Quantized Nanolaminates

– the next step on the road to mass production

Nanolaminate technology offers users the potential to “tailor” thin film production processes in different ways according to their priorities. For some it will be thin film stacks with enhanced optical performance, for others it could be the desire for a “wider” production window, and for others it could be achieving mass production of films only possible until now by IBS at much higher throughput and lower costs. Evatec’s Principal Scientists, **Dr. Silvia Schwyn Thoeny** and **Dr. Stephan Waldner**, report the latest progress on nanolaminate technology through two different cases on CLUSTERLINE® 200 BPM in collaboration with partners from RhySearch and EMPA.

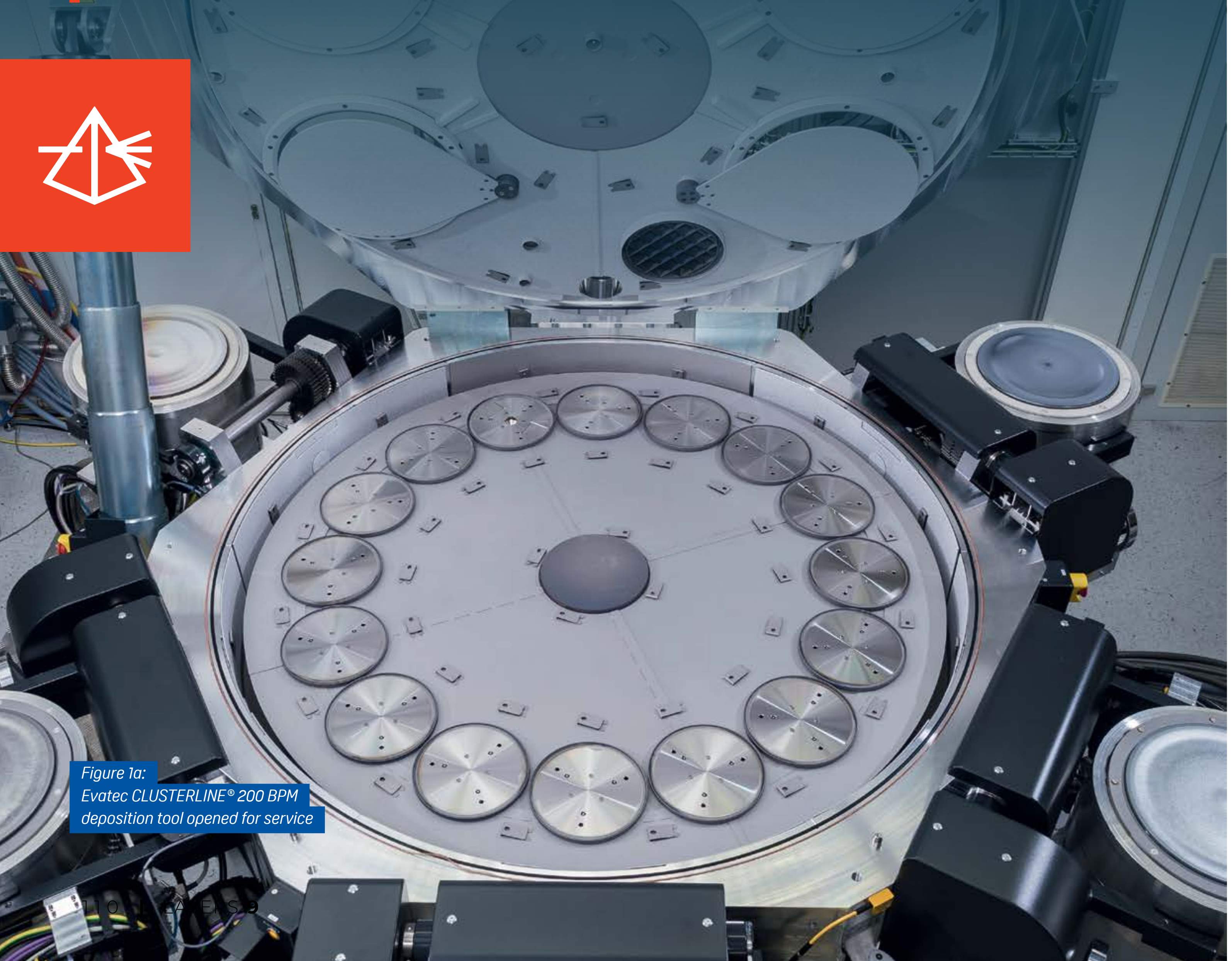

Figure 1a: Evatec CLUSTERLINE® 200 BPM deposition tool opened for service

Production platform set-up

CLUSTERLINE® 200 BPM is a magnetron sputter deposition tool equipped with automatic substrate loading from cassette-to-cassette using a vacuum robot. It can be configured with up to four sputter sources, a plasma source (PSC), plasma emission monitoring (PEM), and an in-situ optical monitoring system with broadband and monochromatic capability.

Figure 1a shows the deposition tool with sputter and plasma sources swung out to service position and the process chamber lid opened. 15 substrates of 200 mm diameter are placed on a turn table. Each substrate is additionally rotating to achieve excellent thickness uniformity without using shaper masks.

When depositing Ta_2O_5/SiO_2 Quantized Nanolaminates (QNL), both sources are running simultaneously (Figure 1b). Both materials are sputtered from metallic targets and oxidized near the sputter target, controlled by the PEM. During one rotation of the table, each substrate receives a thin Ta_2O_5 layer and a thin SiO_2 layer when passing below the respective sources. The thickness of such a layer pair can be adjusted by the table rotation speed, whereas the ratio between high-index and low-index material is defined by the sputter powers (see Figure 2). The PSC is used to pre-condition the substrates or to influence layer stress. Figure 1c) shows a transmission electron microscopy (TEM) analysis of a nanolaminate consisting of Ta_2O_5 (black, approx. 1.35nm thick) and SiO_2 (white, approx. 2.18nm thick) layers.

The flexibility of the tool allows deposition of QNL layers consisting of a variety of materials, e.g. amorphous Silicon (a-Si) and SiO_2. To deposit multilayer optical interference coatings with QNL layers, the process sequence simply consists of the standard high- or low-index material deposition steps and the QNL layer steps.

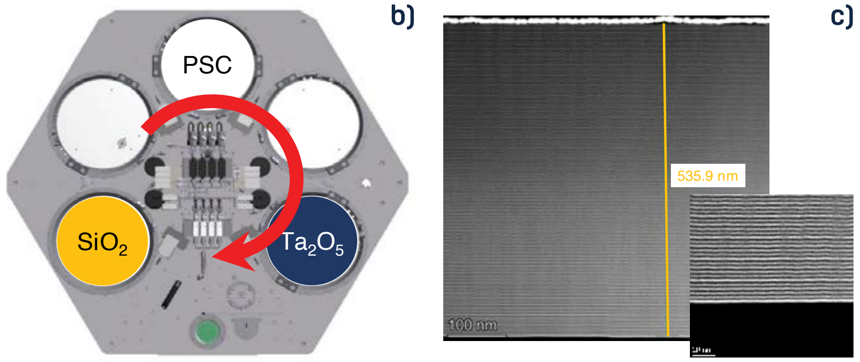

Figure 1: b) Schematic for deposition of Ta_2O_5/SiO_2 QNL; c) TEM analysis of a Ta_2O_5/SiO_2 QNL.

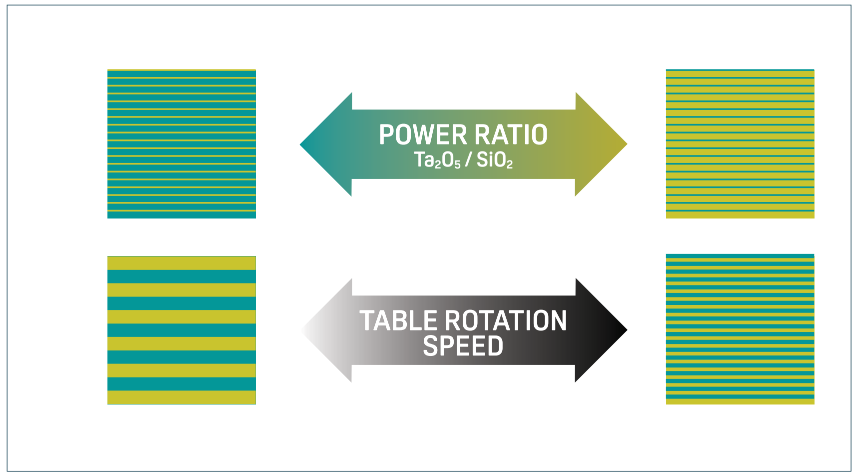

Figure 2: a) Changing the ratio of the power applied to the sputter sources allows tuning of the material composition and therefore the overall refractive index of the QNL. b) By changing the rotation speed of the substrate table, the thickness of a layer pair of the QNL deposited during each rotation can be adjusted without changing the overall material composition.

Why Nanolaminates?

- Improved process stability
- Improved film performance
- Lower cost of ownership in production

Antireflection coatings – A case study

Background
In the early times of optical interference coatings, traditional antireflection (AR) coatings were composed of quarter wave optical thickness layers of three different materials. With the advent of sophisticated coating design programs, antireflection designs with optical thicknesses departing from quarter waves could be designed.

Macleod showed with the concept of "equivalent layers" that any intermediate index layer could be replaced by three layers of high- and low-refractive-index material. This approach typically results in some thin layers, which are very sensitive to small thickness errors and reduced yield. Nevertheless, with the advent of powerful process and optical thickness control, the two-material approach became less complex than handling three materials and thus the two material AR became standard.

In this study, we revisited this assessment using QNL. The effective refractive index of the nanolaminate is defined by the thickness ratio of the high- and low-index material in the nanolaminate layer sequence and can be set to any value within the boundaries of the high- and the low-index material, which in this study are SiO_2 and Ta_2O_5. (Note: In the context of this work, the QNL are not used for the quantization effect and the related shift of the absorption edge, which was shown to occur at individual layer thicknesses of less than 2 nm).

The design study presented here compares antireflection coating designs using QNL layers with intermediate effective refractive index to standard designs using SiO_2 and Ta_2O_5. Compared to the standard design, the QNL approach was expected to show reduced average reflection and improved stability to thickness errors, and would therefore be expected to improve the production yield.

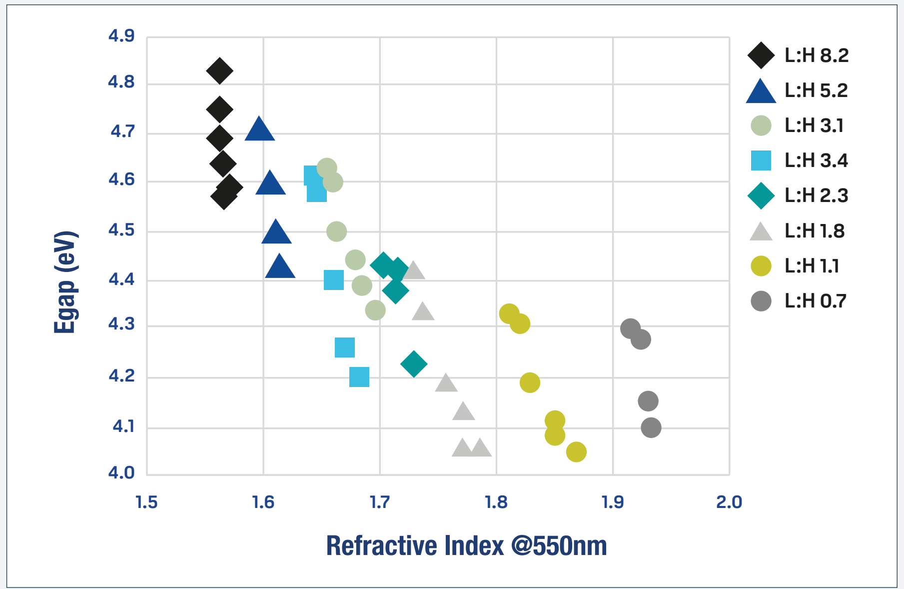

Figure 3: Dependance of gap energy to refractive index

Experiments to develop QNL with a wide range of refractive index
A series of experiments was performed varying the ratio of the thicknesses of Ta_2O_5 to SiO_2. The power on each source was varied as a main parameter. The deposition rates of the nanolaminate layers were typically higher than for the corresponding single material layers, since nanolaminates are sputtered from two sources.

For each H:L ratio, a set of deposition runs with different table speeds was deposited in order to vary the thickness of the individual nanolaminate layers. Typically, table speeds of 1.5 / 3 / 4.5 / 6 / 9 / 12 / 15 s/pass were chosen with the fastest speed of 1.5s/pass leading to the thinnest layers. Figure 3 shows the relation between band gap energy and refractive index.

The gap energy is shifted towards higher energies when the Ta_2O_5 layers become thinner and thereby confirms the occurrence of the quantization effect in the layers. Important in the current context is to note, however, that the effective refractive index can be tuned in a very wide range between the indices of SiO_2 to Ta_2O_5.

Furthermore, the effective refractive index within a series of specific L:H ratios, i.e. average composition, remains in a narrow band with a small trend to lower index for thinner layer pairs.

Design study
Based on these results, three designs of an antireflection coating for the visible part of the spectrum were evaluated. Two of them used refractive indices with intermediate values, while avoiding thin layer thicknesses. Figure 4 shows a comparison of a standard design using SiO_2 to Ta_2O_5 and two alternative designs using intermediate indices based on QNL, one with 3 layers and one with 7 layers. It can be seen from the diagram, that already the 3-layer design leads to a similar average reflection in the range of 450-650nm than the standard design and the 7-layer design improves the performance even more.
Figures 5 a, c & e show the index profiles of the three designs. The 6 layer Ta_2O_5-SiO_2 AR coating contains two very thin layers, which is typical in this type of design. The 3 and 7 layers in the mixed designs have more equal physical thicknesses. As mentioned in the introduction, it is the goal of this work to investigate whether the use of QNLs with intermediate effective refractive index offers enhanced stability against production variations. Thus, the designs were tested by introducing cumulative errors of 2 nm absolute and 1% random RMS deviation in thickness, and of 1% RMS refractive index variation for each material and layer. It can be seen from Figure 5 b, d & f that designs with intermediate index would lead to markedly narrower deviations for the 3 layer and also the 7 layer design.

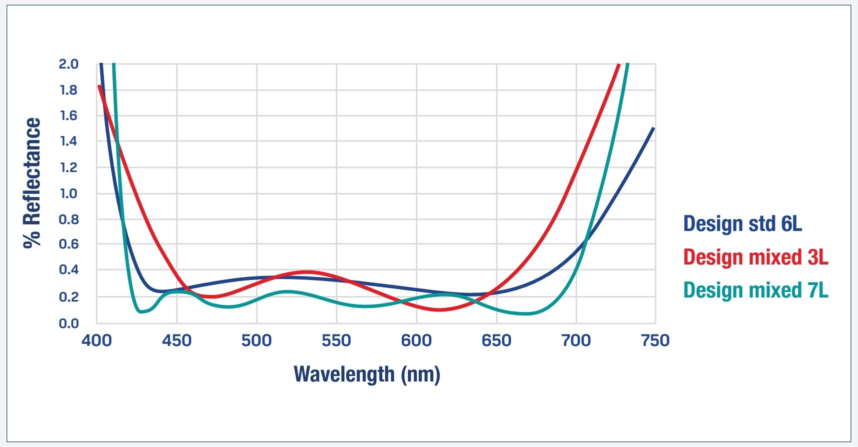

Figure 4: Antireflection designs for a standard 6 layer SiO_2 - Ta_2O_5 and 3 and 7 layer designs using QNL.

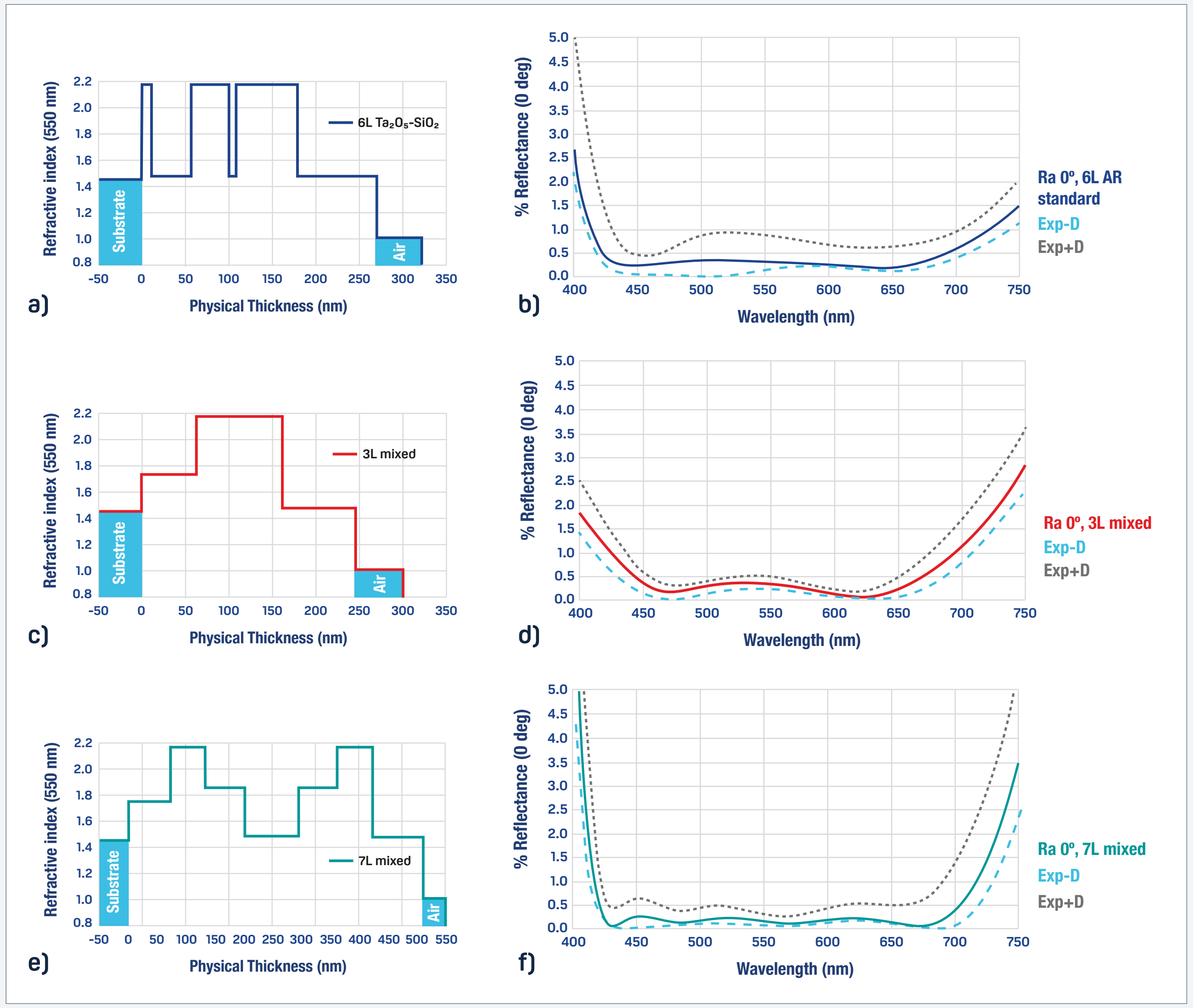

Figure 5: a,c,e) Refractive index profile of 6 layer SiO_2 to Ta_2O_5 and 3 and 7 layer designs using QNL, b,d,f) expected deviation (1σ) of the respective designs due to thickness and refractive index deviations.

Experimental results

Figure 6 shows the actual experimental results achieved on CLUSTERLINE® 200 BPM for the 3 AR coating designs. In each case, the tool produced 10 process runs which were also non-consecutive to test the process robustness in conditions closer to the real world. For comparison of the stability and performance of the produced coatings, the average deviation between the reflection spectra as well as the average reflection over 430 to 680nm was calculated.

Figure 7a shows that both designs using QNL lead to a significantly lower deviation than the standard design. The average reflectance in 7b is almost identical for the standard and the 3 layer mixed design, whereas a much lower value is achieved with the 7 layer mixed design.

> As expected, we see the following important results using QNLs:
>
> 1. Lower average deviation for intermediate-index AR designs
> 2. Lower average reflectance for 7 layer intermediate-index design
>
> This confirms the potential of using intermediate index designs to lower the sensitivity to process deviations relative to standard designs, increasing production robustness or offering the possibility to tailor properties such as angular shift or polarization.

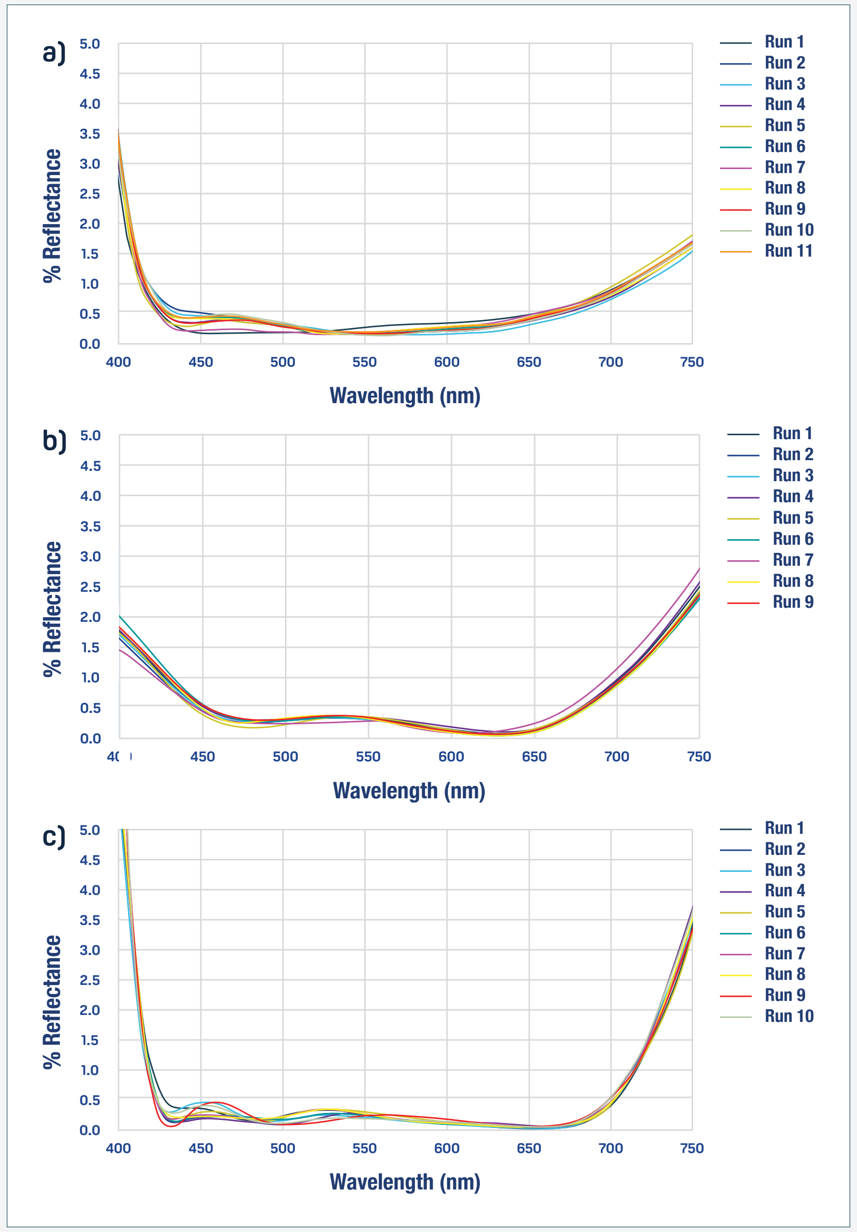

Figure 6: a) Reflection spectra of repeated runs of the standard 6 layer design, b) of the 3 layer design with a QNL material, c) of the 7 layer design with two QNL materials.

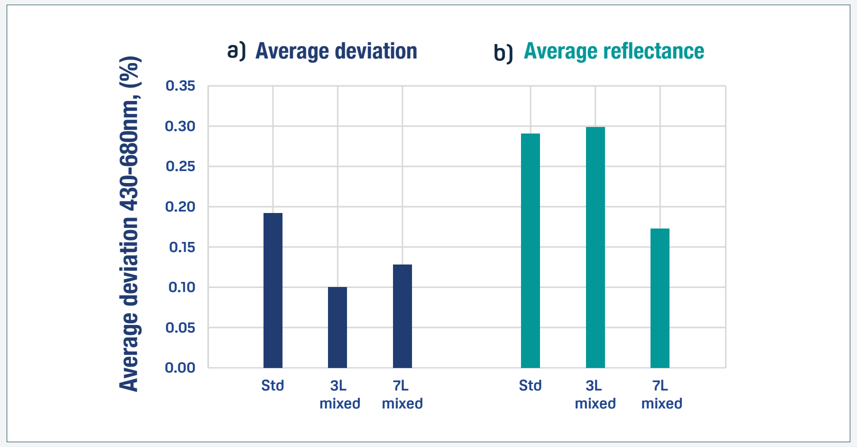

Figure 7: Experimental results comparison – standard vs intermediate index designs: a) average deviation between runs, b) average reflectance over 430…680nm.

LASER Mirrors – A case study

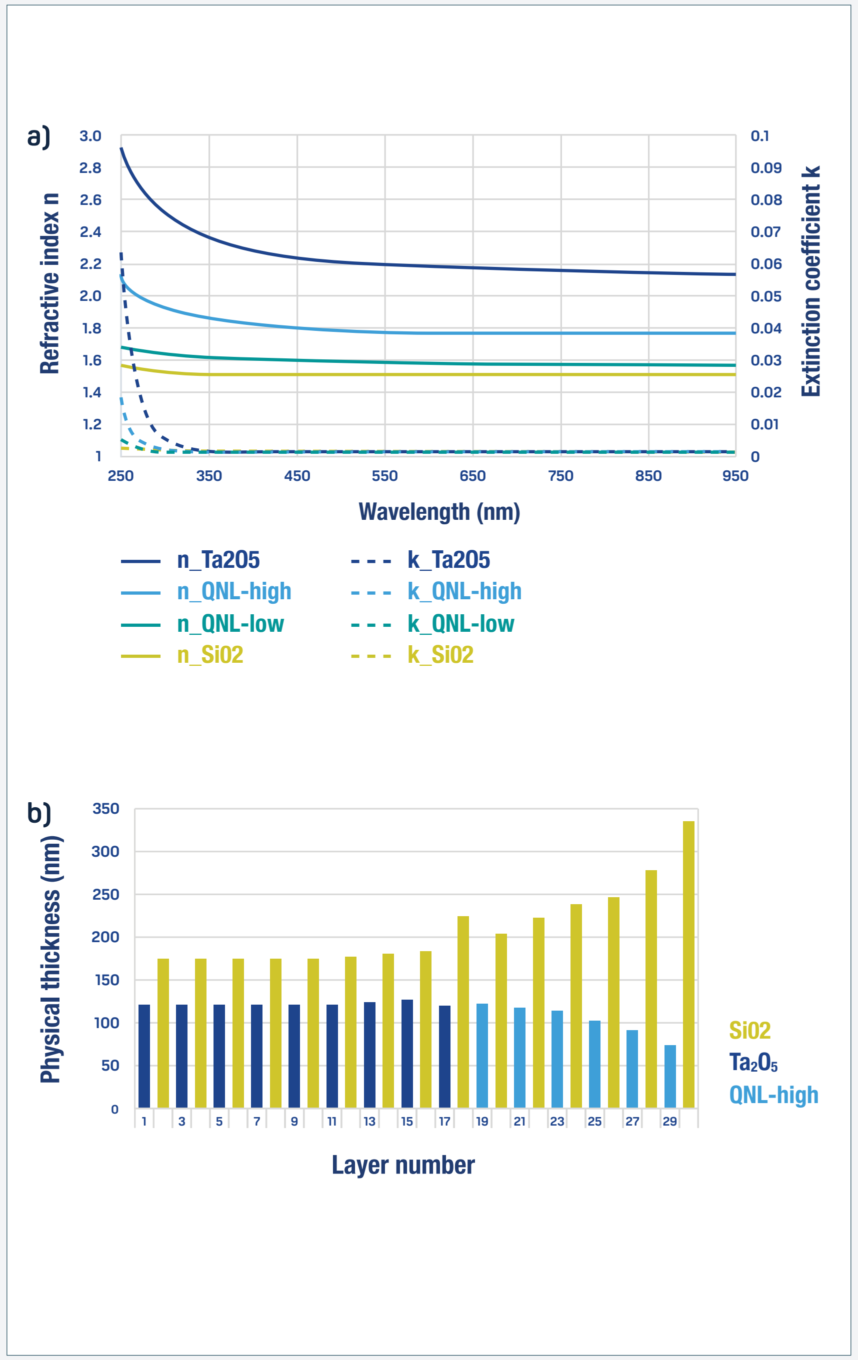

Figure 8: a) Refractive index dispersions SiO_2, Ta_2O_5, and of two Ta_2O_5/SiO_2 QNLs; b) Design thicknesses of a mirror for 1030nm consisting of Ta_2O_5, SiO_2 and a QNL material.

Background

Nanolaminates exhibit a higher band gap energy than what would be expected from a homogeneous mixture of the high- and low-index material. This increased band gap leads to a shift of the absorption edge to shorter wavelengths, as well as to a higher laser-induced damage threshold (LIDT). QNLs therefore enjoyed growing interest in the optical thin film community, but work was mainly performed using ion beam sputtering or atomic layer deposition with relatively low deposition rates and substrate loading capacity. However, if comparable performance of the coatings can be reached, magnetron sputter could offer much reduced cost of ownership for production of QNLs.

In this study, we investigated how multilayer coatings that contain QNLs could be produced on the CLUSTERLINE® 200 BPM sputter deposition tool and compared fs-LIDT results of mirrors for 1030 nm with those produced by standard sputter and IBS techniques.

Design study

QNLs can be used in a multilayer design like an additional material of intermediate refractive index. Figure 8a shows the refractive index dispersions determined from spectrophotometric measurements of two different QNLs, along with the SiO_2 and Ta_2O_5 refractive indices. These material properties can then be used like standard materials in multilayer designs.

As an example, Figure 8b shows the layer thicknesses of a 30 layer mirror for 1030nm at 0° angle of incidence (AOI). For layers 1 to 18 Ta_2O_5 and SiO_2 are used to form the base mirror, whereas QNL and SiO_2 are used for the remaining layers to make use of the postulated higher laser damage threshold of the QNL material. This is justified because high electric field amplitudes are expected near the mirror surface. Additionally, thicknesses were adjusted to lower the E-field amplitudes in the QNL and Ta_2O_5 layers.

When coating the various designs, the thicknesses of the layers were controlled by optical monitoring. The in-situ optical monitoring GSM 1102 equipment measures reflection on a defined substrate at every rotation of the substrate table. Therefore, the monitoring "sees" an additional layer pair of the QNL with every measurement. The measured reflection spectra agree very well with the simulations based on the determined refractive index of the QNL. Figure 9a shows the spectra measured by the optical monitor at the start and end of the standard Ta_2O_5 layer 17, whereas 9b shows the corresponding spectra for the QNL layer 19. This illustrates that the changes in the reflection spectra from start to end of the layers are significant, which allows for precise layer termination by optical monitoring. Therefore, no specific adaptation of the optical monitoring process is required for the QNL layers.

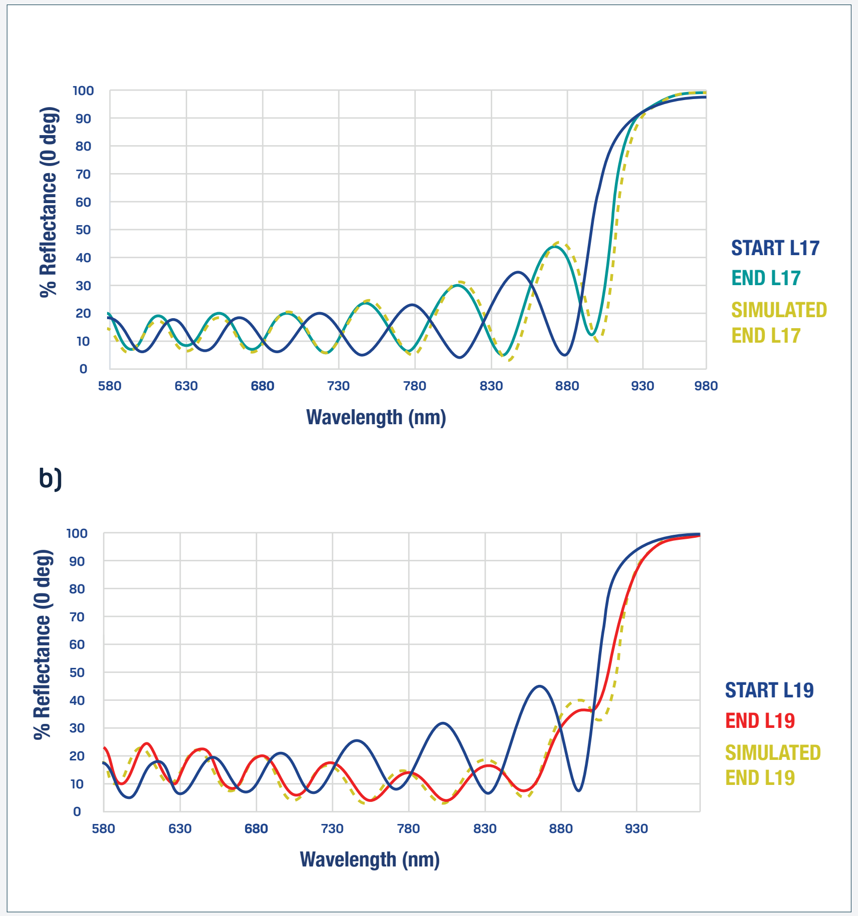

Figure 9: a) Optical monitoring spectra at start and end of the Ta_2O_5 layer 17; b) Start and end spectra of the QNL layer 19.

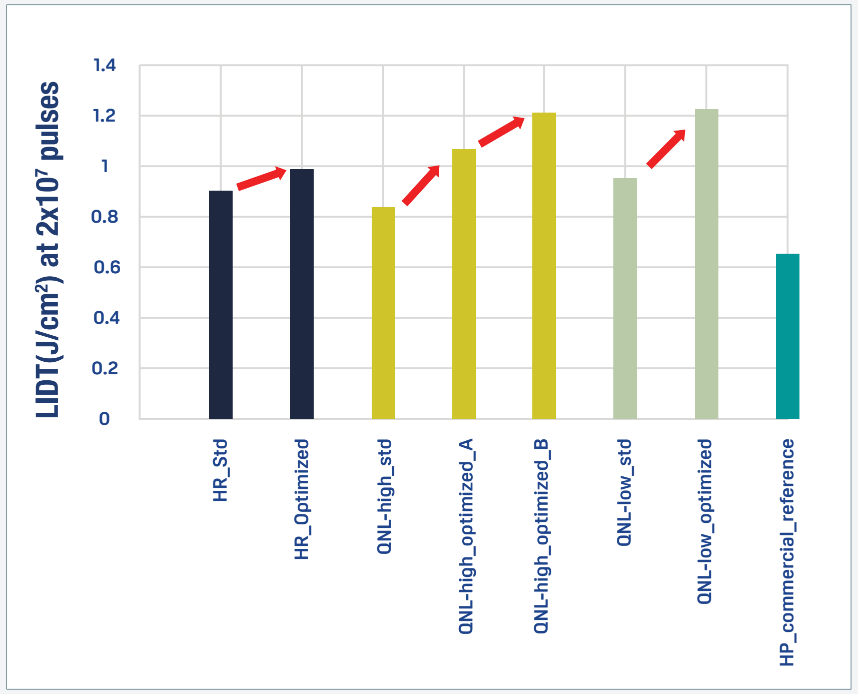

Figure 10: Femtosecond LIDT at 2×10^7 pulses for different mirrors for 1030nm. The arrows indicate the improvement due to the E-field optimization.

Experimental Results

Inspired by the contest of the 2024 SPIE Laser Damage Conference, a series of mirrors for 1030nm at 0° AOI with and without QNL layers were designed, deposited, and characterized. For each material combination, "std" designs composed of quarter-wave layer thicknesses were set up, as well as "optimized" designs, where the layer thicknesses were adjusted to reduce the electric field amplitudes in the higher-index layers close to the mirror surface. The "HR" designs consisted of Ta_2O_5 and SiO_2 layers only, whereas the "QNL-high" designs used Ta_2O_5 for the first part of the design and QNL with high-index for the remaining part. See Figure 8b for layer sequence and thicknesses. Accordingly, "QNL-low" used the lower-index QNL material.

These multilayer designs were deposited as described in the previous sections and then submitted to femtosecond laser damage testing at the LIDT facility at RhySearch. The test conditions were 1030nm laser wavelength with 300fs pulse duration, 200kHz repetition rate, and an effective beam diameter of 32µm. LIDT was determined for 2×10^7 pulse repetitions.

Figure 10 summarizes the LIDT results. The fabricated mirrors achieved an LIDT in the range of 0.83 to 1.23 J/cm². For all material combinations, the "optimized" designs reach higher LIDT than the "std" quarter-wave designs. The two best results are the mirrors which contain QNL layers, whereas the Ta_2O_5 / SiO_2 mirror "HR" does not exceed 1.0 J/cm². For comparison, a commercially available ion-beam sputtered mirror for high power fs-laser applications was included in the test. It reached an LIDT of 0.66 J/cm², i.e. significantly lower than all other samples.

Based on these results, we conclude that tuning the deposition parameters of the CLUSTERLINE® 200 BPM allows for successful adjustment of the refractive index of the QNL and for optimization of the bandgap shift. The QNL layers can be incorporated into multilayer designs using the standard tools for characterization, design, and optical monitoring. LIDT testing of the mirrors showed an excellent performance of the designs containing QNL layers.

NEXT STEPS

We continue to collaborate closely with our partners to refine processes on 6" and 8" substrates. Interested in exploring nanolaminate technology with us? Reach out to your local Evatec sales and service team to discover how we can support your next innovation.

Acknowledgements

The work reported here is a compilation of two studies first reported at the Optica Optical Interference Coatings conference, Tuscon, Arizona in May 2025 https://www.optica.org/events/topical_meetings/optical_interference_coatings with additional material added since that time.

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