Nanostructures versus thin films in the design of antireflection coatings

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ABSTRACT

It is shown how the discussion about antireflection coatings for the visible and near infrared region has been changed dramatically with recent experimental applications of nanostructures that realize media with effective refractive indices less than the 'magic border' of 1.34. Using the so-called binary optics as an example, a glass-like nanostructure similar to the moth-eye structure is theoretically designed as antireflection coating for the visible and near infrared region. With the aim of this example and considering only known design principles of thin-film optics, a connection between nanostructures and thin films regarding their alternative or combined application as antireflection coatings is presented. As summary regarding the nanostructures vs. thin film discussion, a reference list is presented that cited different types of antireflection coatings presented in the past 70 years with respect to their applications, designs, and deposition technologies.

Keywords: Thin film optical coatings, Zero-Order-Grating, Antireflection

1. INTRODUCTION

The application of a single MgF_2-layer as antireflection (AR) coating on transparent lenses highlighted the beginning of thin-film optics 75 years ago.[1] At present, thin-film optics is characterized by the application of multilayer coatings with up to 300 layers and the realization of nearly any desired spectral characteristics.[2] Nevertheless, AR coatings consist still of few layers but countless papers have been published and theoretical discussions have presented also multilayers of 30 or more layers due to the application of multi-cycle or multi-cluster designs.[3-5] Answering the question how to eliminate the reflection at the boundary of two media of different refractive indices by thin films seems to be just only a question of the number of layers in the design of thin film coatings.[6,7] The main feature for the performance of any AR design is the refractive index of the layer directly adjacent to the incident medium. The AR performance of any design is the better the lower the difference is between the refractive index of this layer and the refractive index of the incident medium.[8] There is a 'magic border' of about 1.34 for the lowest available refractive index regarding suitable materials and their deposition methods for the manufacturing of stable AR coatings in the visible spectral range.[9] On high-index substrates applied for infrared optics, the refractive index n=1.32 at 9 µm of a CaF_2 thin film maybe close enough to unity to use such a thin film very well for a broadband AR coating.[10] Improvements of the performance of the AR coatings have been reached successfully by the application of standard materials as MgF_2 and SiO_2 for the outermost layer but exclusively by applying special deposition methods as solgel process or oblique angle deposition.[11-13]

In any case, the characteristic feature of a 'thin-film' AR coating is its stratified structure normal to the substrate surface and the homogeneity of this stratified structure lateral versus the complete surface. But one has to consider another practical solution of the AR problem in form of lateral modifications in the depth of the substrate surface or a thin film deposited on the substrate. The basic for such an AR solution is the fact, that the lateral dimensions of these surface modifications have to be less than the shortest wavelength of the wavelength range for which the optical application is provided.[14] Considering the application of transparent gratings in optics there has been always the possibility, that diffraction orders do not occur if the grating period is less than the wavelength of its application.[15] The grating is called a zero-order-grating and its lateral modification is a sub-wavelength structure.[16]

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Because some different terms exist to characterize this effect as surface-relief structure, porous microstructure, or sub-wavelength structure, here we prefer the term ‘nanostructures’ due to the fact that most of the interesting applications of such solutions for the AR problem require structures in the dimension of nanometres from 10 nm to 1000 nm, both in lateral and in vertical direction with respect to the substrate surface. However, the real dimensions of these nanostructures depend strictly on the wavelength range of their applications and in this sense, the application of nanostructures as AR coating can be expanded, e.g. up to the millimetre scale for the very topic THz imaging and spectroscopy.[17]

The two methods of AR coating designs differ in their technical realisation but they he same optical principle. Whatever method is applied there is a transition region between ambient medium and substrate and this region is defined by its effective refractive index or a refractive index profile and its overall physical thickness. But nobody can say that there is a solution better than another one. However, different AR designs and different technical realizations can be used for the same application and give the same results. In this sense, a discussion under the viewpoint ‘nanostructures versus thin films’ may help to understand the principles without consideration of a complex theory. In Section 2 we describe AR designs made by nanostructures or homogenous thin films or inhomogeneous thin films in general and section 3 gives some remarks to the oblique incidence in general and a straightforward design approach. In Section 4 we discuss the meaning of the refractive index profiles in particular and consider oblique incidence with respect on the concept of the so-called reflectionless potentials.

2. THE AR PROBLEM AND THE EFFECTIVE REFRACTIVE INDEX

Consider a transparent and plane optical element as a substrate with the refractive index n_S and an incoming radiation from the ambient medium with the refractive index $n_0 < n_S$. Ignoring the second surface of the substrate, the AR problem is defined by the fact that the reflectance at the boundary of ambient medium and substrate is unwanted and has to be reduced to a defined value within a given spectral range. This spectral range is defined by its lower and upper boundaries λ_l and λ_u which gives a bandwidth $BW = \lambda_u/\lambda_l$ and a reference wavelength $\lambda_0 = 2\lambda_l\lambda_u/(\lambda_l + \lambda_u)$ within this spectral range. If the BW is equal or greater than about 1.5 we talk about a broadband AR problem.

As mentioned in the introduction, a transition region with an effective refractive index n_{eff} and a physical thickness t is placed in between the ambient medium and the substrate. In a first approach and without loss of generality we can assume normal incidence, media without any losses, and materials with constant refractive indices. Then, there is a zero-reflection at the reference wavelength if the transition region is made by a thin film with a homogeneous refractive index that fulfills the root-condition $n_{film} = \sqrt{n_S n_0}$ and a physical thickness that fulfills the quarter-wave-condition $t = \lambda_0/4/n_{film}$ as it is described in the basics of thin film optics.[9] The same ideal solution of the AR problem is possible if the transition region is made by a one-dimensional (1D) grating into the substrate depth, with grating period $\Lambda \leq \lambda_0/n_S$, grating height $h = \lambda_0/4/n_S$, and filling factor f as ratio of the width of the solid part of the grating to the grating period as it is described in some fundamental papers concerning the equivalence of gratings and homogeneous layers.[18,19]

Additionally, a third solution of the AR problem is possible if the transition region is a thin film having an inhomogeneous refractive index normal to the substrate surface but a constant index lateral versus the substrate surface. This gradient index is more or less stepless but the gradient follows any index profile in dependence of the depth into the transition region. Ideally, there is a continuous increasing of the refractive index from that of the ambient medium to that of the substrate. However, if such an inhomogeneous thin film is applied as AR coating, the physical thickness t of the transition region depends on the index profile and it is in any case $t > \lambda_0/4/n_{ave}$ where n_{ave} is the geometric mean of all the refractive index values that occur within the gradient index.[20]

Regardless of all the known methods for the description of the propagation of electromagnetic waves in stratified media and gratings as Matrix Method, Rigorous Coupled Wave Approach, Effective-Medium Theory, Green’s Function Surface Integral Equation Method, or Finite-Difference Time-Domain Method, a straightforward approach for this transition region can be made by the following assumption: If we consider coherent waves of the same wavelength within a small pixel of such a transition region with a lateral length of half the wavelength and a vertical height of a quarter of the wavelength - then we can assume that phase differences occurring between the different waves within the pixel are the necessary condition and boundaries of different refractive indices are simply a sufficient condition. That means whatever approach is applied within the defined area of the transition region – homogeneous thin film or inhomogeneous thin film or nanostructure – the application as solution for an AR problem requires a phase change by changing the refractive index in any way within the defined pixel. To describe these effects one can use the method that is accustomed to one and we prefer the matrix method.[9]

Considering a 2D-grating there is usually not an exact definition whether the grating is convex or concave with respect to the substrate surface. In this sense, the consideration of a transition region is obviously where the refractive index changes someway vertical and lateral within this region. Additional to the grating-relief structures it is possible to apply holes of different refractive index with respect to the index of the ambient material or the material of the transition region is someway a mixture of two materials having different refractive indices.[21-23] In any case, the dimensions of the holes or the structures of different refractive indices are within the quarter-wave dimensions that characterize the optical thickness of the transition region with respect to the wavelength range of the AR design and we can use the term 'nanostructure' also for such surface modifications. Furthermore, a periodicity is not required necessarily to apply a nanostructure as AR coating. Stochastic distributions of grating periods, nipple diameters, or structure heights are possible and have been applied successfully.[24-29]

3. OBLIQUE INCIDENCE AND A STRAIGHTFORWARD DESIGN APPROACH

Under nearly normal incidence, the three possibilities of an AR design regarding the modification of the transition region with respect to the refractive index may realize the same spectral characteristics within the desired wavelength range; however, there are great differences in the spectral characteristics outside this AR range. Homogeneous thin films produce periodic characteristics with higher-order reflection bands and broadband AR designs have to consider this effect, particularly. Inhomogeneous thin films give remarkable differences at shorter wavelength in dependence of the index profile. Nanostructures loss their AR characteristics at longer wavelength due to the mismatch of the thickness condition and produces scattered light at shorter wavelength due to the occurrence of higher orders of diffraction. Therefore, detail analysis is needed if the conditions have to be discussed that constrict the application of a nanostructure with respect to the wavelength region or its geometrical dimensions and other methods than the matrix method have to be applied strictly.[15,16]

Under oblique incidence, all the AR characteristics shift generally towards shorter wavelength due to the dependence of the phase thickness on the angle of incidence but the characteristics change differently versus the wavelength and the angle of incidence with respect to the TE and TM modes. If we merge from a 1D structure to a 2D one, we can assume approximately the structure has the same period in x and y direction or it is concentrically around the z-axis and so there are quarters or cylinders, if the 1D structure is rectangular; pyramids and cones, if the structure is as a saw-tooth; or there are any shapes of rods or nipples, if the 1D structure does not change linearly in z direction normal to the surface. In any case, under oblique incidence the effective index of the 2D-structure becomes an average one and there are approximate solutions for standard models.[19,25] Generally, there is a nipple-shaped model developed by the nature in form of the corneal nipples of the moth-eye, known in the fields of biology and ophthalmology since 1962.[30,31] Applying such a nanostructure in the organic material of the moth-eye with a refractive index similar to the BK7 glass, the increase in transmission is merely 4.2 % at normal incidence and about 10 % at 60 degree angle of incidence which are values that do not improve the transmission in principle. However, moths are nocturnal insects and the avoidance of any light reflex from their eyes - seen from any direction within a wide opened cone - seems to be the main effect for the natural development of the moth-eye nanostructure.[32] In this sense, the application of a nanostructure as AR 'coating' gives an essential advantage over thin-film coatings with respect of lower reflection at higher angles of incidence or higher cone angles of the incident light beam.

The single quarter-wave optical thickness solution for an AR coating is in any case a low-cost solution and the standard thin-film application are still the single layer of MgF_2 and the 4-layer Quarter-Have-Quarter Design.[33] Broadening the AR region in the visible wavelength region by homogeneous thin films is also a standard application if the layer number is restricted to 10 or 11 layers. AR designs with essential more layers have been restricted to theoretical discussions though they offer excellent broadband AR characteristics.[7,8] At AR designs for high-index substrates as ZnSe, Ge and Si, homogeneous thin films as step-down application of an inhomogeneous index-profile have been applied successfully since the early 1980s.[7,34,35] Particularly, a so called 'flip-flop' solution has been manufactured as nearly perfect simulation of an index-profile by application of pairs of very thin layers of an high-index and a low-index material.[36] The application of Si as the material most used in the semiconductor industry and the techniques applied for the manufacturing of semiconductor electronics give the opportunity to apply 'nanostructures' for AR designs on the Si surface, directly.[19,37] But these nanostructures were not the brilliant result of human research, they have been copied from the above-mentioned moth-eyes. Pushed by the intense research and development in the field of photovoltaic, moth-eye nanostructures have been analysed and applied by different techniques in AR designs for the visible and the near infrared spectral range.[38-40] Additionally, AR coatings made by nanostructures have been applied successfully also on plastic substrates, light-emitting diodes and fibre communication optics.[41-45]

The single quarter-wave AR design usually does not fulfil the requirement on a broadband AR coating regardless of which refractive index modification the transition region at the substrate surface is made. As mentioned-above, gradient-index AR solutions and their approximation by homogeneous multilayers are the basics for broadband and omnidirectional AR designs both at the visible and in the infrared spectral range. But this 'broadening' of both the spectral range and the angle range is possible only by increasing the overall thickness of the coating.[46] Consider the transition region used for the application both of thin films and nanostructures as AR coatings, we can state generally as a 'thickness principle': The broader the spectral AR range shall be the thicker the AR transition region has to be. But the index profile in dependence of the thickness of this transition region has to quantized in multiples of the quarter-wave optical thickness defined by the wavelength range of the AR design.[47] Applied on nanostructures this principle requires high aspect ratios as the ratio of the height of the structures to their width at the basis. In this sense, both the spectral and the angle performance of nanostructures as AR coatings are determined and restricted by the used technology to produce, e.g. 'nanopillars' on GaN, 'nanowires' on Si, or 'sponge-like' structures on Zeonex.[25,27,48] To overcome these problems determined by technological difficulties and not by the AR designs themselves, the idea was to combine homogeneous thin films deposited on the substrate with a nanostructures on top of them.[49] This concept combines the technological advantages of the application of homogeneous thin films with the 'optical' advantages of nanostructures.[50]

Summarizing this general discussion we can draft the following straightforward approach to design an AR 'coating' as a mixture of thin films and nanostructures:

1. Set the residual reflectance R_0 at the reference wavelength λ_0 slightly higher than zero. This gives a 'target index' n_T instead of the refractive index of the incident medium.[47]
2. Choose any step-down index profile between the substrate index and the target index in p steps what results in (p-1) new refractive indices.[51]
3. Set the optical thickness of each step equal to $\lambda_0/4$.
4. Check the new refractive indices next to the substrate for available materials to apply them for homogeneous thin films or choose homogeneous Herpin-index thin-film equivalents by suitable high-index and low-index materials.[52]
5. Apply the remaining refractive indices or at least the outermost one by an effective index and choose a realizable nanostructure for this index.
6. Set the lateral dimensions of the nanostructures equal or less to the lower wavelength of the spectral AR range and the height of the structure according to the quarter-wave thickness and the number of steps that involve effective indices.
7. Calculate the required filling factors for the different steps and approximate any way the step-profile of the real structure for simulation and discussion.

4. THE INDEX PROFILE AND THE REFLECTIONLESS POTENTIAL

Broadband AR coatings require an AR transition region with an optical thickness of at least two but typically 3 to 5 quarter-waves. In principle, the multiple of the quarter-wave equals the number of steps of the index profile and both nanostructures and inhomogeneous thin films apply a step-down profile from the substrate index to the refractive index of the incident medium. Such a profile involves the advantage that the reflection does not increase higher than the reflection of the uncoated substrate, abandoned the effects that occur at nanostructures with respect to their physical dimensions. Considering homogeneous thin films for AR designs in the visible spectral range particularly, broadband AR range have been reaches only if the refractive index follows a step-up/step-down profile but outside the AR spectral range the reflection jumps up to essential higher values than it occurs at the uncoated substrate.[53] Generally, and known since nearly 50 years, the spectral range for an AR application using a step-up/step-down profile is only half the spectral range of direct step-up profiles.[54,55] However, the so-called multi-cycle or multi-cluster AR designs substitute also a step-down index profile of unavailable refractive indices and inside the AR spectral region the reflection differs by the number of cycles or by the profile type, e.g. the so-called maximally flat designs do not show any ripple in the spectral characteristic and Chebychev designs show equal ripples.[56,57,37]

There are also Quintic, Gaussian, and other profiles and it has been discussed what profile gives the best omnidirectional AR quality.[58] However, this is in any case a theoretical discussion because on the one hand the index profiles of nanostructures are primarily defined by the manufacturing method and on the other hand stochastic structures yield acceptable practical results but they can be described by simply models only, e.g.[31,40] In this sense, it is reasonable to remember an old solution called 'reflectionless potential' and/or to study the latest theoretical approaches concerning squeezing devices based on transformation optics.[59-61] We have designed such a 'super' AR coating (SARC) for the visible and near infrared spectral range on a BK7 glass according the straightforward approach given in Section 3. We applied a special step-up/step-down index-profile based on only one vanishing solution of the reflectionless potential described in ref. [60]. Figure 1 shows the refractive index profile, Figure 2 the spectral characteristic, and Figure 3 the angle characteristic. Between 400 nm and 800 nm and 0 degree and 60 degree, the reflectance is not higher than 1 %. We assume that this profile can be realized by 5 homogeneous thin films next to the substrate and two nanostructures with effective indices next to the air. Without loss of generality we assume for this first design constant refractive indices and act on the assumption that the nanostructure fulfills all the requirements on its physical dimension. The SARC was a first approach and it seems possible to get designs much better than this one. We will present further solutions and first practical solutions of this SARC-type in a further paper.

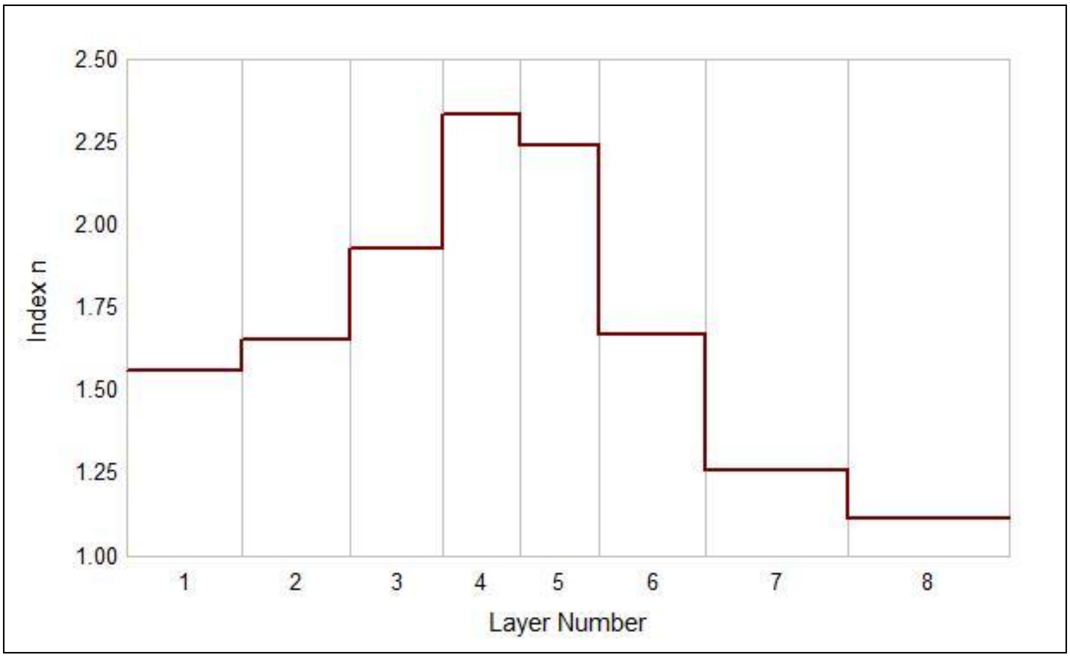

Figure 1: Refractive-index profile of the SARC-1. The abscissa is in Nanometres and sub-divided in the layer number. The refractive indices are constant versus the wavelength and the values are 1.52 for the substrate, 1.56, 1.93, 2.33, 2.24, 1.67, for the homogeneous layers 1 to 5, 1.26, and 1.11 for the last two layers 7 and 8 as effective indices, and 1.0 for air as the ambient medium.

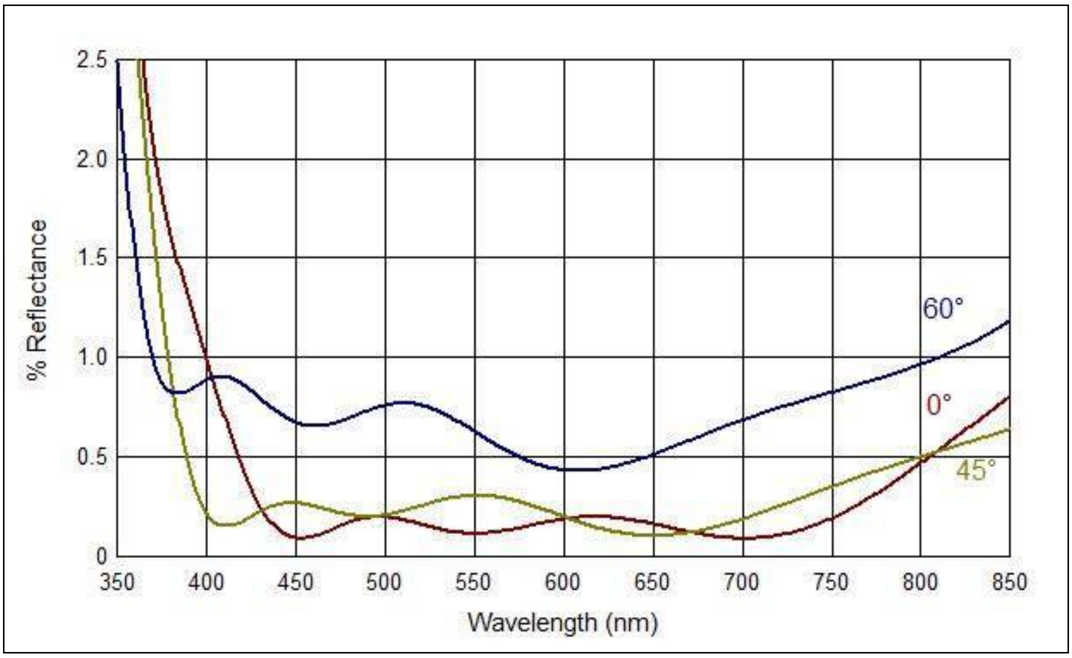

Figure 2: Reflectance vs. wavelength plot at 3 angles of incidence for random polarization as spectral characteristic of SARC-1.

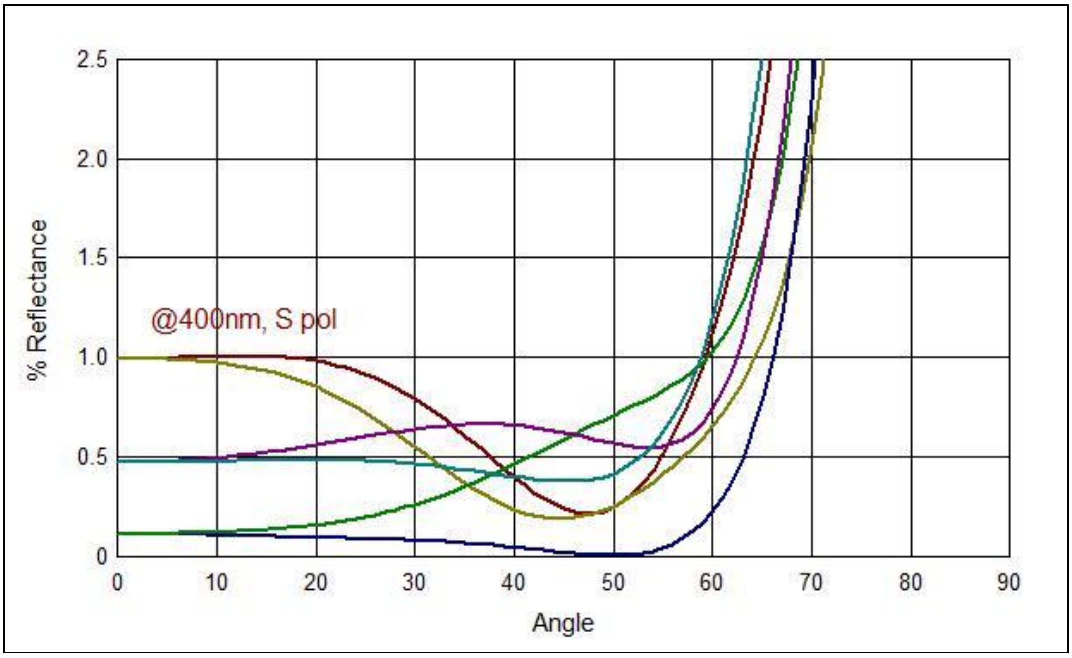

Figure 3: Reflectance vs. angle of incidence plot at three wavelengths 400 nm, 550 nm, 800 nm, and for S and P polarization as angle characteristic of SARC-1. The maximum at 1 % at 15 degree is indicated as the spectrum for S polarisation at 400 nm.

5. CONCLUSION

We conclude that the intensive discussions both on multi-cluster thin-film AR coatings and AR moth-eye nanostructures during the last 10 years result in a dead heat in the ‘game nanostructures versus thin films in the design of antireflection coatings’. The successful manufacturing of an AR coating for a special application may decorate only a ‘temporal or local’ winner in this game. Standard optics is still coated as 50 years ago but also special broadband AR designs with up to 20 homogeneous layers are applied. Developments for AR coatings on plastics, large area displays, solar cells, semiconductor chips, fibre-ends, LEDs, and both deep UV and THz optics have been pushed by the idea to reduce reflection covering the natural moth-eye principle in a technical realizable nanostructure. Theoretical base for any AR coating both with homogeneous thin films and with nanostructures is the application of a transition region between the bulk substrate und the ambient medium that is characterized by a gradient-index profile. This fact gives the background for the combination of homogeneous thin films and nanostructures in one AR coating and a straightforward method is presented to realize this concept. Remembering the known principle of reflectionless potentials it seems possible to produce broadband omnidirectional antireflection coating without any novel design principle. As an example, a special AR design for the visible and near infrared range on BK7 glass is presented which shows residual reflectance < 1 % in the wavelength range 400 nm to 800 nm und the angle of incidence range from 0° to 60°.

Moth-eye nanostructures have been covered from the nature and AR designs for omnidirectional incidence do not require new principles. In this sense, it seems that there is not any remarkable ‘white area’ that challenges somebody to search for a new AR design principle. Theoretically, the AR problem should be solved completely, but practically, a big run has been started presenting cheaper and more stable methods to manufacture AR designs made of thin films or of nanostructures or of a mixture of both.

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