Supplementary Materials: Optical and Chromaticity Properties of Metal-Dielectric Composite-Based Multilayer Thin-Film Structures Prepared by RF Magnetron Sputtering

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1. Tooling Factor Optimization for the Single-Layer Ultrathin Ag Layer Growth

The thickness of ultra-thin Ag layers plays an important role in the design and manufacture of spectrally selective optical coatings for various applications. This is the reason it is necessary to have an optimized growth process for silver layers. We conducted multiple deposition runs and evaluated the transmission spectra (obtained by using Agilent Cary 5000 spectrophotometer) by comparing with the modeled spectra (simulated by using OptiLayer Pro and the known dielectric constants of sputter-deposited thin silver layers (Reference 25 in the main manuscript)) to calibrate and fine-tune the tooling factor (TF %). Multiple fitting experiments of the measured transmission spectra to the modeled transmission spectra allowed fine-tuning of the tooling factor. Figure S1 shows a sketch-type process-flow diagram for tooling factor calibration process.



Figure S1. Schematic diagram of the process-flow used to obtain the tooling factor calibration to enable accurate in-situ thickness measurements during the growth of ultra-thin Ag layers.

Figure S2 presents the transmission spectra of the as-deposited Ag layers, compared and closely fitted against the modeled transmission spectra of the corresponding Ag layers.





Figure S2. Details of the multiple spectral fitting experiments conducted to finalize the materials tooling factor for the thin Ag layers deposition.

2. Calculation of the Volumetric Fraction of Added MgF₂, Optical Fitting of the Transmission Spectra, and Results for the Absorption Coefficient Spectra of the Single-Layer Ultrathin Ag + MgF₂ Composite Layers

The estimated amounts of the volumetric fraction of added MgF₂ were quantified by using the measured partial deposition rates from both (Ag and MgF₂) sputtering sources calibrated during the preliminary deposition processes. The basic formula used to calculate the added vol.% fraction of MgF₂ in metal-dielectric composite-type layers is as follows:

$$Vol. \% of MgF_2 = \frac{Deposition rate of MgF_2}{Total deposition rate (Ag + MgF_2)}$$
(1)

For example, if the partial deposition rates were 5.7 nm/min for Ag and 0.3 nm/min for MgF₂ then the volumetric fraction of added MgF₂ is

Vol. % of
$$MgF_2 = \frac{0.3}{(5.7+0.3)} \times 100 = 5\%$$
 (2)

Figure S3 presents the transmission spectra of the as-deposited MDC layers, compared and closely fitted against the modeled transmission spectra of respective MDC layers. The MDC layer modeling with OptiLayer involved using the weighted average of the known dielectric permittivity functions of both component materials. The left column shows the results of transmission fitting to the optically

equivalent pure-Ag layers; the right column shows the calculated optical absorption coefficients for each MDC layer, as well as their upper and lower limits, based on considering a maximum 5% error in the actual physical thickness.



Figure S3. Spectral fitting of the optical transmission of MDC layers to the modeled MDC layers and the calculated absorption coefficient spectra of the MDC layers.

Figure S4 shows the calculated absorption coefficients of MDC layers of slightly different layer thickness, compared to that of a pure silver layer. It can be clearly noticed that significant changes in

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the absorption coefficient were observed (across the visible spectral range, between 350–750 nm) in the MDC layers.



Figure S4. Optical absorption coefficient spectra of MDC layers of slightly different thickness compared to the absorption of pure thin-film Ag layer.



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