_2 single-layer with Ti metal as source material - characterization using OptiChar software

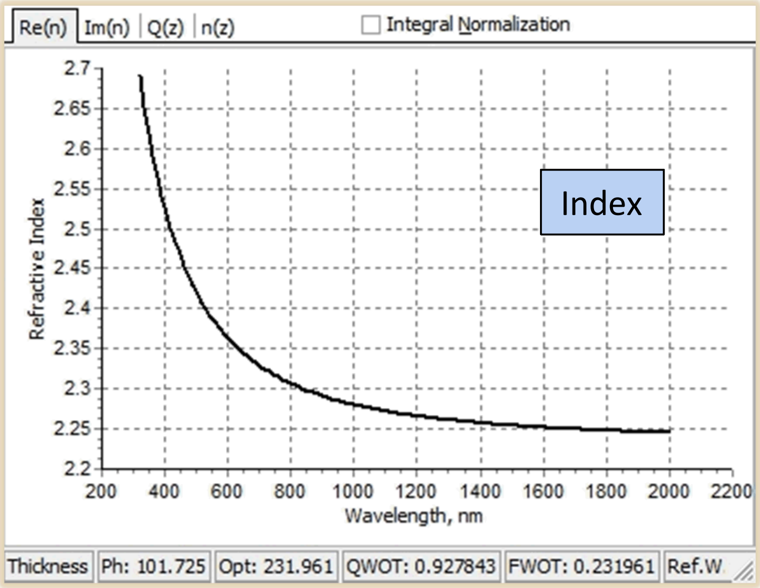


Data Fitting

Red – spectral scan

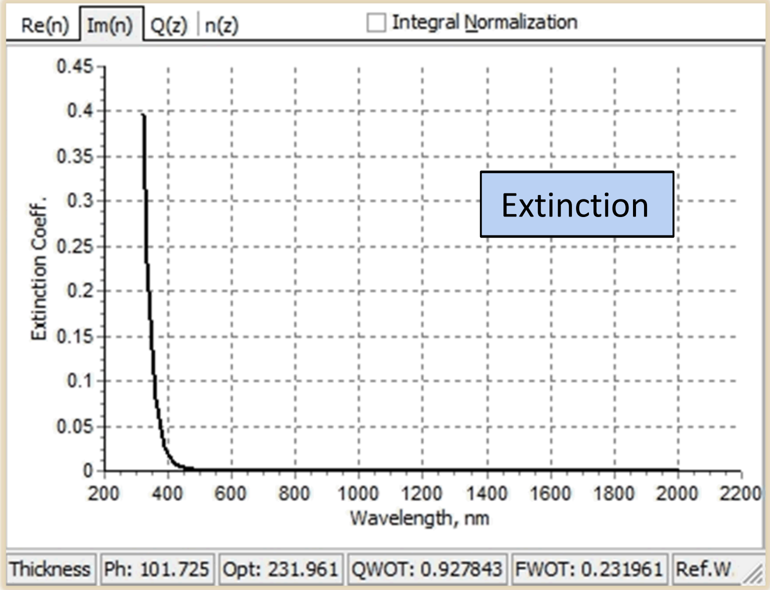
Black – OptiChar fit

D = 0.693337



Index

Thickness | Ph: 101.725 | Opt: 231.961 | QWOT: 0.927843 | FWOT: 0.231961 | Ref.W



Extinction

Thickness | Ph: 101.725 | Opt: 231.961 | QWOT: 0.927843 | FWOT: 0.231961 | Ref.W

Formulas:

Cauchy: $n(\lambda)=A_0+A_1/\lambda^2+A_2/\lambda^4$
- $A_0=2.233316$
- $A_1=0.046818$
- $A_2=0$

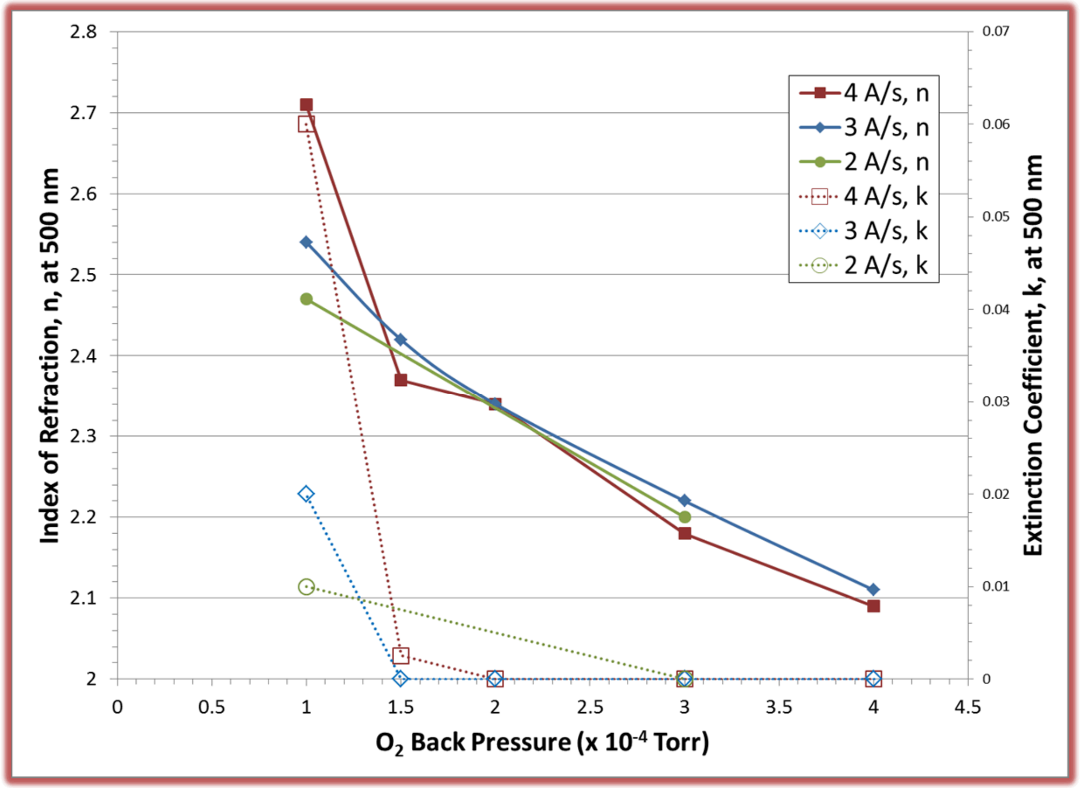
Exponential: $k(\lambda)=B_1\exp(B_2\lambda^{-1}+B_3\lambda)$
- $B_1=72946.15213$
- $B_2=0$
- $B_3=-37.880308$

The complex refractive index is n + ik. The optical extinction coefficient is $4\pi k/\lambda$ where λ is the wavelength of the light

TiO_2 single-layers with Ti metal as source material

Initial tests indicated higher indices at chamber temperature of 200 °C and IAD beam current/voltage of 600 mA/400 V

Varied the O_2 back pressure and deposition rate to find the highest index and lowest absorption TiO_2 layers



Lower O_2 back pressures lead to
higher layer indices &
higher extinction (absorption)
due to incomplete oxygenation of Ti
Good for HR bandwidth – Bad for LIDT

Higher O_2 back pressures lead to
lower layer indices &
lower extinction
due to complete oxygenation of Ti and excess O_2 trapped in the porous, amorphous layer structure
Bad for HR bandwidth – Good for LIDT

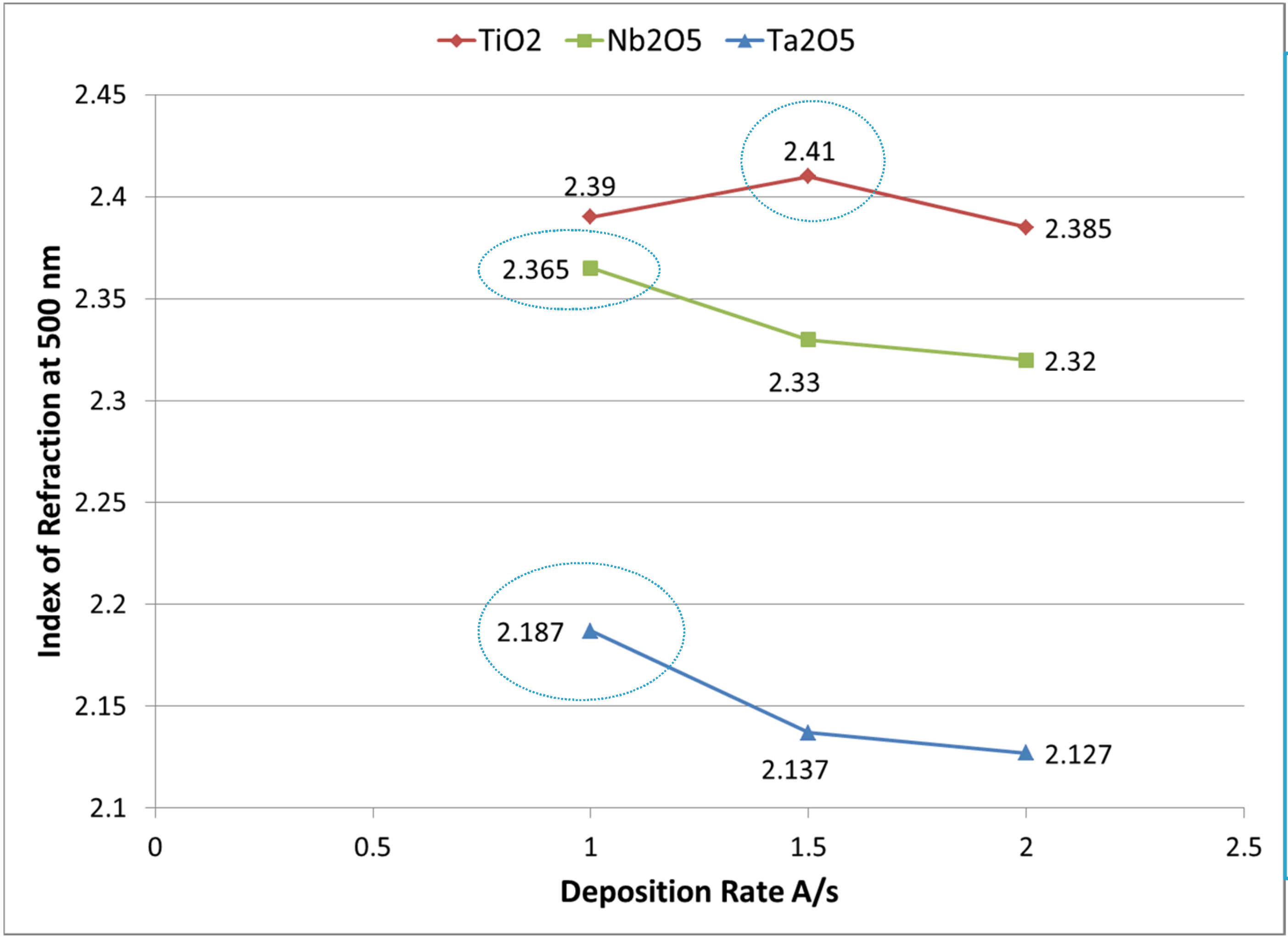
Optimal TiO_2 deposition settings:

- O_2 back pressure: 1.5×10^{-4} Torr
- Deposition rate: 3 Å/s

Optimal index at 500 nm = 2.42

Reaction of Ti with O_2 to form TiO_2 occurs on the surface of the thin film as it forms

Refractive index vs. deposition rate for single layers of TiO_2 (with Ti_3O_5 as source material), Nb_2O_5, and Ta_2O_5



IAD settings:
Beam Voltage, 400 V
Beam Current, 600 mA
45 sccm O_2
7 sccm Ar

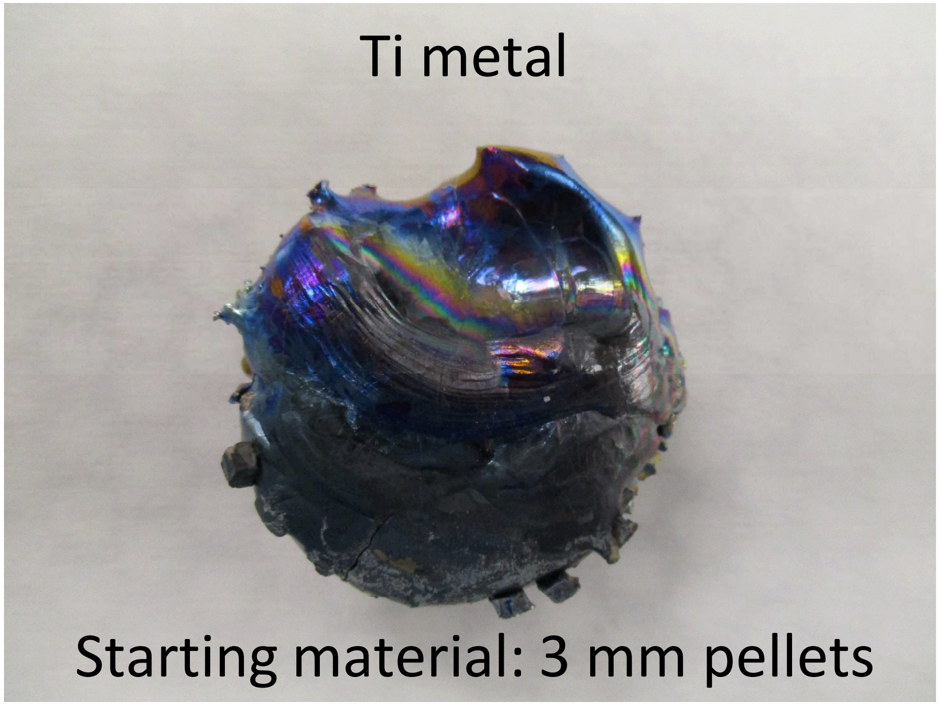
Pressure from ion source:
~ 1.8 e-4 Torr

Deposition pressure:
~2.2 e-4 Torr

Chamber temperature:
200 ^{o}C

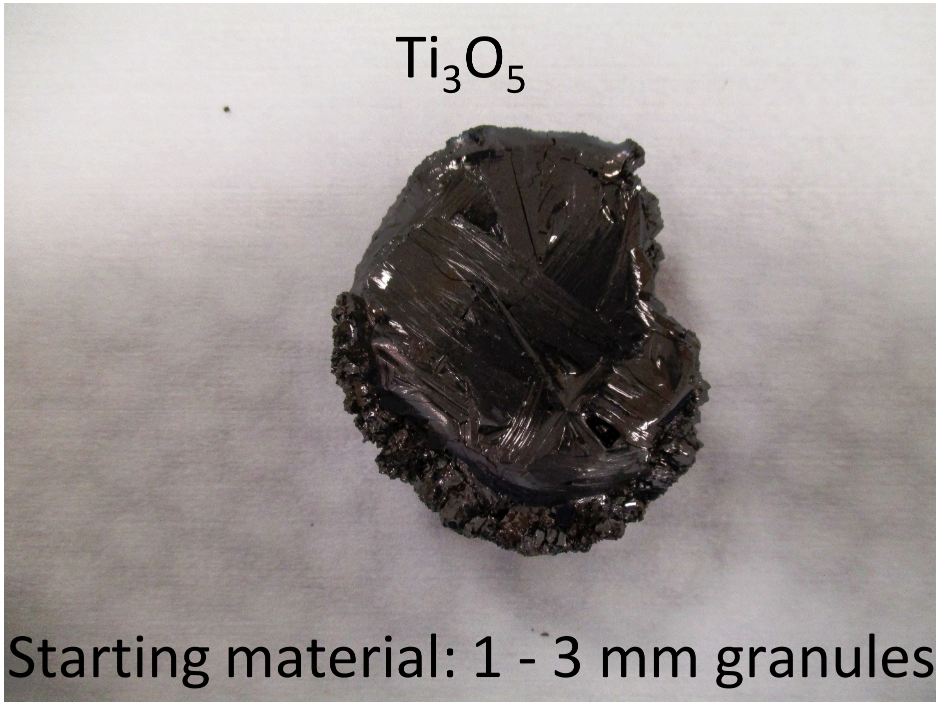
Layer thickness:
125 nm

Ti metal and Ti_3O_5 source materials



Ti metal

Starting material: 3 mm pellets



Ti_3O_5

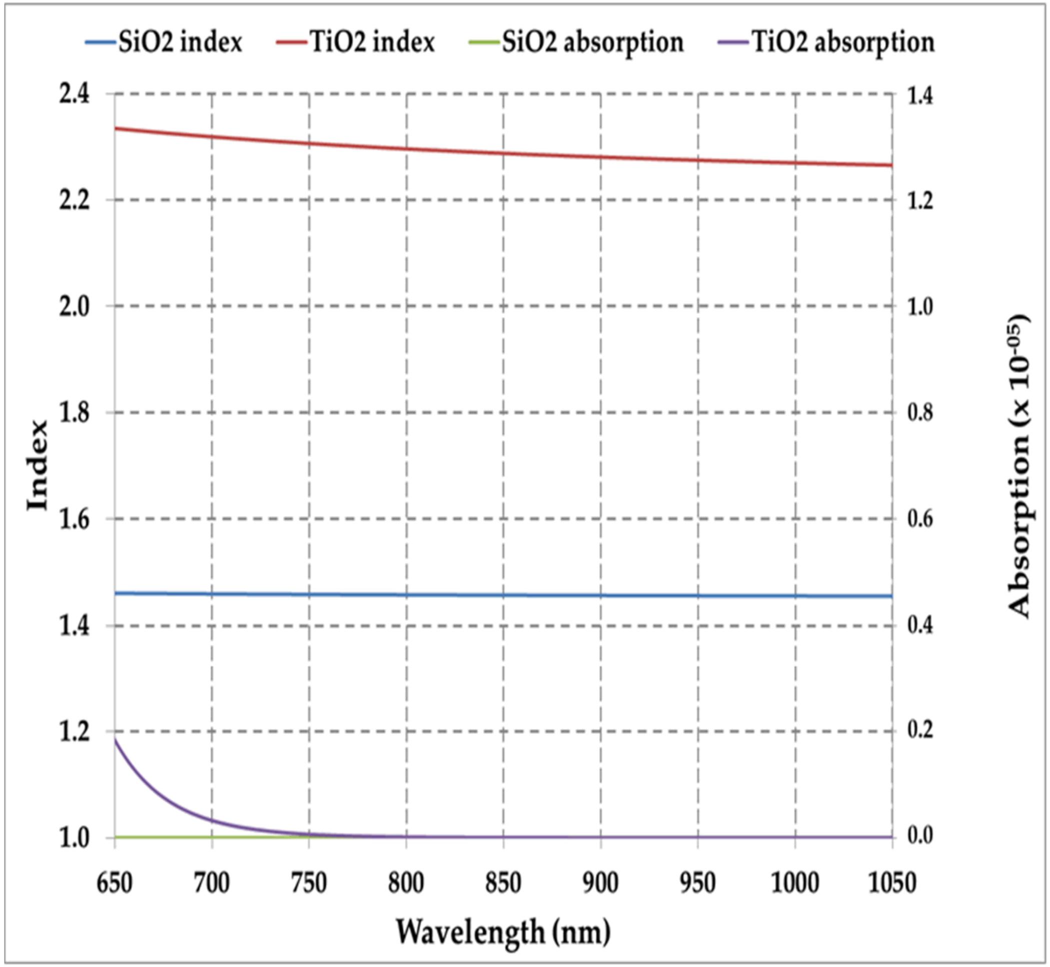
Starting material: 1 - 3 mm granules

Ti metal under e-beam heating develops a relatively thick, uniform melt on top and undergoes a stable melt-to-vapor process

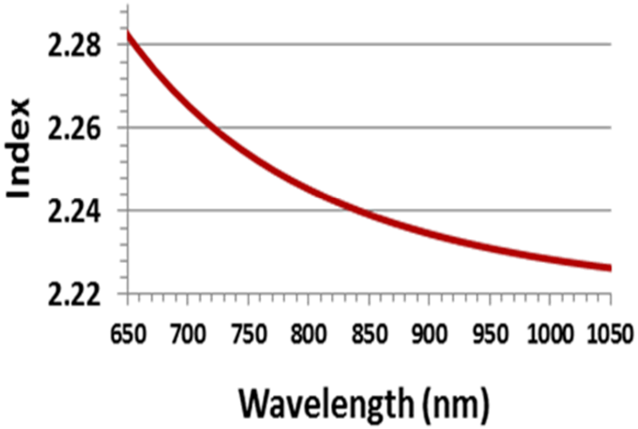
Ti_3O_5 under e-beam heating develops a thin, semi-solid, randomly structured melt on top

- Higher e-beam currents lead to unstable deposition characterized by spitting of oxide clusters
- This makes high deposition rates unfavorable for Ti_3O_5

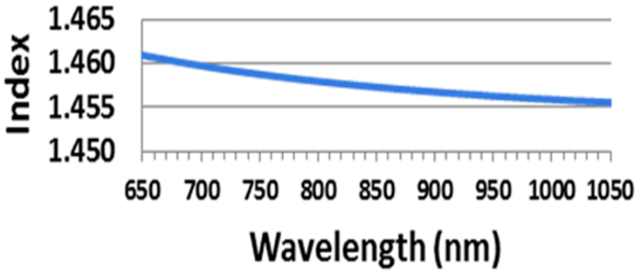
High and low index layer choices for BBHR coating: TiO_2 with Ti as source material, and SiO_2



TiO_2



SiO_2



Sandia BBHR coating design for fs-pulse high energy lasers
(Work in collaboration with and supported by Rutherford Appleton Laboratory, UK)

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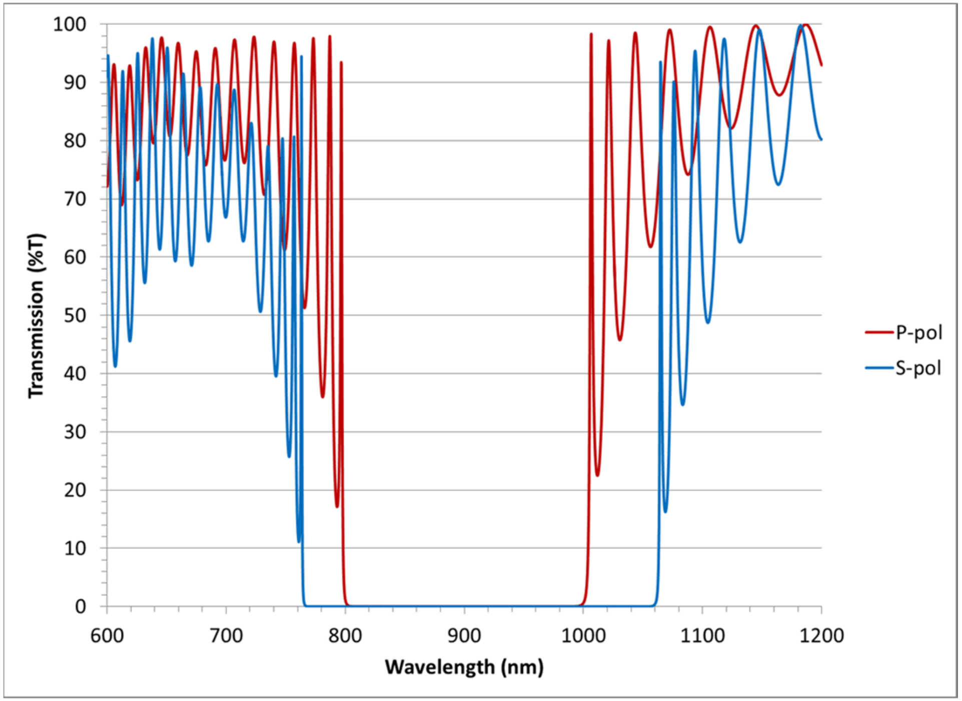
BBHR Coating Design Goals:

- Reflectivity → R > 99.5% for 45^o AOI, Ppol
- Operational Bandwidth → 800 – 1000 nm (900 nm line center)
- LIDT → > 800 mJ/cm^2 for fs class pulses
- Group Delay Dispersion (GDD) → < 20 fs^2 over the operational bandwidth

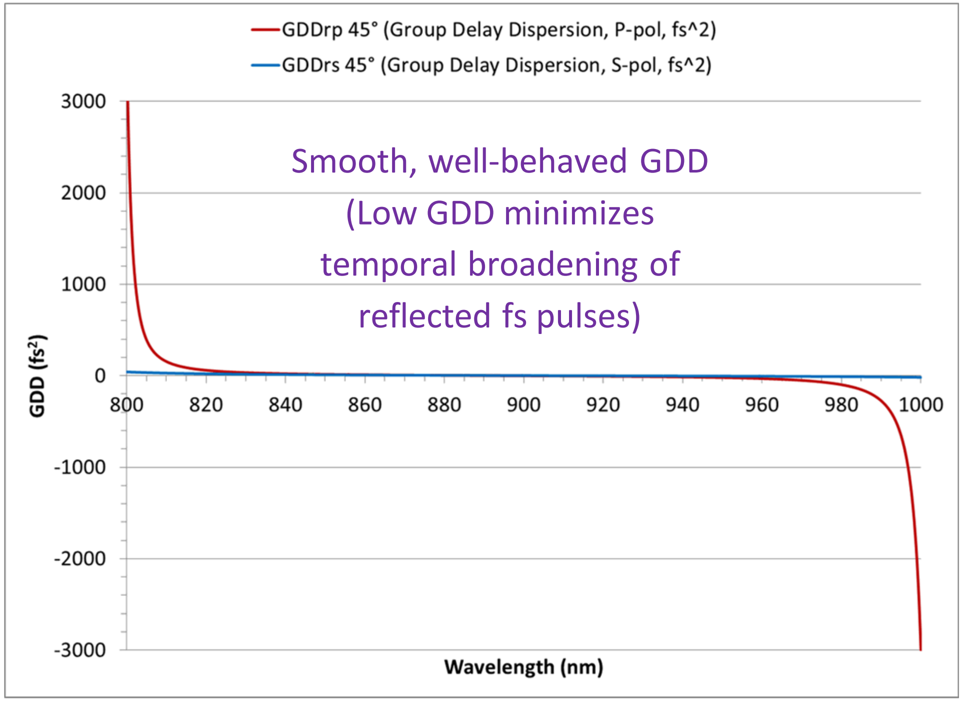
We designed and produced an all-dielectric multilayer, quarter wave type BBHR coating

- based on TiO_2/SiO_2 layer pairs

- based on opposite (reversed) chirped TiO_2 versus SiO_2 layer thicknesses from outer to inner layers for low GDD



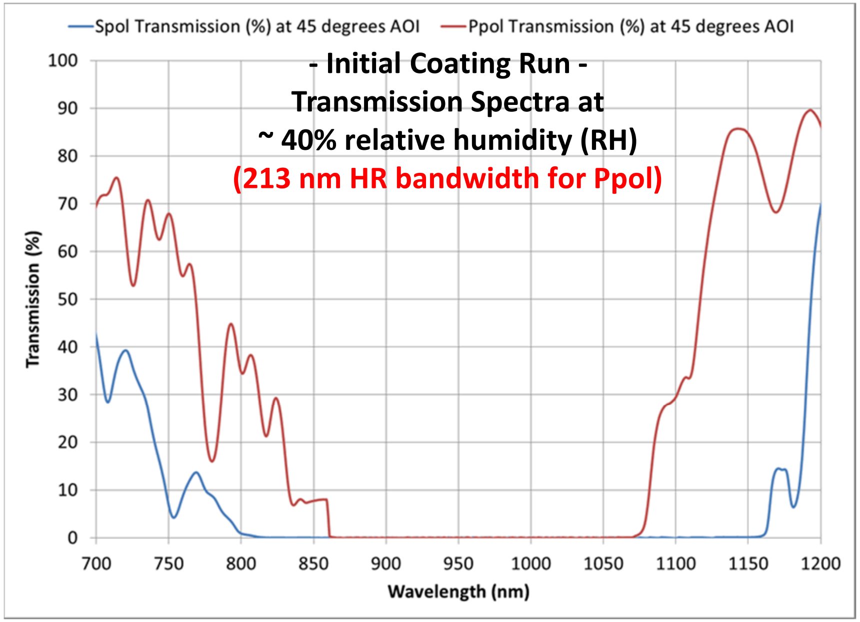
Transmission at 45^o AOI, Ppol and S pol, for the fs-pulse BBHR coating design



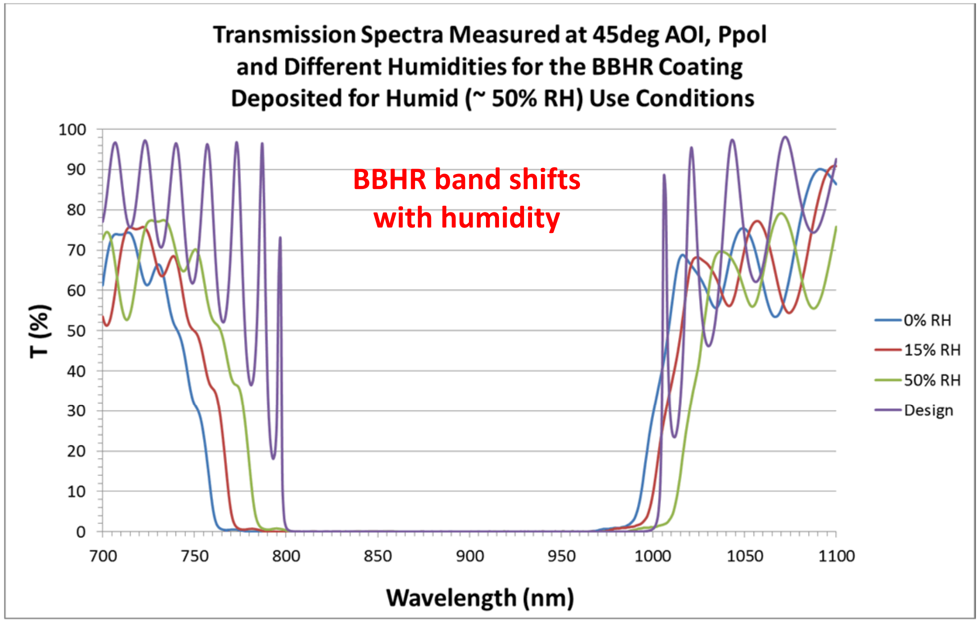
Smooth, well-behaved GDD (Low GDD minimizes temporal broadening of reflected fs pulses)

GDD on reflection for 45^o AOI, S and P pol, from 800 nm to 1000 nm for the fs-pulse BBHR coating design

Results from the initial set of coating runs for Sandia’s BBHR coating design for fs-pulse high energy lasers

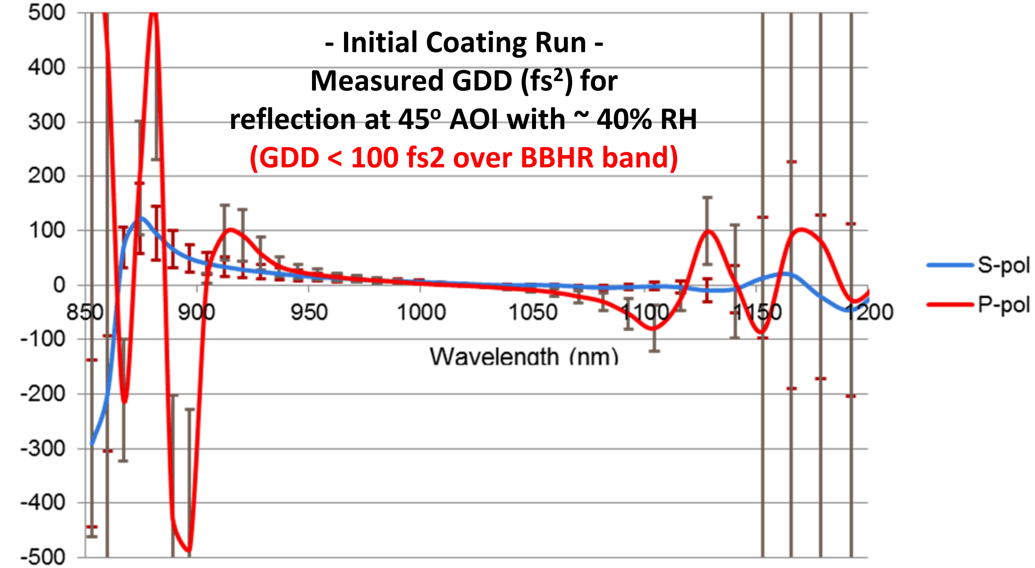


- Initial Coating Run -
Transmission Spectra at
~ 40% relative humidity (RH)
(213 nm HR bandwidth for Ppol)



Transmission Spectra Measured at 45deg AOI, Ppol and Different Humidities for the BBHR Coating Deposited for Humid (~ 50% RH) Use Conditions

BBHR band shifts with humidity



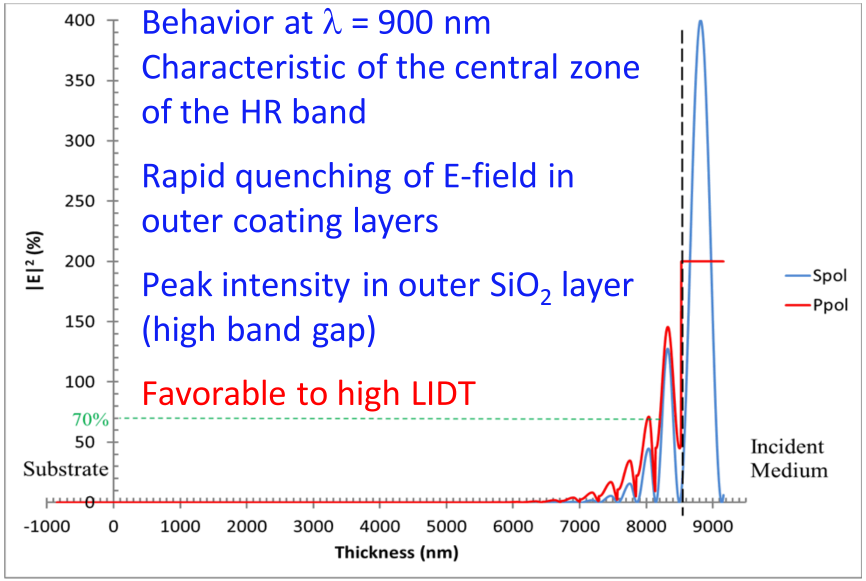
- Initial Coating Run -
Measured GDD (fs²) for reflection at 45° AOI with ~ 40% RH
(GDD < 100 fs2 over BBHR band)

Optical E-field behaviors for the BBHR coating design

Slow quenching of E-field deep into coating layers

Higher intensity maxima ($<180\%$) occur in SiO_2 layers (high band gap)

Favorable to high LIDT



Behavior at λ = 900 nm
Characteristic of the central zone of the HR band

Rapid quenching of E-field in outer coating layers

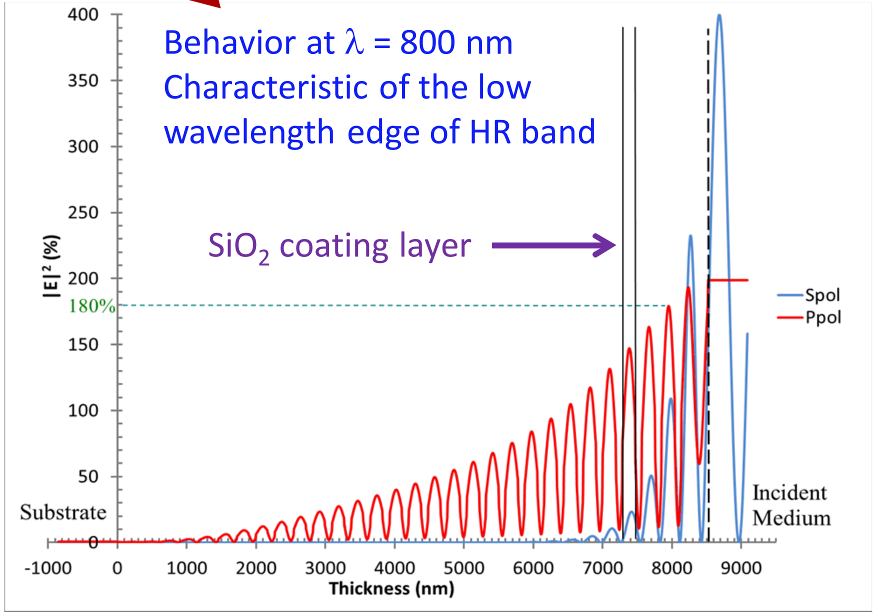
Peak intensity in outer SiO_2 layer (high band gap)

Favorable to high LIDT

Slow quenching of E-field deep into coating layers

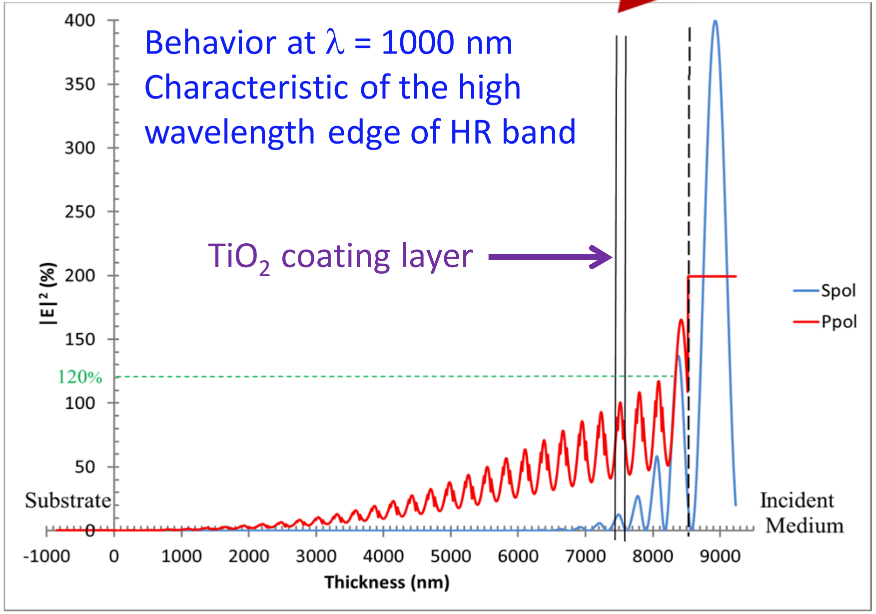
Lower intensity maxima ($<120\%$) occur in TiO_2 layers (low band gap)

Favorable to high LIDT



Behavior at λ = 800 nm
Characteristic of the low wavelength edge of HR band

SiO_2 coating layer →



Behavior at λ = 1000 nm
Characteristic of the high wavelength edge of HR band

TiO_2 coating layer →

The vertical dashed lines indicate the coating boundary with the incident medium

LIDT test results for Sandia TiO_2/SiO_2 BBHR coatings

Coating	Test Center Wavelength & BBHR Band	Test Pulse Duration	Test AOI	Test Protocol	LIDT (J/cm²)	LIDT Reference
Coating 1	1064 nm 940 – 1171 nm	3.5 ns	45°	NIF-MEL	**28**	[18]
Coating 1	1064 nm 940 – 1171 nm	10 ns	45°	ISO 11254-1 (1-on-1)	**17.5**	[18]
Coating 2	1064 nm 971 – 1203 nm	3.5 ns	45°	NIF-MEL	**19**	[19]
Coating 2	1064 nm 971 – 1203 nm	10 ns	45°	ISO 11254-1 (1-on-1)	**12.7**	[19]
Run 071	1064 nm 819 – 1108 nm	800 ps	0°	NIF-MEL	**11**	[15]
Run 071	1064 nm 819 – 1108 nm	8 ps	0°	NIF-MEL	**1.25**	[15]
Run 071	1064 nm 808 – 1097 nm	800 ps	19°	NIF-MEL	**9**	[15]
Run 071	1064 nm 808 – 1097 nm	8 ps	19°	NIF-MEL	**1.5**	[15]
Run 072	1053 nm 866 – 1169 nm	675 fs	0°	ISO 11254-1 (1-on-1)	**1.34**	[15]
Run 072	1053 nm 866 – 1169 nm	675 fs	0°	ISO 11254-2 (10-on-1)	**1.15**	[15]
Run 072	1053 nm 818 – 1060 nm	675 fs	40°	ISO 11254-1 (1-on-1)	**1.15**	[15]
Run 072	1053 nm 818 – 1060 nm	675 fs	40°	ISO 11254-2 (10-on-1)	**0.8**	[15]

Test Polarization

Ppol for non-normal AOI

NIF-MEL Protocol

Dense raster scan method

Damage definition: propagating damage, or 25 or more non-propagating damage sites

- By Spica Technologies, Inc.

ISO 11254 Protocols

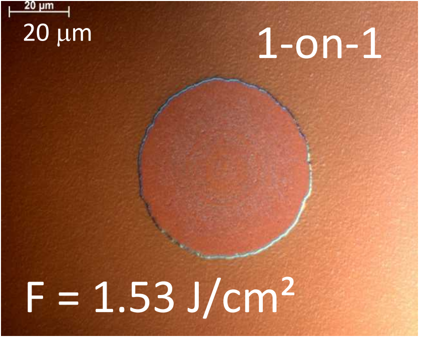
damage frequency method, single shot (1-on-1) or 10-shot (10-on-1)

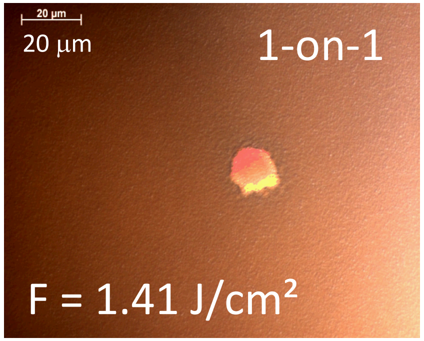
Damage definition: permanent surface change

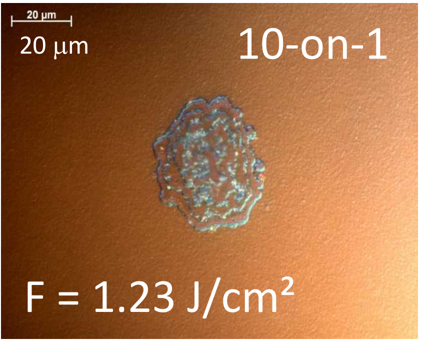
- By Quantel USA (Coatings 1 & 2) and CEA-CESTA, Le Barp, France

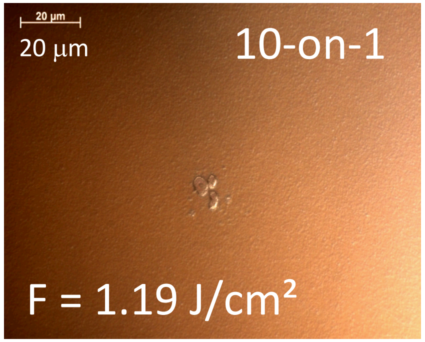
Morphology of 675-fs pulse laser-induced damage to the TiO_2/SiO_2 BBHR coating of Run 072

Damage for 0° AOI





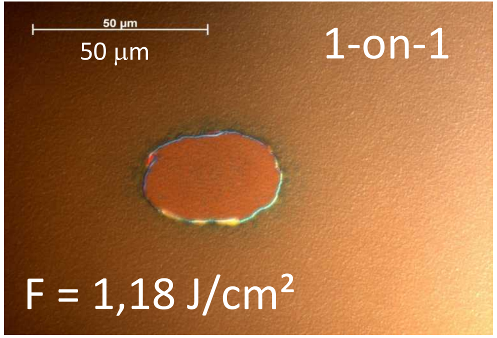


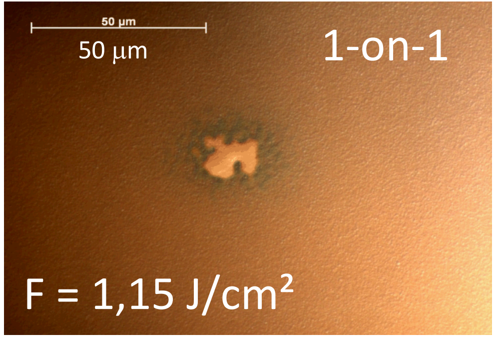


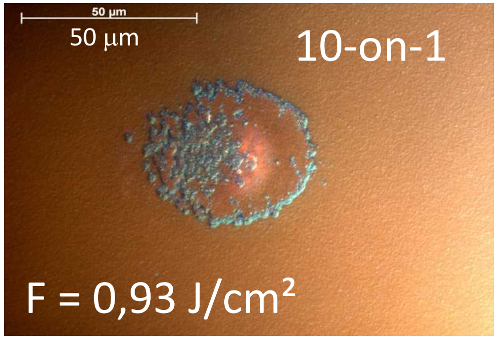
Fluences for 100% damage frequency

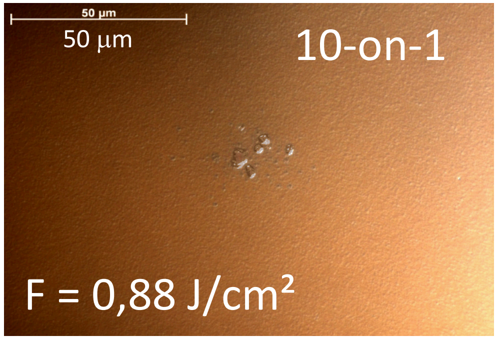
Fluences slightly above LIDTs

Damage for 40° AOI, Ppol









Fluences for 100% damage frequency

Fluences slightly above LIDTs

Abrupt transition from LIDT fluences to fluences for 100% damage frequency indicates intrinsic damage

Fatigue effect for 10-on-1 damage – successive laser shots create electronic defects that lower intrinsic LIDTs

All damage morphologies indicate delamination of the outer coating layers

LIDT tests and morphology images by CEA-CESTA, France

Conclusions

- Sandia successfully produces uniformly thick, high LIDT nano-scale thin films on meter-scale optics for high energy lasers
- TiO_2/SiO_2 layer pairs are an excellent choice for BBHR coatings because of their strong high/low index contrast
- Sandia has developed e-beam evaporation deposition processes to produce optimally high index TiO_2 thin films
- BBHR coatings must afford low GDD despite large index variations over broad HR bands
  - to reflect fs laser pulses without degradation of their temporal profiles or durations
- Sandia's BBHR coating design based on reverse-chirped thickness TiO_2/SiO_2 layer pairs
  - achieves a 200 nm HR bandwidth centered at 900 nm
  - exhibits low GDD, and E-field behaviors favorable to high LIDTs
- Sandia's TiO_2/SiO_2 BBHR coatings as deposited
  - meet HR bandwidth goals
  - show promise of low GDD
  - are characterized by reasonably high LIDTs from the ns to fs laser pulse regimes