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Performance of Antireflection Coatings Consisting of Multiple Discrete Layers and Comparison with Continuously Graded Antireflection Coatings

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The performance of discrete multilayer and continuously graded antireflection coatings for omnidirectional broadband applications are compared. It is shown that in practical cases where refractive index choices are constrained, discrete antireflection coatings can surpass the performance of continuously graded coatings by taking advantage of interference effects, which continuously graded coatings are expressly designed to avoid. A four-layer antireflection coating designed using a genetic algorithm is fabricated, and is experimentally shown to have reflectivity lower than what is achievable for continuously graded designs.

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Designs for broadband omnidirectional antireflection (AR) coatings are generally motivated by the hypothesis that smooth and gradual variation of the refractive index over an extended distance will produce the best possible AR characteristics. AR coatings with refractive index profiles that vary continuously from the substrate refractive index to the ambient value are known to have excellent performance. However, in virtually all applications, the refractive index of the AR coating is constrained by the availability of materials with refractive indices matching the substrate and the ambient. Nevertheless, experimentally realized broadband omnidirectional AR coatings have used refractive index profiles intended to approximate continuous grading. Here it is shown that, contrary to common assumption, AR coatings consisting of only a few discrete layers can significantly outperform AR coatings of *any* continuously graded profile over a broad range of wavelengths and incident angles. Even compared with infinitely thick continuously graded AR coatings, the discrete multi-layer approach has a lower angle- and wavelength-averaged reflectivity.

The pursuit of low-reflectivity coatings has fascinated researchers for more than a century.[1–9] While it is straightforward to attain low reflectivity for normal incidence and a narrow wavelength range, achieving omnidirectional and broadband AR characteristics is far more challenging, but also more relevant for many applications. In 1880, Lord Raleigh analyzed reflections of waves from graded interfaces between two dissimilar media, and realized that "the transition may be so gradual that no sensible reflection would ensue".[1] That is, for an infinitely thick, continuously graded AR coating, Fresnel reflectivity approaches zero. Significant research has focused on finding refractive index profiles which minimize reflection for a given AR coating thickness. A linear profile is a reasonable starting point, but other profiles, such as the quintic profile, have been found to give superior performance.[3,4)]

Recently, oblique-angle deposition has been used to demonstrate nanoporous films with a refractive index approaching that of air.[9,10] These nanoporous films are stable, showing no signs of deterioration over time or in response to aqueous environments. Besides allowing very low refractive index values, oblique-angle deposition and other techniques can be used to precisely tune the refractive index within a broad range by varying a material's porosity.[11,12] In principle, this combination enables AR coatings in which the refractive index is continuously graded. However, a fundamental limitation prevents implementation of true continuously graded air-ambient AR coatings matching Raleigh's description: there exist gaps in the range of attainable refractive indices for transparent materials. For example, while refractive indices as low as 1.05 have been reported,[9] values below this have not been reached, though they would be required for a true continuous AR coating where air is the ambient medium. In addition, transparent materials generally are not able to match the refractive index of an absorbing substrate, because absorbing materials have a complex refractive index whereas for transparent materials the refractive index is necessarily real. Thus, in the vast majority of practical applications, it is simply not possible to realize a graded AR coating as envisioned by Rayleigh, because portions of the required range of refractive index values are unavailable.

One might suppose that an infinitely thick, continuous graded-refractive-index approach is still the best option, despite unavoidable discontinuities in refractive index. Following this assumption, experimentally realized AR coatings are frequently intended to approximate continuous profiles.[2,6–9] In this paper, however, we demonstrate that such coatings are, in fact, *not* the optimum for minimum angle-averaged and wavelength-averaged reflectivity for virtually all practical cases. We show that discrete multilayer coatings enable reflectivity far below values attainable by continuously graded coatings. Specifically, we show that interference of light — a phenomenon which is expressly avoided in continuously graded AR coatings — enables discrete multilayer coatings to deliver superior performance, particularly for broadband and omnidirectional applications. Approximating our step-graded refractive index profile with a continuously graded profile has been confirmed to worsen the reflection properties of the AR coating, i.e., the interference effects that continuously graded coatings expressly avoid, are weakened. This result is relevant to virtually all thin-film coatings that have practical uses in photonics applications. Specifically, it applies to common components such as solar cells, sensors, detectors, lenses, and solid-state lighting devices. We consider light incident from a low-refractive-index ambient upon a high-refractive-

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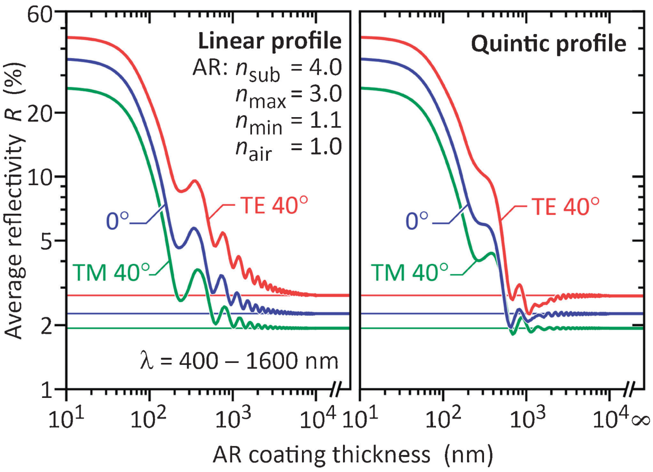


Fig. 1. Calculated wavelength-averaged reflectivity as a function of angle for linearly graded and quintic refractive-index profiles at 40° and normal incidence. Substrate refractive index is 4.0, the ambient index is 1.0, and the AR coating refractive index is limited to values between 3.0 and 1.1.

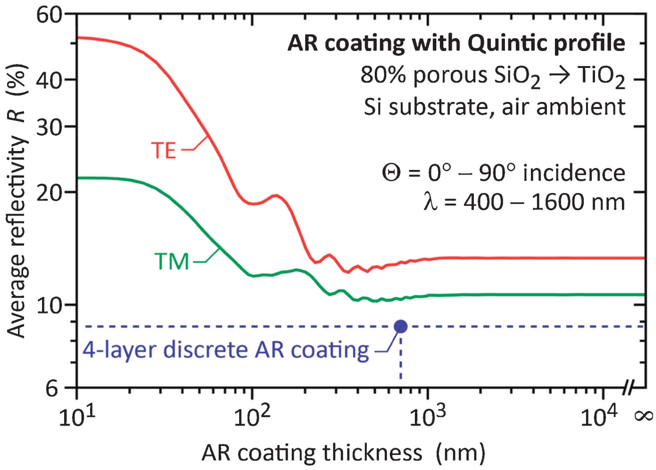


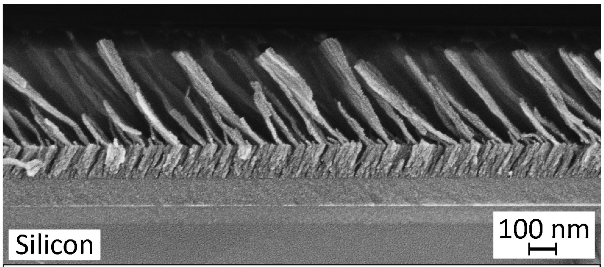
Fig. 2. Calculated angle- and wavelength-averaged reflectivity of an AR coating with a continuously graded quintic profile as a function of the coating thickness. Also shown is the averaged reflectivity of a discrete four-layer AR coating with thickness of approximately 700 nm. The discrete multilayer coating outperforms the continuously graded quintic profile for any thickness of the quintic coating.

index substrate. In the limit of a transparent, infinitely thick, continuously graded AR coating, the reflection coefficient is $R = 1 - (1 - R_1)(1 - R_2)/(1 - R_1 R_2)$, where R_1 and R_2 are the angle- and wavelength-dependent reflection coefficients, at the interfaces between (1) the ambient and the adjoining material of the AR coating, and (2) the substrate and the adjoining material of the AR coating, respectively. When these interfacial regions are also continuously graded, then the total reflection coefficient will be zero. However, in cases where perfect refractive index matching is not possible, the discontinuity in refractive indices results in unavoidable reflections. The calculation of a wide variety of continuously graded profiles reveals that all of them asymptotically approach the reflection values shown for an infinitely thick coating. Figure 1 shows the reflection coefficient for a linearly-graded and quintic AR coating without perfect refractive index matching, and demonstrates that even in the limit of infinitely thick graded-index coatings, there is a limiting value of the reflectivity which is independent of the functional form of the refractive index profile.

To demonstrate the high performance of optimized multilayer AR coatings, we consider light incident from air upon silicon, with the requirement that the AR coating be composed of nanoporous SiO_2 and mixed SiO_2/TiO_2. We limit the porosity of SiO_2 to 80%, which yields a refractive index around 1.1. We include all incident angles (0–90°) and wavelengths between 400 and 1600 nm, and material dispersion is taken into account. Designing such multilayer AR coatings requires care, as the configuration space of a multilayer AR coating has many local minima for reflection, and makes finding the true optimum AR coating a challenge. To design optimized multilayer AR coatings, we use a genetic algorithm, which prevents capture within local minima and is well suited to the design of optical coatings.[13,14]

Figure 2 shows the calculated wavelength- and angle-averaged reflection coefficient as a function of thickness for a continuously graded quintic AR coating, and also shows the performance of a 700-nm-thick four-layer AR coating designed to minimize the reflection coefficient optimized over all wavelengths and angles. The results demonstrate the superiority of discrete multilayer coatings: An AR coating consisting of four discrete layers outperforms the continuously-graded quintic profile AR coating for *any* thickness of the quintic coating. As the thickness becomes large, the averaged reflection coefficient of the quintic AR coating approaches 13.59% for the transverse electric (TE) polarization, compared to 8.86% for the four-layer design. For transverse magnetic (TM) polarization, continuous grading yields 10.76% average reflection compared to 8.60% for the four-layer design. As mentioned earlier, in the limit of infinite thickness all continuously graded AR coatings have identical performance, and therefore the optimized four-layer design is superior to any conceivable continuously graded AR coating. Compared to the optimized four-layer design, the angle- and wavelength-averaged reflection coefficient of an infinitely-thick continuously graded AR coating is a remarkable 53 and 25% higher, respectively, for TE and TM polarizations.

The four-layer AR coating is experimentally realized using co-sputtering for TiO_2/SiO_2 layers, and oblique-angle deposition for nanoporous SiO_2 layers. An scanning electron microscopy (SEM) image of the AR coating along with layer specifications is shown in Fig. 3. Layers of the AR coating are fabricated by co-sputtering of TiO_2/SiO_2 and oblique-angle deposition of SiO_2 using e-beam evaporation. Deposition angles for the nanoporous SiO_2 are 85 and 60° for the 80 and 30% porous layers, respectively.[11] Film thicknesses and refractive indices are characterized using variable angle spectroscopic ellipsometry. The porosity of a material is determined from the measured refractive index and a linear volume approximation in conjunction with an analytic expression for porosity as a function of deposition angle.[11,12] The reflection coefficient is measured as a function of angle using a broadband white light source and an optical spectrum analyzer.



Layer 4	80% porous SiO_2	386 nm	$n = 1.09$
Layer 3	30% porous SiO_2	142 nm	1.32
Layer 2	68% SiO_2/32% TiO_2	95 nm	1.77
Layer 1	TiO_2	73 nm	2.42

Fig. 3. SEM image of the four-layer AR coating on a silicon substrate, shown with layer composition, thickness, and refractive index data. The refractive index is given at a wavelength of 485 nm.

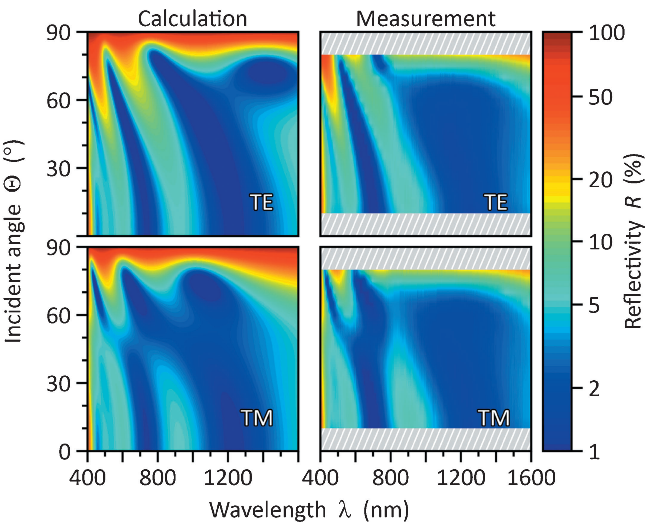


Fig. 4. Calculated and measured angle- and wavelength-averaged reflectivity of the four-layer AR coating on a silicon substrate.

Figure 4 shows both calculated and measured reflectivity as a function of wavelength and incident angle for TE and TM polarization for the four-layer AR coating, and reveals local minima in reflectivity that are indicative of the interference which allow performance beyond the level of continuously-graded coatings. Experiment and calculation show excellent agreement. The measured average reflectivity between 10 and 80° and between 400 and 1600 nm is 5.29 and 4.63% for TE and TM polarization, which are very close to the calculated values of 4.93 and 4.32%, and to our knowledge, are the lowest values ever reported.

The underlying reason that this four-layer AR coating outperforms even an infinitely thick continuously graded coating is found in the interference of light reflected at different interfaces. Just as in a "perfect" normal incidence quarterwave coating, by properly optimizing the thicknesses and refractive index contrasts of a discrete multi-layer AR coating will result in destructive interference for reflected light, and therefore low reflection coefficient. For broadband (400–1600 nm) and omnidirectional (0–90°) AR coatings, this is highly surprising since one would expect that any interference effects over a wide range of wavelengths and angles will average out. High performance is achieved by taking advantage of the very phenomenon that continuous AR coatings are specifically designed to eliminate. Therefore, only by discarding the continuous AR coating profiles as a starting point can performance of an AR coating be truly maximized.

In summary, we have shown that the performance of continuously-graded AR coatings is fundamentally limited in virtually all practical cases where perfect index matching is not available. Further, it is theoretically demonstrated that discrete multilayer AR coatings can outperform continuously-graded AR coatings, by taking advantage of interference effects arising from reflections within the coating — something which is expressly avoided in continuously-graded designs. Using a genetic algorithm, an optimized a four-layer coating is demonstrated which offers substantially lower reflectivity than even a hypothetical, infinitely thick continuously graded coating. The broadband and omnidirectional reflection characteristics of an experimentally realized four-layer (TiO_2/SiO_2)/nanoporous SiO_2 coating are in excellent agreement with theory.

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