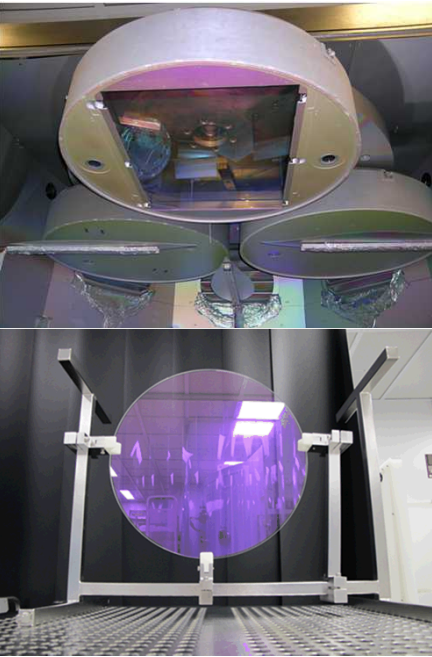


Broad Bandwidth Coating Development: Coating Design Report

SAND2014-15551PE



Submitted to Central Laser Facility,
Rutherford Appleton Laboratory, Science
and Technology Facilities Council of the
United Kingdom, July 2014



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John Bellum and Ella Field

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1. Introduction

This report is a deliverable of Task 1 of Sandia Project # 174545 titled “Broad Bandwidth Coating Development,” which is a Work for Others contract (Agreement FI016121017) between Sandia National Laboratories and the Science and Technology Facilities Council of the United Kingdom. It describes the coating modelling and design development for a **broad bandwidth high reflection (BBHR)** coating that meets specifications suitable for high laser-induced damage threshold (LIDT) mirrors for fs-class laser pulses at the Rutherford Appleton Laboratory, and that can eventually be produced on meter-class optical substrates.

The report is organized as follows:

- Section 2: Design Goals
- Section 3: Coating Design and Production Considerations
- Section 4: Proposed Design
- Section 5: Summary
- References

2. Design Goals

- Table 1 summarizes Rutherford Appleton Laboratory's specification goals for the BBHR optical coating.

Table 1: Design Goals of the BBHR Optical Coating	
Item:	Specification:
Reflectivity	high reflection (> 99.5%)
Angle of Incidence (AOI)	45 degrees
Polarization	P pol and S pol (separate, not averaged)
Operational Bandwidth	800 - 1000 nm
LIDT Requirement	> 800 mJ/cm ² (surface) at 40 fs and > 5 J/cm ² (surface) at 3 ns under vacuum of 10 ⁻⁶ mbar
Group Velocity Dispersion	Second order < 20 fs ² over the operational bandwidth Third order < 100 fs ³ over the operational bandwidth

3. Coating Design and Production Considerations

In this section, we discuss our strategy towards designing a BBHR coating that satisfies the challenging goals of Table 1.

Meeting the 800 – 1000 nm operational bandwidth specification, especially for the case of P pol, depends on having a sufficiently large refractive index contrast between the alternating high and low index coating layers. We have addressed this demand by developing the processes for depositing high index layers of TiO_2 , Ta_2O_5 and Nb_2O_5 using Sandia's large optics coater, and have produced 42-layer HR coatings for 45° AOI, P pol at a central wavelength of 1054 nm that have bandwidths greater than 200 nm [1, 2]. **TiO_2 offers the highest refractive indices of the layers that we have deposited.**

We have produced TiO_2 layers by means of e-beam evaporation of either Ti metal or Ti_3O_5 under reactive, ion-assisted deposition (IAD) conditions using a back pressure of oxygen in the coating chamber. We deposit low index layers by means of e-beam evaporation of SiO_2 under IAD conditions. IAD leads to denser coating layers with less stress mismatch to fused silica optical substrates. This in turn makes the coatings very mechanically stable and resistant to delamination in vacuum such as the level of vacuum mentioned in Table 1. **None of our IAD HR coatings on large, fused silica optics have ever delaminated under vacuum conditions in Sandia's Z-Backlighter beam trains, and this gives us confidence that these same IAD coating processes will lead to equally delamination-free coatings based on our proposed BBHR design.**

3. Coating Design and Production Considerations, cont.

TiO₂ and SiO₂ Layer Models

For TiO₂, we have used the OptiChar application of the OptiLayer thin film design software to determine index and absorption properties from analysis of transmission spectra of single layers that we have deposited. The transmission spectra of SiO₂ single layers don't lend themselves to the OptiChar analysis because of the close match of the SiO₂ layer index to that of glass. So, we characterize the index and absorption of our SiO₂ single layers based on our experience over more than 10 years of depositing SiO₂. We find that the Cauchy relationship,

$$n(\lambda) = A_0 + A_1/\lambda^2 + A_2/\lambda^4$$

is an accurate description of the refractive index, indicated here by n , as a function of wavelength, λ , for both our SiO₂ single layers, as characterized by experience, and our TiO₂ single layers, as characterized by OptiChar analyses.

We base our BBHR coating design on TiO₂ layers evaporated from Ti metal as the starting material because this coating process is very well behaved without the occurrence of spit from the evaporation melt, and affords double the TiO₂ deposition rate compared to evaporation from Ti₃O₅ as the starting material. For such TiO₂ layers, we find $A_0 = 2.224101$, $A_1 = 0.044657$, and $A_2 = 8.2192065 \times 10^{-4}$ for the Cauchy parameters, and absorption that is close to negligible and well characterized in terms of an exponential decrease with increasing wavelength. For SiO₂ layers, we find $A_0 = 1.452228$, $A_1 = 0.003672198$, and $A_2 = 3.9035249 \times 10^{-8}$ for the Cauchy parameters, with absorption that is negligible. These indices of refraction and absorption behaviors are shown in Figs. 1 – 4 over 600 nm – 1200 nm and also over the 800 nm – 1000 nm operational bandwidth, and are the basis for our BBHR coating design.

Figure 1. Refractive index and absorption for TiO₂ layers. In this graph, $n = \text{Re}(n) + i\text{Im}(n)$ indicates the complex index of refraction where the refractive index is $\text{Re}(n)$ and the absorption coefficient is $4\pi[\text{Im}(n)]/\lambda$.

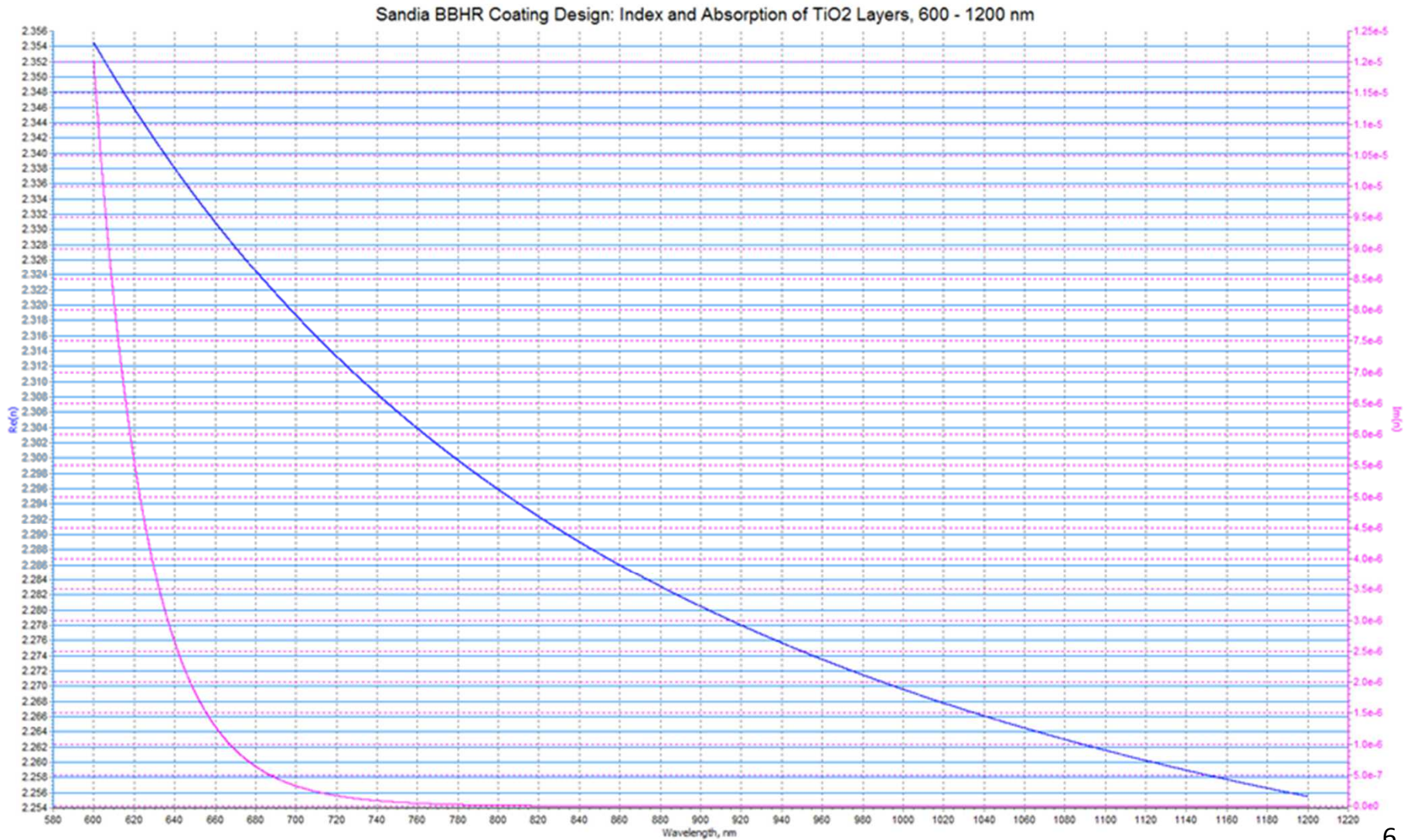


Figure 2. As Fig. 1, but for 800 – 1000 nm.

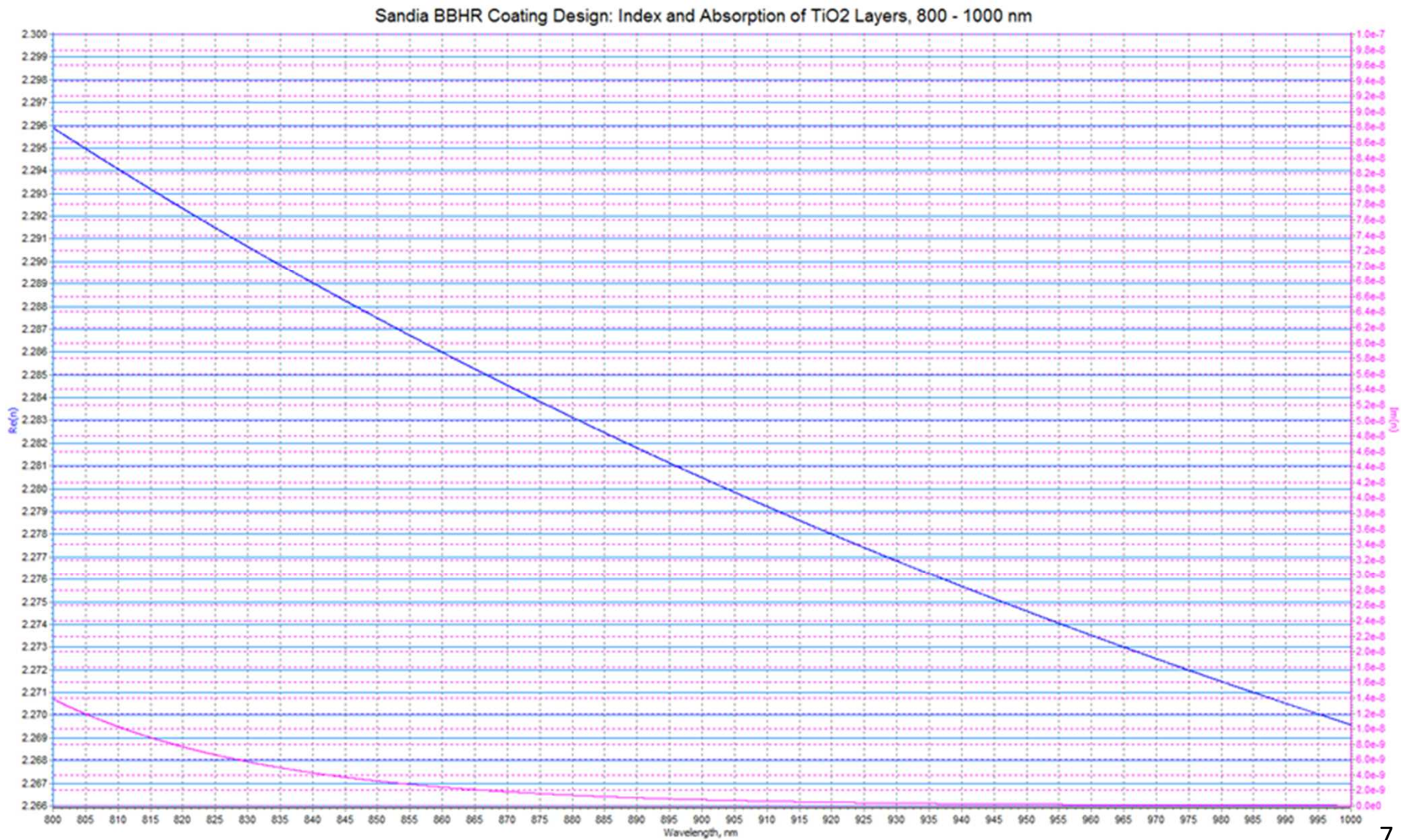


Figure 3. As Fig. 1, but for SiO₂ layers.

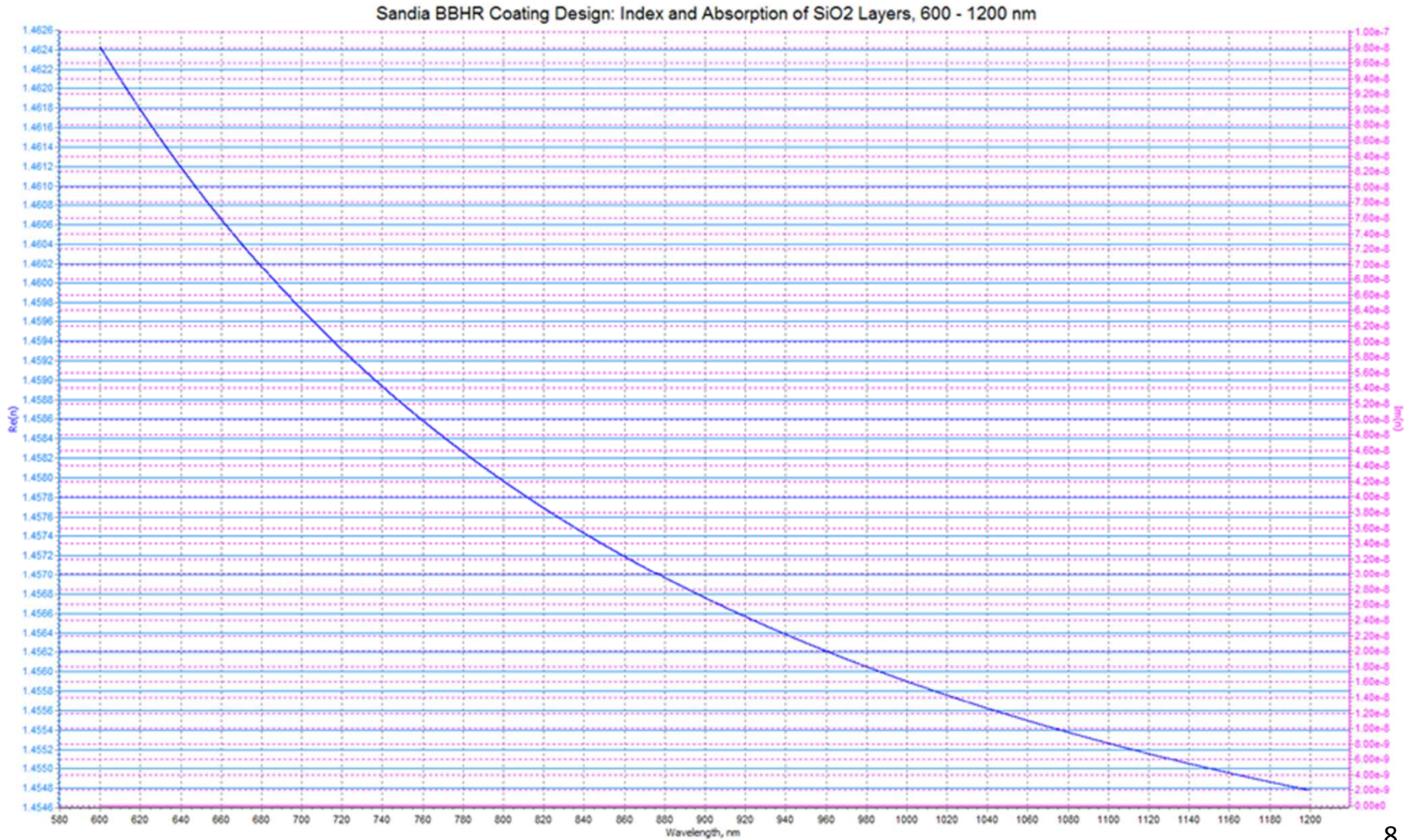
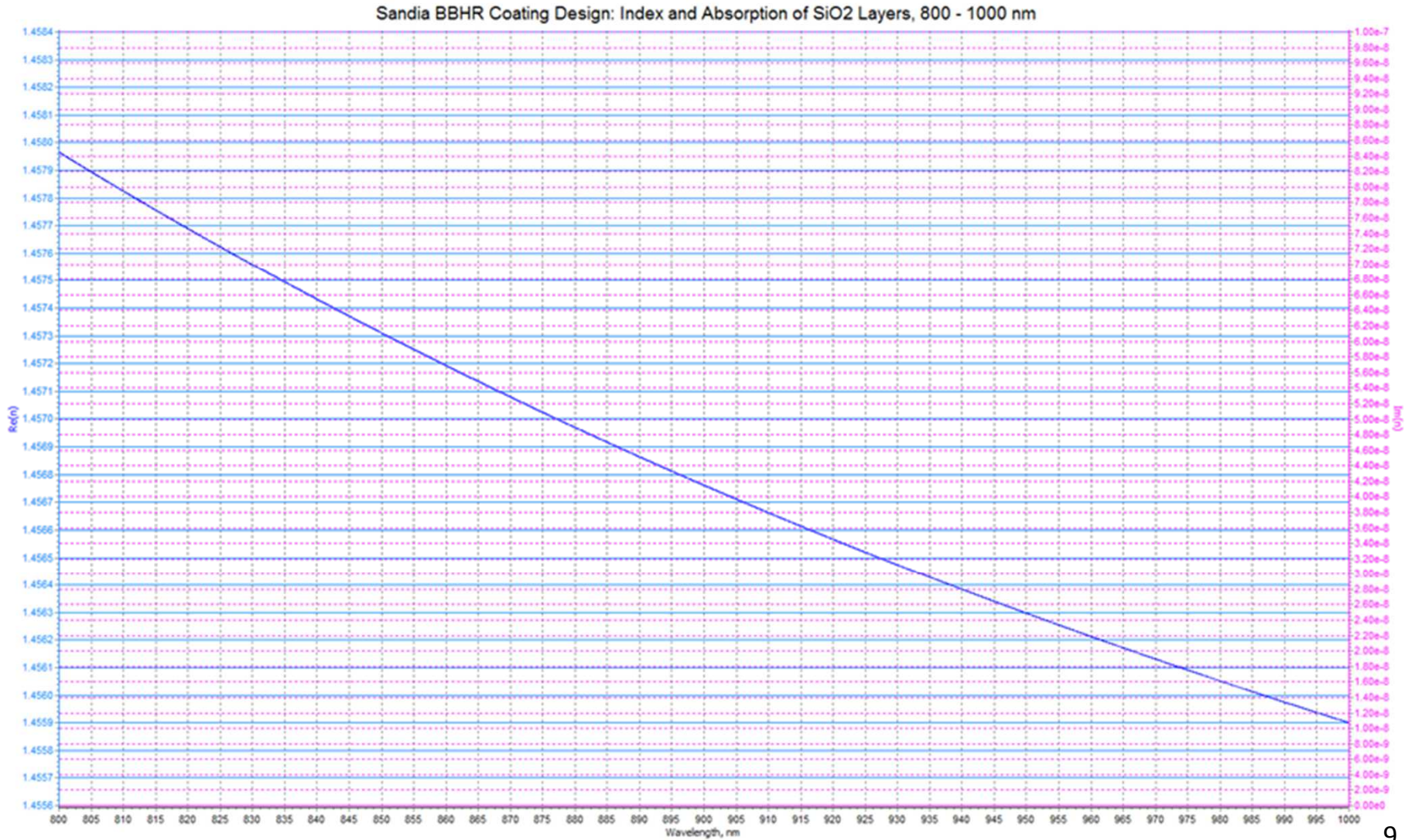


Figure 4. As Fig. 1, but for SiO₂ layers and 800 nm – 1000 nm.



3. Coating Design and Production Considerations, cont.

High Reflection and Group Delay Optimization

The design goal of $> 99.5\%$ reflectivity at 45° AOI in either S pol or P pol means that light of all frequencies in the operational bandwidth must propagate into and back out of the HR coating layers with no more than 0.5% loss of intensity. Quarter-wave stack designs offer the best prospects for such HR coatings. The wide variation of layer refractive indices over the broad operational bandwidth means, however, a requirement of an equally wide variation of quarter-wave layer thicknesses to accommodate the broad range of wavelengths and refractive indices. Meeting the BBHR requirement forces a competition between the wide range of effective quarter-wave layer thicknesses and of optical wave propagation velocities across the operational bandwidth. At the heart of this competition is the non-linear dependence on wavelength of the layer refractive indices (see Figs. 1 - 4), leading to group delay (GD) behavior that is non-linear with respect to wavelength, and to related deviation of the group delay dispersion (GDD) from being constant as a function of wavelength.

In any HR coating design, each coating layer plays a different role for each wavelength across the broad spectra of fs-class optical pulses as they propagate into and back out of the coating. For an optimal coating design, the differing layer roles combine to provide the broadest HR bandwidth while minimizing the non-linearity of GD and the associated non-constant behavior of GDD as a function of wavelength. The BBHR coating design challenge is to achieve an optimal compromise between the high and low index quarter-wave layer thicknesses in order to meet the specification goals as closely as possible. This means that the negative impact of deviations from the mirror's performance specifications should not only be minimal but also acceptable or, even better, correctable. **In this regard, we consider in our BBHR design relaxing the reflectivity requirement from $> 99.5\%$ to $> 99\%$. We also consider GD that varies as nearly linearly and smoothly as possible with wavelength, and GDD that is close to being $< 20 \text{ fs}^2$ in magnitude and in addition varies smoothly over the broad operational bandwidth.**

3. Coating Design and Production Considerations, cont.

Laser Damage Considerations

The coating materials TiO₂ and SiO₂ exhibit some of the highest LIDTs. Our TiO₂/SiO₂ coatings for broad bandwidth HR at 45° AOI, P pol with 1054 nm central wavelength exhibit a LIDT under use conditions at 1064 nm of 19 J/cm² according to the NIF-MEL protocol with 3.5 ns pulses and 12.7 J/cm² according to the ISO 11254-1 protocol with 7 ns pulses [1]. LIDTs with fs pulses will be much lower and more dependent on intrinsic, band-gap related material properties. Consequently, TiO₂ will be more vulnerable than SiO₂ to laser damage since its band gap is considerably less than that of SiO₂. Therefore, an optimal coating design with respect to LIDT for fs pulses will be one for which the strongest electric field intensity peaks in the coating occur within SiO₂ layers. We take this into account in our BBHR coating design.

Coating Uniformity

A final design consideration is how well the designed coating lends itself to being produced uniformly on large, meter-class optics. We have gained considerable experience in meeting the large optics coating production challenges due to both increasing number of coating layers and increasing substrate dimension [3], and routinely deposit coatings of > 30 thin film layers uniformly on meter-class optical substrates. This is possible because of the 2.4 m x 2.4 m x 1.8 m size the Sandia large optics coater, and the combination of e-beam evaporation with IAD, planetary motion of the large optics during deposition, and specially designed masking just below the coating plane. **We consistently produce multi-layer coatings with +/- 0.5 % uniformity over 94 cm diameters of our planetary fixtures, and are confident we can produce the BBHR coating according to our proposed design on a large optic with similar uniformity.**

4. Proposed Design

Layer Thickness Optimization

For our high and low index layers (see Figs. 1 - 4), the physical thicknesses that match quarter-wave optical thicknesses within the 800 nm – 1000 nm operational bandwidth are from 91.56 nm to 115.92 nm in the case of TiO_2 layers and from 156.86 nm to 196.44 nm in the case of SiO_2 layers. We used the OptiLayer thin film design software and its numerical algorithms that optimize designs based on targeted performance properties such as HR and GDD to explore designs. A given algorithm can lead to different optimal designs depending on the starting design and performance targets. Our starting designs were various numbers of high/low index layer pairs and various combinations of layer thicknesses that lie within the ranges for our TiO_2 and SiO_2 quarter-wave layers over the 800 nm – 1000 nm band.

We arrived at the proposed design with a limited optimization process starting from 33 high/low index layer pairs in which the $\text{TiO}_2/\text{SiO}_2$ layer thicknesses increased/decreased linearly from the outermost to innermost layer pairs. The extent of linear thickness increase/decrease matched the extent of thicknesses for quarter-wave layers over the 800 nm – 1000 nm operational band, with the $\text{TiO}_2/\text{SiO}_2$ layer thicknesses accordingly at 91.56 nm/196.44 nm for the outermost layer pair and at 115.92 nm/156.86 nm for the innermost layer pair. We further modified this starting design by doubling the thickness of the outermost SiO_2 layer, making it a half-wave for 1000 nm wavelength light. That modification ensures that the highest peak electric field intensities over the HR bandwidth occur in this outermost half-wave or near half-wave SiO_2 layer. This is a condition that favors enhanced laser damage resistance of the coating since SiO_2 , with its higher band gap, is more resistant to laser damage by fs-class laser pulses than TiO_2 .

4. Proposed Design, cont.

The overall combination of quarter-wave layer thicknesses with the high and low index layers arranged in opposite ramp, or “chirped,” progressions proved to provide the best initial mix of quarter-wave layer roles capable of accommodating the full 800 nm – 1000 nm range of wavelengths in a balanced way. The limited optimization from this 33 layer-pair starting design resulted in our proposed 30 layer-pair design, which has layer thicknesses that are slightly modified compared to the starting thicknesses but, aside from a few exceptions, do not strongly deviate from an opposite “chirp” layer thickness behavior. The limited optimization led to improved GD and GDD behaviors without compromising the HR performance or prospects for a high LIDT.

The P and S pol transmission spectra at 45° AOI for our proposed BBHR coating design are shown by Fig. 5 for the 600 nm to 1200 nm spectral range, and by Fig. 6 for the 800 nm – 1000 nm operational band. The transmission spectra are well-behaved.

- The bandwidth for P pol reflectivity exceeding 99% is 200.29 nm (from 800.34 nm to 1000.63 nm) and the bandwidth for P pol reflectivity exceeding 99.5% is 198.02 nm (from 801.22 nm to 999.24 nm).
- The bandwidth for S pol reflectivity exceeding 99% is 295.88 nm (from 765.23 nm to 1061.11 nm) and the bandwidth for S pol reflectivity exceeding 99.5% is 294.30 nm (from 765.64 nm to 1059.94 nm).

Figure 5. Transmission at 45° AOI, P and S pol, from 600 nm to 1200 nm for the Sandia BBHR coating design.

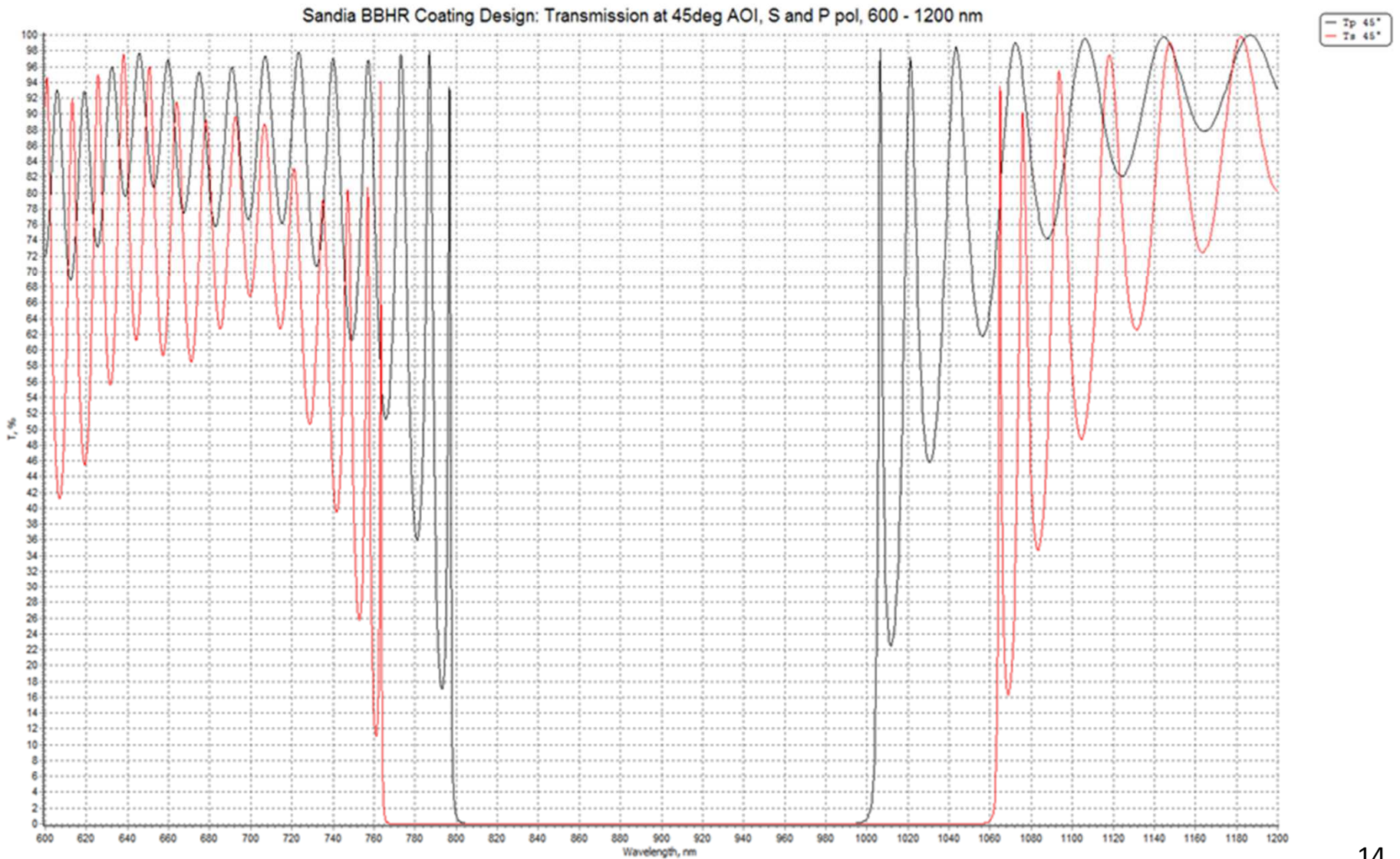
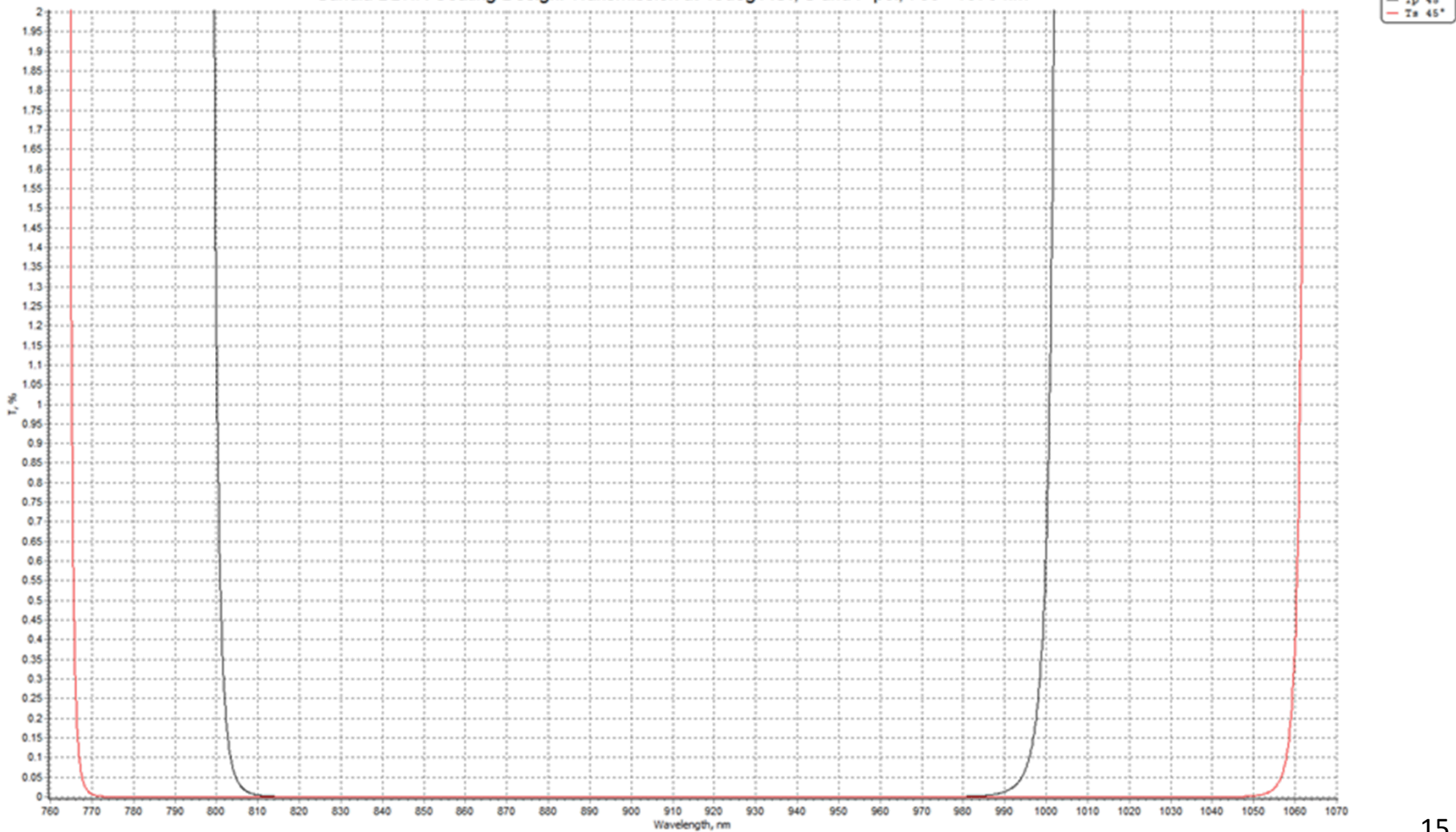


Figure 6. As Fig. 5, but from 760 nm to 1070 nm.

Sandia BBHR Coating Design: Transmission at 45deg AOI, S and P pol, 760 - 1070 nm



4. Proposed Design, cont.

GD and GDD Behavior

Figure 7 shows P and S pol GDs over the operational bandwidth for reflection at 45° AOI from our proposed BBHR coating. The GD variation with wavelength is very smooth overall and fairly flat, with the P pol GD between ~ 6 fs and ~ 10 fs in the interval of 816 nm to 982 nm and the S pol GD between ~ 3 fs and ~ 7 fs over the entire operational band. Beyond the 816 – 982 nm spectral range for P pol, the GD rises smoothly to ~ 35 fs at 800 nm and to ~ 30 fs at 1000 nm. The corresponding GDDs are shown in Fig. 8 and on an expanded scale in Fig. 9. The GDDs are definitely not constant with wavelength but are within ± 20 fs² from 843 nm to 949 nm for P pol and from 822 nm to 1000 nm and beyond for S pol, and are within ± 50 fs² from 823 nm to 969 nm for P pol and over the entire operational bandwidth and beyond for S pol. GDD rises smoothly in magnitude as wavelengths approach the low and high wavelength extremes of the operational band. For P pol, GDD rises from 50 fs² at 823 nm to ~ 3500 fs² at 800 nm, and drops from -50 fs² at 970 nm to -3500 fs² at 1000 nm. The S and P pol 3rd order behaviors of the GDD are shown in Fig. 10. They are always negative and are well behaved. For P pol, the magnitudes are < 100 fs³ over nearly the entire operational band, from 804 nm to 993 nm, and rapidly but smoothly rise outside this band to ~ 2000 fs³ at 1000 nm and at 800 nm. For S pol, the magnitudes are all < 1 fs³ over the entire operational band. Because of this smooth variation with wavelength of GD, GDD and the 3rd order of GDD, the prospect of being able to compensate for large GDD (such as > 20 fs² or > 50 fs²) where it occurs over the operational band is promising.

Figure 7. Group delay on reflection for 45° AOI, S and P pol, from 800 nm to 1000 nm for the Sandia BBHR coating design.

Sandia BBHR Coating Design: Group Delay for 45deg AOI, S and P pol, 800 - 1000 nm

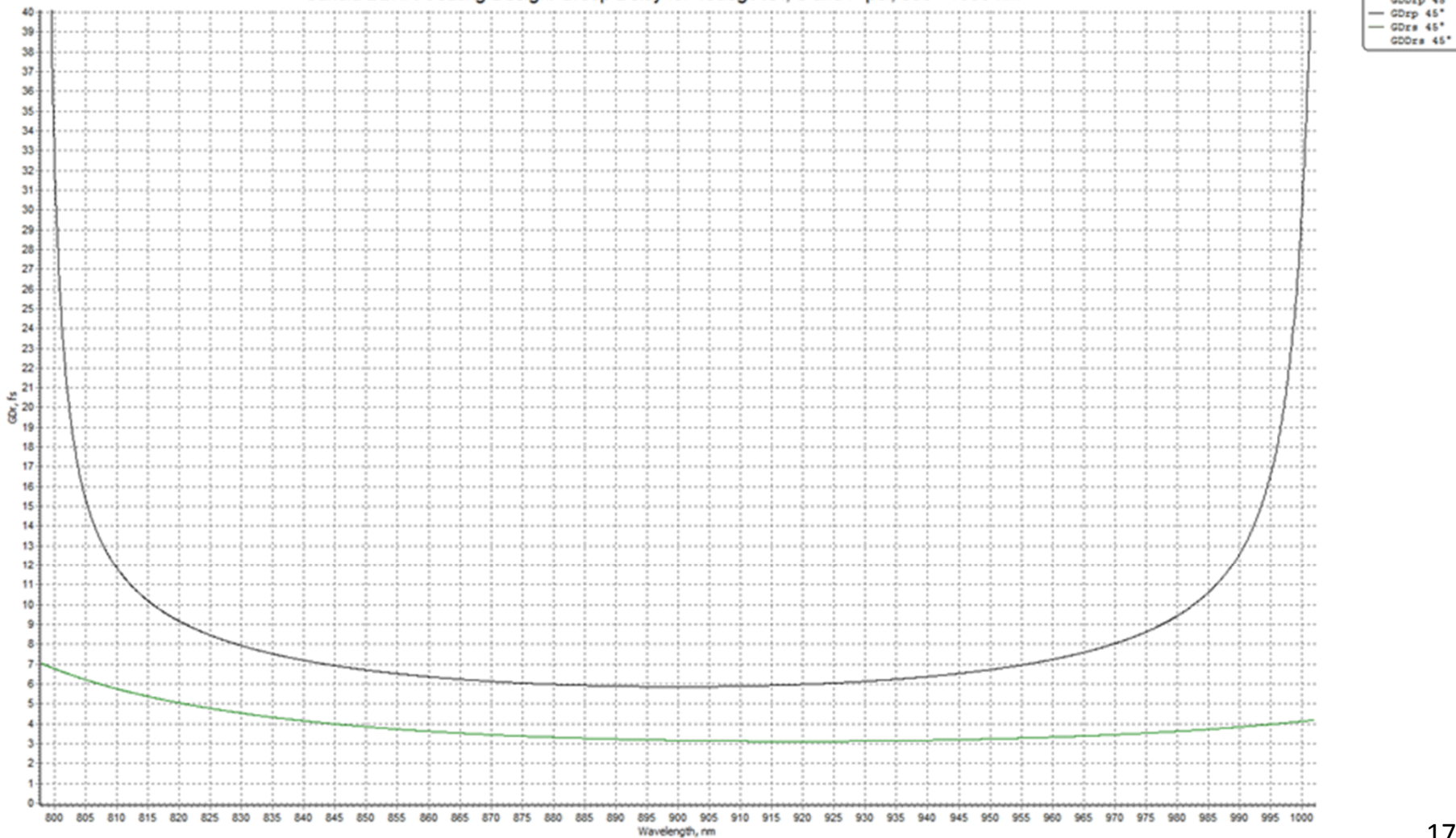


Figure 8. Group delay dispersion on reflection for 45° AOI, S and P pol, from 800 nm to 1000 nm for the Sandia BBHR coating design.

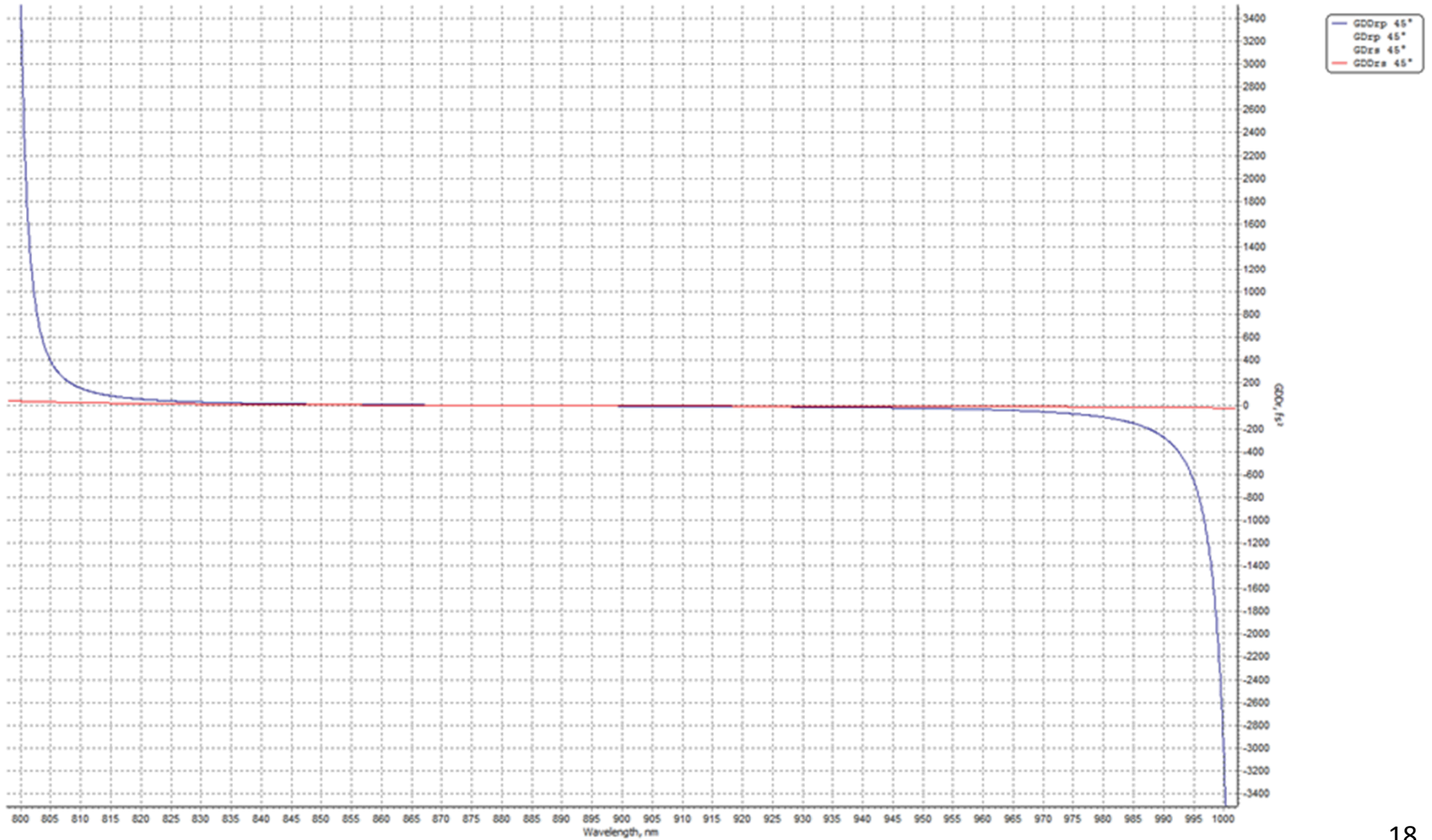


Figure 9. Group delay dispersion on reflection for 45° AOI, S and P pol, from 800 nm to 1000 nm and between + 60 fs² and - 60 fs² for the Sandia BBHR coating design.

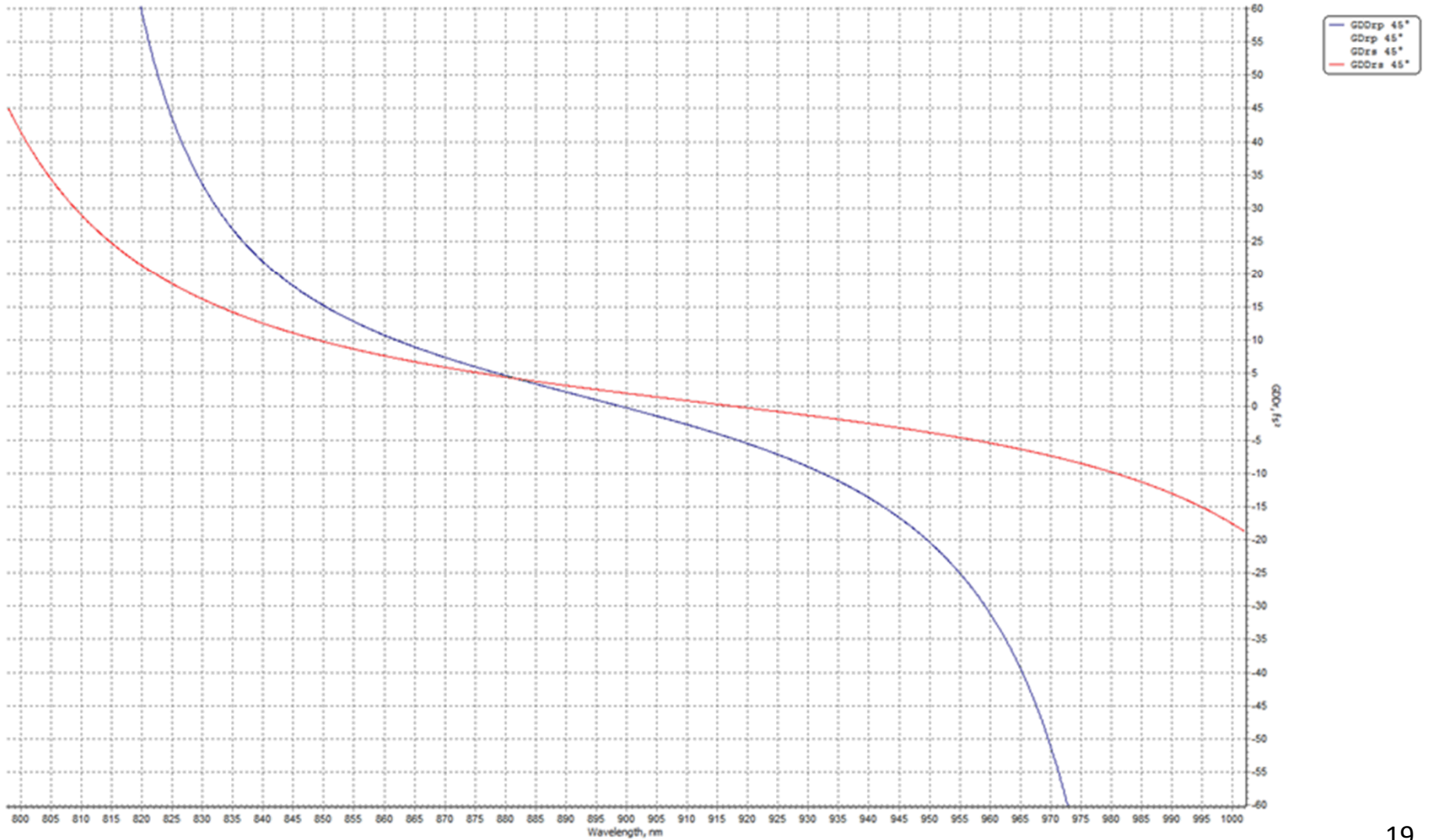
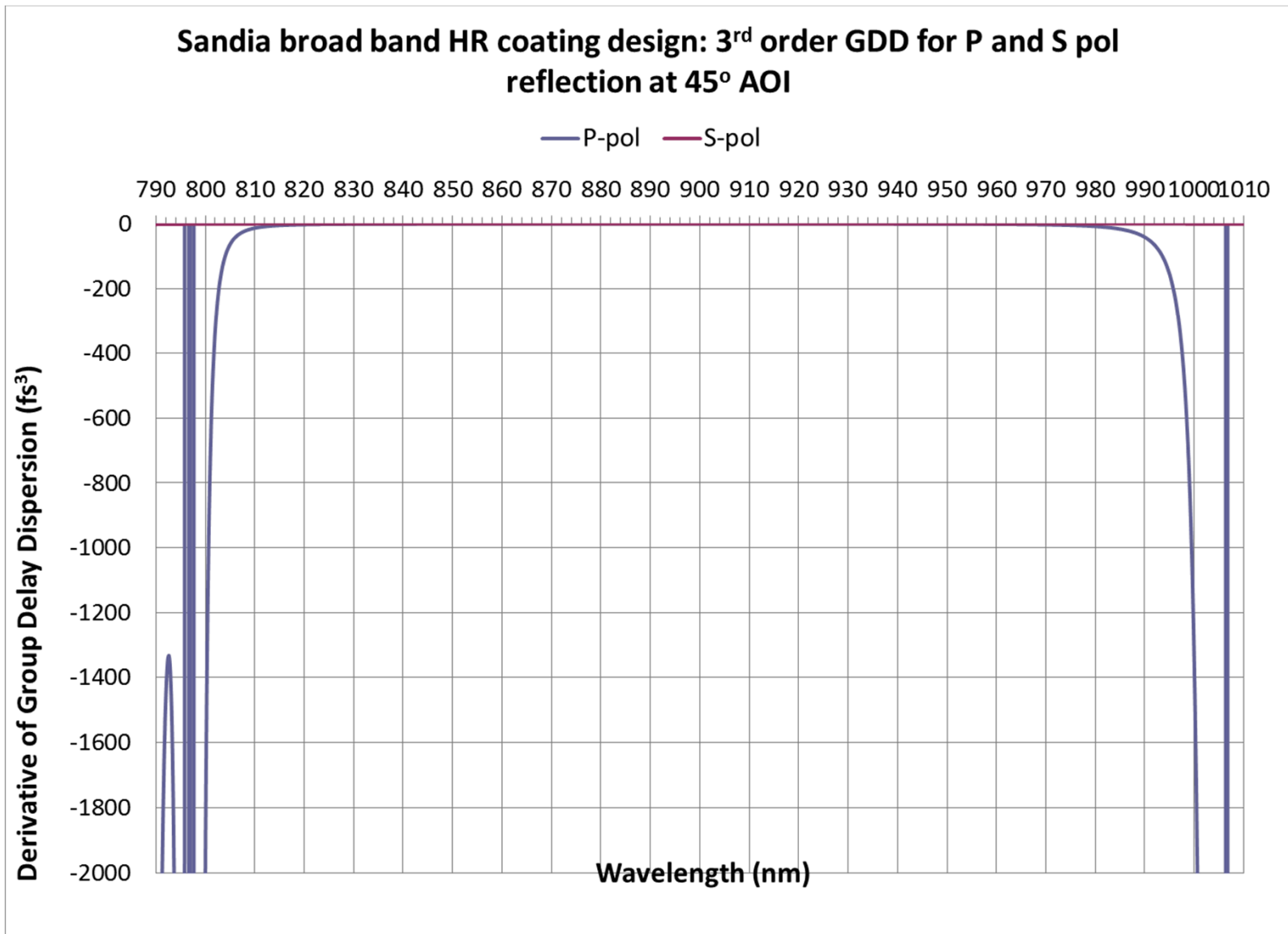


Figure 10. 3rd order of group delay dispersion on reflection for 45° AOI, S and P pol, from 800 nm to 1000 nm for the Sandia BBHR coating design.



4. Proposed Design, cont.

Electric Field Behavior

The electric field intensity peaks for both S and P pol are highest in the outer thick SiO₂ layer for all operational wavelengths, and rapidly quench within the outer coating layers to very low values. This quenching leads to intensity peaks that are < 20% of the incident intensity within the outer ~ 12 layers over the entire operational band for S pol and over most of the operational band, from ~ 810 nm to ~ 990 nm, for P pol. For (S pol/P pol), the maximum intensity peaks in the outer thick (~ half-wave) SiO₂ layer are less than about (210%/180%) and as low as about (120%/140%) of the incident intensity from ~ 810 nm to 1000 nm. The S and P pol electric field intensity behaviors in the coating for the ~ 810 nm to 1000 nm spectral range are similar to those shown in Fig. 11 for a wavelength of 900 nm. From 810 nm down to 800 nm, the highest peak intensities in the outer thick SiO₂ layer increase for both S and P pol, and, for these wavelengths as well as for 990 – 1000 nm wavelengths, the quenching of the intensity peaks becomes much more gradual for P pol. As shown in Fig. 12 for 1000 nm light, the P pol intensity peaks quench down to < 20% of the incident intensity over ~ 30 outermost layers. For 800 nm wavelength light, Fig. 13 shows that the outer layer peak intensity is ~ 190% of the incident intensity for P pol and at ~ 230% of the incident intensity for S pol, and the intensity peaks quench down to < 20% of the incident intensity over ~ 39 outermost layers for P pol. The LIDT impact of this higher peak intensity behavior for both S and P pol in the outer layer and for P pol deep within the coating at 800nm – 810 nm is mitigated by a shift at these wavelengths of the intensity peaks so that they occur in the middle of SiO₂ layers (see Fig. 13). An opposite shift occurs at 990 nm – 1000 nm, placing the intensity peaks in the middle of TiO₂ layers (see Fig. 12), but the intensities of these peaks are more moderate. These electric field behaviors overall are favorable for achieving high LIDTs.

Figure 11. Optical electric field intensities (as % of incident intensity) within the coating for 900 nm wavelength light at 45° AOI, S and P pol, for the Sandia BBHR coating design. The shaded areas on the left and right indicate the optical substrate (left) and the air or vacuum incident medium (right), and the vertical dashed lines indicate coating layer boundaries.

Sandia BBHR Coating Design: E-Field Intensity Behaviours at 45deg AOI, S & P pol, for 900 nm wavelength

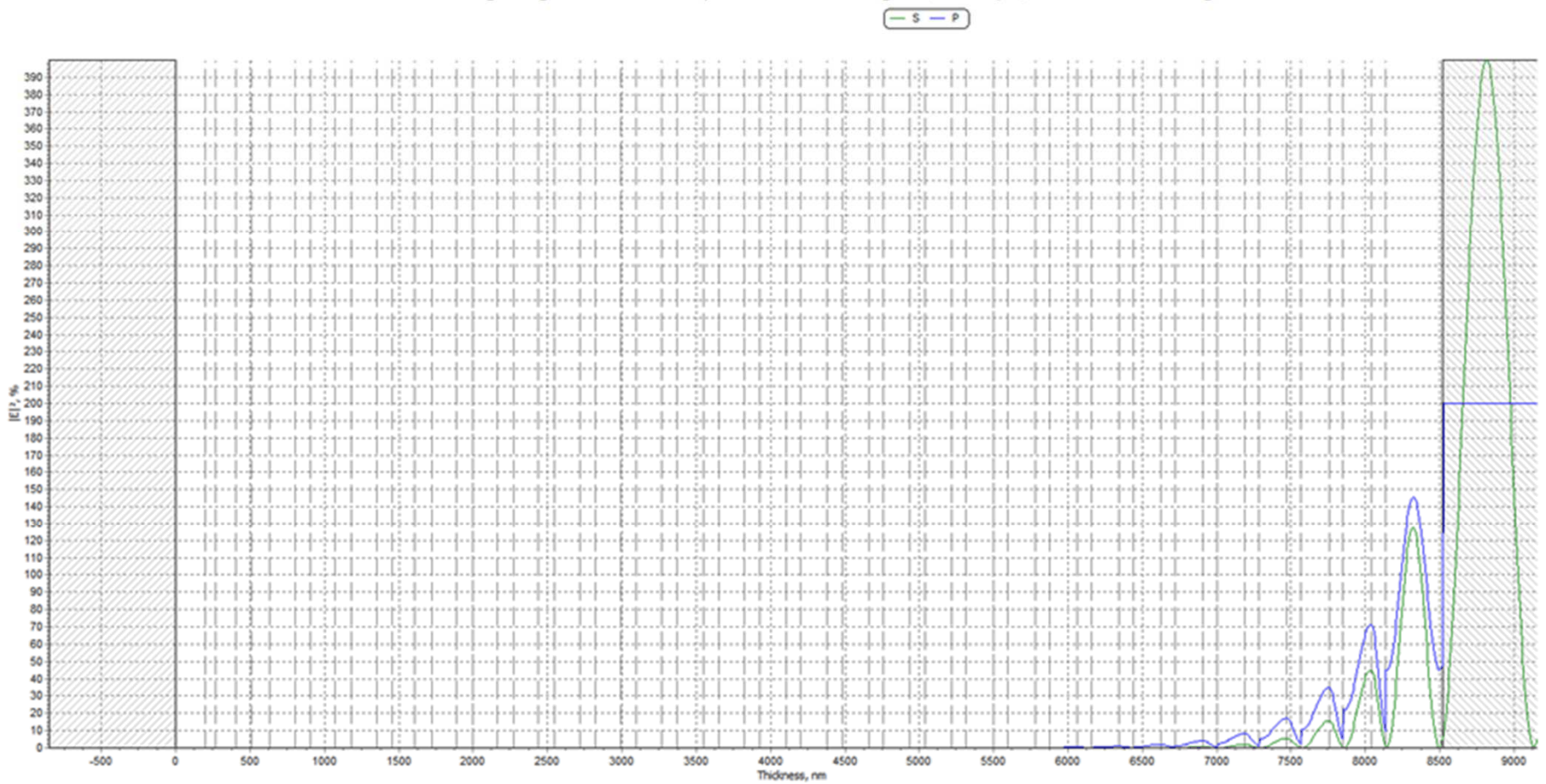


Figure 12. As Fig. 11, but for 1000 nm wavelength incident light.

Sandia BBHR Coating Design: E-Field Intensity Behaviours at 45deg AOI, S & P pol, for 1000 nm wavelength

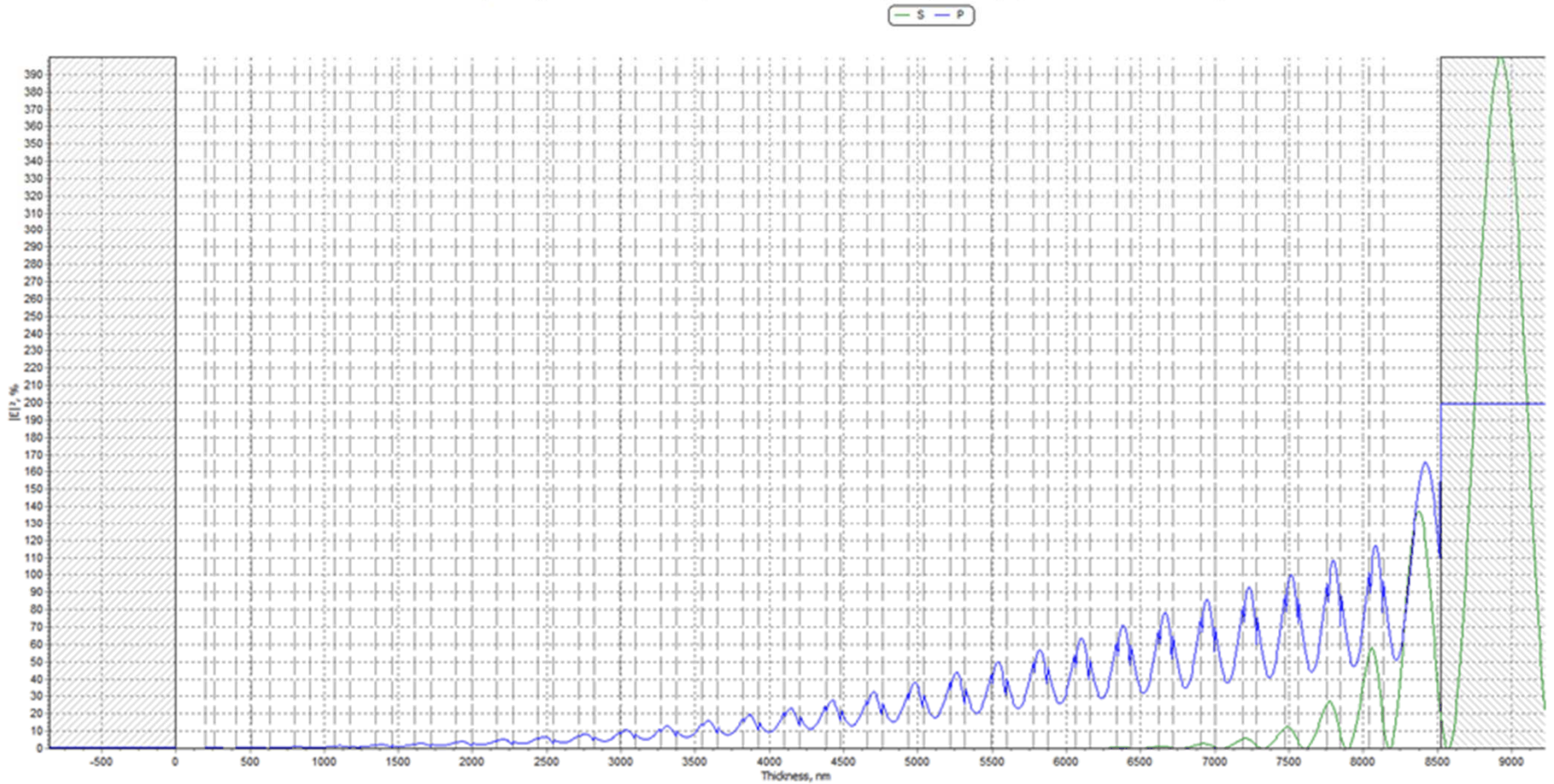
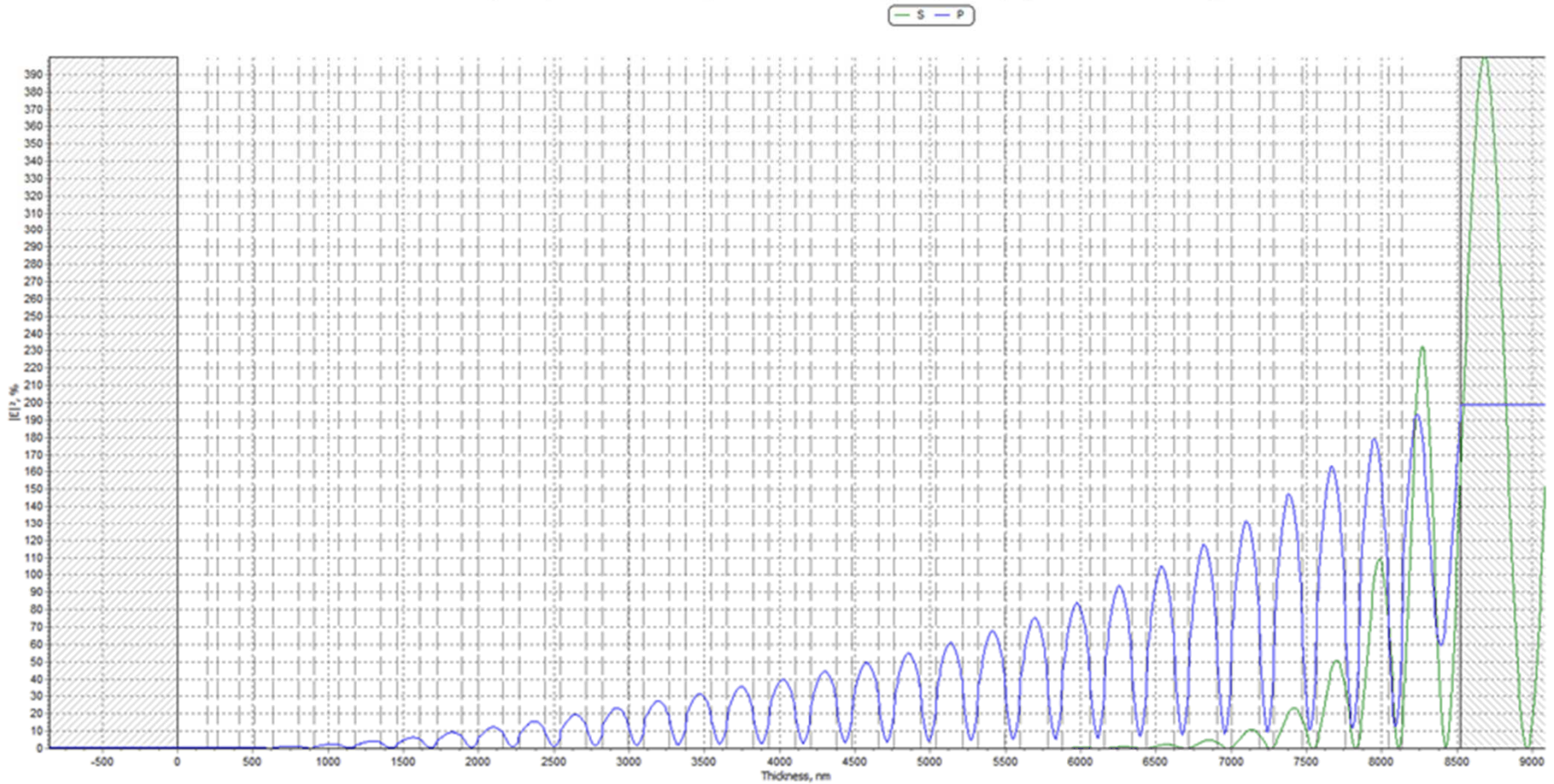


Figure 13. As Fig. 11, but for 800 nm wavelength incident light.

Sandia BBHR Coating Design: E-Field Intensity Behaviours at 45deg AOI, S & P pol, for 800 nm wavelength



5. Summary

- We have developed a BBHR coating design consisting of 30 $\text{TiO}_2/\text{SiO}_2$ layer pairs whose performance specifications behave smoothly and deviate only slightly from the Rutherford Appleton Laboratory design goals of Table 1.
- Furthermore, because of their smooth behaviors, the slight deviations from design goals very likely can be compensated.
- Table 2 summarizes the spectral bandwidth performance characteristics of this BBHR coating design.
- Our considerable experience in producing multi-layer dielectric optical coatings on meter-class optics supports the eventual goal of depositing this proposed BBHR coating on a large dimension mirror substrate.

5. Summary, cont.

Table 2: Spectral Performance Characteristics of the Proposed BBHR Optical Coating for 45° AOI

Coating Property	P pol	S pol
Reflectivity	<p>> 99.5%, 801.22 – 999.24 nm (198.02 nm bandwidth)</p> <p>> 99%, 800.34 – 1000.63 nm (200.29 nm bandwidth)</p>	<p>> 99.5%, 765.64 – 1059.94 nm (294.30 nm bandwidth)</p>
Group Delay	<p>Smooth, well-behaved: ~ 6 fs < GD < ~ 10 fs, 816 - 982 nm GD @ 800 nm = ~ 35 fs GD @ 1000 nm = ~ 30 fs</p>	<p>Smooth, well-behaved: ~ 3 fs < GD < ~ 7 fs, 800 - 1000 nm</p>
Group Delay Dispersion	<p>Smooth, well-behaved: - 20 fs² < GDD < 20 fs², 843 – 949 nm - 50 fs² < GDD < 50 fs², 823 - 969 nm GDD @ 800 nm = ~ 3500 fs² GDD @ 1000 nm = ~ - 3500 fs²</p>	<p>Smooth, well-behaved: - 20 fs² < GDD < 20 fs², 822 - 1000 nm - 50 fs² < GDD < 50 fs², 800 - 1000 nm</p>
3 rd Order Group Delay Dispersion	<p>Smooth, well-behaved: - 100 fs³ < 3rd order GDD < 0 fs³, 804 - 993 nm 3rd order GDD = ~ - 2000 fs³ @ 800 nm and @ 1000 nm</p>	<p>Smooth, well-behaved: - 1 fs³ < 3rd order GDD < 0 fs³, 800 - 1000 nm</p>

5. Summary, cont.

The optical electric field intensity peaks within the proposed BBHR coating are optimal for achieving the highest LIDTs for fs-class as well as ns-class laser pulses. These LIDTs need to be measured for an actual coating over the 800 – 1000 nm operational band. We have positive LIDT results for ns-class pulses at 1064 nm from our 42 layer $\text{TiO}_2/\text{SiO}_2$ HR coating for 45° AOI, P and S pol, over a ~ 200 nm spectral band centered at 1054 nm. In that case, the measured LIDTs at 1064 nm for ns-class pulses, P pol, were 19 J/cm^2 (NIF-MEL protocol) and 12.7 J/cm^2 (ISO 11254-1 protocol) [1]. This result makes us confident that our proposed BBHR coating will easily meet the goal in Table 1 of $> 5 \text{ J/cm}^2$ LIDT for 3 ns pulses over the 800 – 1000 nm operational band. For fs-class pulses, the LIDT will be largely governed by the band-gap properties of TiO_2 and SiO_2 . We expect the highest achievable LIDTs for fs-class laser pulses with the proposed BBHR coating design because of the behavior of its electric field intensity peaks that quench rapidly into the outer coating layers or are of moderate strength or located in the higher band gap SiO_2 layers.

References

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3. John Bellum, Patrick Rambo, Jens Schwarz, Ian Smith, Mark Kimmel, Damon Kletecka and Briggs Atherton, “Production of Optical Coatings Resistant to Damage by Petawatt Class Laser Pulses,” in *Lasers - Applications in Science and Industry*, Krzysztof Jakubczak (Ed.), ISBN: 978-953-307-755-0, available from: <http://www.intechopen.com/articles/show/title/production-of-optical-coatings-resistant-to-damage-by-petawatt-class-laser-pulses> (InTech Open Access Publisher, Rijeka, Croatia, 2011).