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Design and Preparation of Antireflection Films on Glass Substrate

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Abstract

Antireflection coatings (ARC) have been modeled and prepared experimentally for optical and electro-optical applications in the visible spectrum (0.4–0.7 μm) to reduce reflections from the surface of glass. Reflection of BK7 bare glass substrate varies from 3.5 to 6.0% over the desired spectrum. This value is reduced reasonably by integrating multilayer thin films, based on dielectric materials, behaving as ARC. Three different three-layer configurations are deposited on glass substrate. The reflection profiles of theoretical and experimental curves are compared and analyzed to demonstrate the use of these multilayered structures. The physical parameters and deposition conditions of the three configurations are optimized before multilayer deposition. The prepared samples have shown very good results.

Key Words: Antireflection coatings, multilayer structures

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1. Introduction

Antireflection coatings have had among the greatest impact on optics; and even today, in sheer volume of production, they still exceed all other types of coatings [1]. In some applications antireflection coatings are used to reduce surface reflections, while in others, to also increase transmittance. As it is commonly modeled, radiation incident upon the surface of an optical material is considered composed of reflected, transmitted, absorbed and scattered fractions; and the fraction of available energy that is distributed between the reflected and transmitted components is determined by the indices of refraction. Antireflection coatings can range from a single layer having virtually zero reflectance at just one wavelength, to a multilayer system of many layers having virtually zero reflectance over a wide spectral range.

Investigations of the optical properties of visible and infrared optical materials for preparing reflective and antireflective (AR) coatings have long been developed for a multitude of applications and have significantly increased over the past few years. This has been facilitated by the advances and growth in computing facilities, coupled with advanced data acquisition and software capabilities [2]. AR coatings have many applications, e.g. lasers, eye glasses, lenses, solar cells, IR diodes and spectral filters, etc. [3]. The antireflection coating depends for its operation on more or less complete cancellation of light reflected at both the upper and lower of the two surfaces of the thin film. Although many recipes have been proposed for antireflection coatings, still the research is going on for the optimization of material properties and deposition conditions to come up with exciting results.

In present work, we have modeled three different configurations of three-layer antireflection coatings on BK7 glass substrates. Theses configurations are optimized for their performance by varying the refractive index, thickness (away from the traditional quarter–half wave-thick configurations) and deposition conditions of individual layers. These modeled configurations are then deposited experimentally under vacuum and are tested for their performance. Four materials, namely magnesium fluoride, cryolite, alumina and zirconia have been used to deposit these configurations. All of these materials are low cost and common to the thin film industry, with additional potential applications in thin film multilayered structures [4]. It is usually recommended to deposit oxides such as zirconia and alumina by high energy process to make their layers less absorptive due to oxygen deficiency [5–9]. We have employed the most simple deposition technique (resistive heating) and produced layers with negligible absorption, as demonstrated in the analysis of the various configurations. The scope of this research work is to produce wide-band antireflection coatings with minimum number of layers and materials using a simple deposition technique, so as to be cost effective for local industry.

2. Theory

The simplest antireflection coating is a single layer deposited on a substrate [10]. The antireflection property of this layer depends on the cancellation of light at the upper and lower of the two surfaces. Denoting the refractive indices of air, film and substrate as n_o, n_1 and n_s, then the intensity of the radiation reflected at the upper and lower surfaces of the coating should be equal in order to cancel each other. This condition means that the ratios of the refractive indices at each boundary should be equal, that is $n_o/n_1 = n_1/n_s$, with film thickness, $n_1 t_1 = \lambda/4$ [11]. This configuration will give only one minimum in the reflectance profile. For more minima, more layers are required. The same theory is used to calculate the expressions for two- and three-layer antireflection coatings [12]. Similarly, multiple layers are used to achieve more minima in the reflectance profile for broadband antireflection coatings [13, 14].

We have modeled and prepared antireflection coatings for use on glass substrates. These coatings are modeled at a design wavelength of $\lambda_o = 0.510\ \mu$m.

Matrix calculations determine the spectral transmittance and reflectance profile for multilayered structures on a substrate. The loss-free transmittance and reflectance for the multilayer assembly can be calculated from this product matrix by [1] via the equations

$$T_q = \frac{4n_s n_o}{(n_o AA + n_s DD)^2 + (n_o n_s BB + CC)^2} \tag{1}$$

$$R_q = \frac{(n_o AA - n_s DD)^2 + (n_o n_s BB - CC)^2}{(n_o AA + n_s DD)^2 + (n_o n_s BB + CC)^2} \tag{2}$$

The reflectance, transmittance and absorptance are then related by the relation $R + T + A = 1$. The solution of this matrix theory is a laborious job for multilayer coatings. Based on the matrix theory, we have developed a software program to design and simulate the performance of multilayer coatings. The software is developed in visual C++. Basic parameters like individual layer thickness, design wavelength, refractive index of the layer, extinction co-efficient, incident angle and spectral range are given as input to the software. The program then generates the out put in tabular and graphical form in terms of transmittance, reflectance and absorptance profiles. These input parameters can be varied to optimize the results and to carryout necessary variations in the design. The detail working of the software is given in [1].

3. Coating Materials

We have used alumina (Al_2O_3), zirconia (ZrO_2), cryolite and magnesium fluoride (MgF_2) for the preparation of multilayer AR coatings. Oxides are an important class of coating materials, because they generally form hard, abrasion resistant, and chemically and environmentally stable films with a good variety of refractive indices and spectral ranges of high transmission. Al_2O_3 is a medium index, low absorption material usable for coatings in the near-UV (<300 nm) to IR (~5 μm) regions. Completely oxidized alumina films are absorption-free over the range above 0.3 μm to at least 5 μm. Slight dissociation and oxygen loss occurs during evaporation. Adhesion is good to glass, most other oxides, some polymers, and to metals such as aluminum and silver [15]. Typical applications include near-UV laser AR and dielectric mirror designs.

ZrO_2 is a high index, low absorption material usable for coatings in the near UV (300 nm) to IR (~8 μm) regions. Typical applications include near UV laser AR and dielectric mirror designs. Zirconia is among the oxide compounds whose tendency for inhomogeneous index gradients can be reduced by the introduction of foreign oxides. Additives such as TiO_2, SiO_2, MgO_2modify the crystalline growth behavior [16, 17].

MgF_2 is still the dielectric material with the highest consumption. This is based on a combination of material properties like low refractive index and film durability [18]. It is insoluble and hard if deposited on hot substrates at reduced pressure around of 10^{-6} mbar. The temperature of the substrate should be at least 250 °C for high film hardness and density on glass or zinc sulfide substrates [14].

Cryolite is also a low index material with typical application in multilayer structures behaving as optical filters [19]. Cryolite decomposes to AlF_3 and NaF during evaporation. The Composition of deposited layer varies depending on evaporation temperature. It is of advantage to evaporate at low pressures at least of the order of 10^{-5} mbar with high substrate temperature in the range 250–300 °C [14]. We have used this material along with MgF_2 as a low index material for our ARC's.

4. Experimental Details

Leybold A700Q deposition system with oil diffusion pump and liquid nitrogen traps have been used to carryout the deposition of thin films of MgF2, Al_2O_3,ZrO_2 and cryolite on glass substrate. The substrate was given an ultrasonic bath in isopropyl alcohol for 30 minutes followed by glow discharge cleaning inside the chamber before the evaporation of films. The method of evaporation is resistive heating with high current source. These films are deposited at a pressure of the order of 10^{-6} mbar using liquid nitrogen. The substrates were heated in the temperature range 250–300 °C to form a hard, stable surface with good adhesion and free of film voids [20]. The substrates were also given planetary movement during the deposition process to achieve uniform thickness. Extra care was exercised to avoid splashing or spitting of material before and during the deposition process. For this, evaporation boat current was varied very slowly until the evaporation was started. Once the evaporation was initiated, the boat current was increased to the desired rate. At this point mechanical shutters were removed for deposition to commence.

The experimental conditions such as evaporation rate, film thickness, substrate temperature etc., were optimized by depositing the layers individually by carrying out several runs (as per design parameters) and testing them before making the multilayered structures. Thickness was controlled with thickness monitor during the deposition process and was then checked by Å-scope interferometer with analytical verification [21]. The coating materials were loaded simultaneously to carry out the deposition process without breaking the vacuum. The reflection profiles of the individual films and multilayer structures were obtained by using Ocean Optics SD2000 Spectrophotometer.

5. Results and Discussion

Three different configurations based on cryolite (configuration 1), MgF_2 (configuration 2) and three materials (configuration 3) are deposited on BK7 glass substrate to reduce the magnitude of reflection from the substrate. Measured reflection of bare BK7 glass is shown in Figure 1. There is variation in the value of reflection from 6% to 3.5% over the given spectrum (0.4–0.7 μm). Measured reflection is quite high at short wavelengths as compared to the theoretically computed curve (Figure 1). All the three configurations are optimized for their thickness, indices of the layers and deposition conditions to improve their performance.

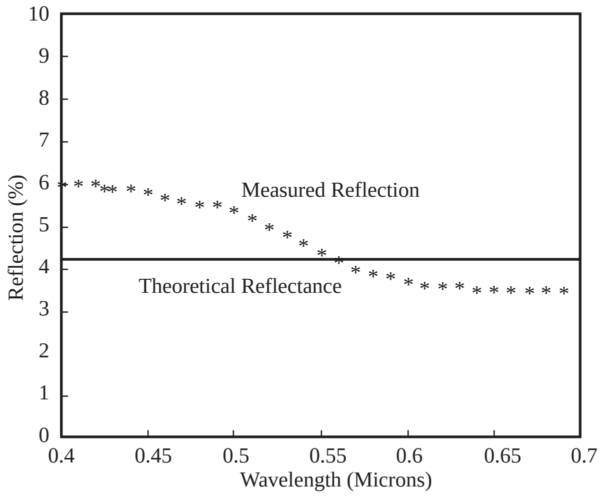

Figure 1. Reflection profile of bare BK7 glass substrate.

Configuration 1. This configuration consists of three layers of cryolite and Al_2O_3 in the order air/cryolite/Al_2O_3/cryolite/substrate. These three layers were then deposited step-by-step starting from a single layer. In-situ cleaning was carried out by glow discharge process before the deposition of the films. Table 4 shows the deposition conditions used for this configuration.

The theoretical and measured reflections of single double and three layer samples are shown in Figures 2, 3 and 4. It can be seen from these figures that experimental and theoretical curves for single layer cryolite on glass are very close to each other. The reflection profile of the experimental curve is very similar to the theoretical one, except that it shows higher reflection values than those expected from theory. Further, minimum value of reflection is observed at the design wavelength in both curves. In case of the single layer, only one minimum in the reflection curve could be achieved as expected [21]. However, increasing the number of layers would result in more than one minimum position in the reflection profile. Figure 3 shows the theoretical and experimental profiles of a two layer (cryolite/Al_2O_3) coating on glass. Again, minimum reflection is observed at the design wavelength. The experimental curve is extremely close to the theoretical curve in the range 0.49 μm to 0.7 μm. The experimental curve in general exhibits much improved performance as compared with the single layer configuration. However, the reflection values at the shorter wavelengths are slightly higher than reflections associated with the medium and long wavelength regions. Reflection profile of the 3 layer configuration is shown in Figure 4. The configuration is basically a sandwich organization, in which Al_2O_3 is sandwiched between two layers of cryolite. For Al_2O_3, higher current was employed once the evaporation started, accompanied by low deposition pressure and high substrate temperature to avoid adsorption of water in the film. The experimental curve exhibits better reflection values compared with the theoretical curve in the long wavelength region. However, high reflection values are observed in the short wavelength region. The magnitude of reflection at the short wavelength is higher compared to the two-layer configuration, but the trend toward increase in the reflection at shorter wavelength is similar to the two-layer configuration. Further, experimental curve is seen to follow the same trend exhibited by the theoretical curve at the shorter wavelength. But the theoretical curve is exhibiting nearly zero reflectance in the region between 0.45–0.575 μm. The higher reflection values at the shorter wavelengths are attributed to small variations in thickness and refractive index of the materials, leading to higher reflection values. Flattening effect is prominent from 0.5 μm and beyond. This effect will give rise to a broad and flat transmission profile; but at wavelengths shorter than 0.5 μm the rise in reflection tends to affect the flattening effect of the curve over the entire spectrum. Two- and three-layer structures with cryolite have been found to be more sensitive to the reflection profile of the substrate. These structures have shown high reflection values at short wavelengths of the spectrum compared to mid- and long wavelengths. This behavior is very similar to what is observed for the measured reflectance profile of the bare glass substrate. Design data for single, two- and three-layer film structures is given in Table 1. Refractive index values are calculated by Manifacier method [22] for each individual film using following relation:

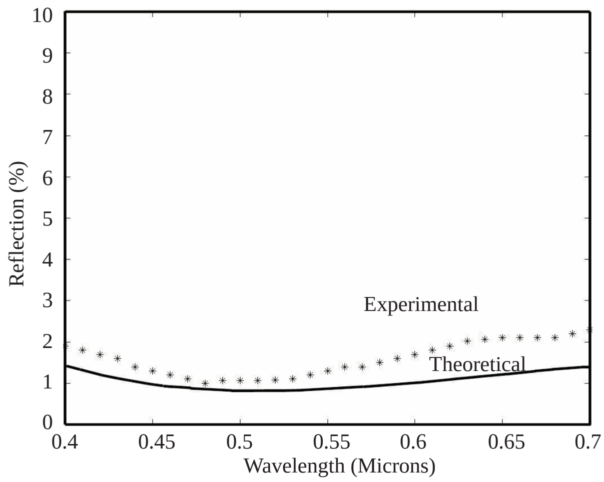

Figure 2. Reflection profile of single layer cryolite coating on glass substrate.

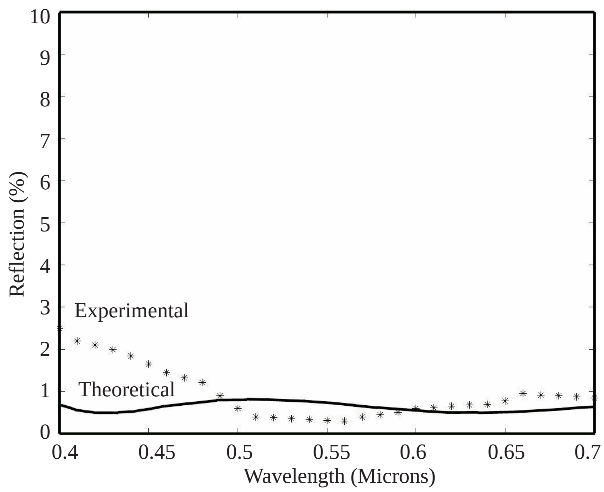

Figure 3. Reflection profile of two layer Cyolite/Al_2O_3 coating on glass substrate.

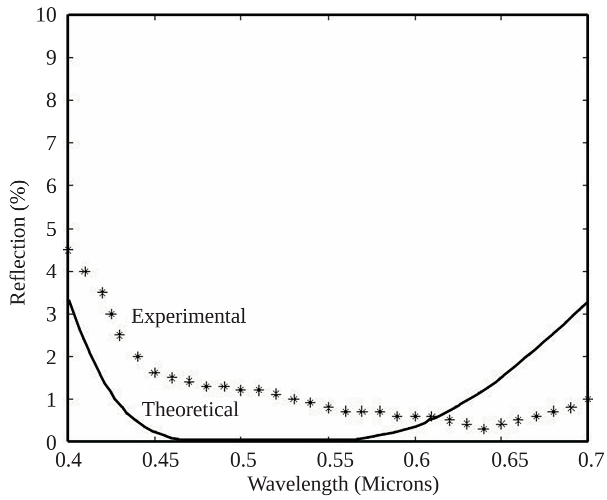

Figure 4. Reflection profile of three layer cryolite/Al_2O_3/cryolite coating on glass substrate.

$$n = \left[N + \left(N^2 - n_0^2 n_1^2\right)^{1/2}\right]^{1/2} \tag{3}$$

Where

$$N = \frac{n_0^2 + n_1^2}{2} + 2n_0 n_1 \frac{T_{\max} - T_{\min}}{T_{\max} T_{\min}} \tag{4}$$

Here, n_o is the refractive index of the incident medium, n_1 is the refractive index of the substrate, $T_{\max}$ and $T_{\min}$ are maximum and minimum values of the observed transmission spectrum of the layer.

Configuration 2. This is another sandwich-type configuration where Al_2O_3 is sandwiched between two layers of MgF_2, in the order Air/MgF_2/Al_2O_3/MgF_2/substrate. The design data for the configuration is given in Table 2 and the deposition conditions are given in Table 5.

Table 1. Design data for configuration 1.

Structure	Material	Refractive Index (n)	Thickness (μ)
Single Layer	Cryolite	1.35	0.0893
Two-Layer	Cryolite	1.35	0.0893
	Al_2O_3	1.62	0.1488
Three-Layer	Cryolite	1.35	0.0944
	Al_2O_3	1.62	0.0787
	Cryolite	1.35	0.1889

Table 2. Design data for configuration 2.

Structure	Material	Refractive Index (n)	Thickness (μ)
Single Layer	MgF_2	1.38	0.0873
Two-Layer	MgF_2	1.38	0.0873
	Al_2O_3	1.62	0.0744
Three-Layer	MgF_2	1.38	0.0924
	Al_2O_3	1.62	0.0787
	MgF_2	1.38	0.1848

Table 3. Design data for configuration 3.

Structure	Material	Refractive Index (n)	Thickness (μ)
Three-Layer	MgF_2	1.38	0.0996
	ZrO_2	2.05	0.1341
	Al_2O_3	1.62	0.0849

Table 4. The deposition conditions used in the formation of films for Configuration 1.

	Condition	Value
Glow Discharge	Chamber pressure during glow discharge	8×10^{-6} mbar
	Time for glow discharge	8 min
Cryolite Deposition	Chamber pressure during deposition	5×10^{-6} mbar
	Evaporation time	1 min 43 s
	Substrate temperature	280 °C
Al_2O_3 Deposition	Chamber pressure during deposition	6×10^{-6} mbar
	Evaporation time	1 min 3 s
	Substrate temperature	260 °C

Table 5. The deposition conditions used in the formation of films for Configuration 2.

	Condition	Value
Glow Discharge	Chamber pressure during glow discharge	6×10^{-6} mbar
	Time for glow discharge	4 min
MgF_2 Deposition	Chamber pressure during deposition	5×10^{-6} mbar
	Evaporation time	2 min 13 s
	Substrate temperature	275 °C
Al_2O_3 Deposition	Chamber pressure during deposition	6×10^{-6} mbar
	Evaporation time	1 min 3 s
	Substrate temperature	260 °C

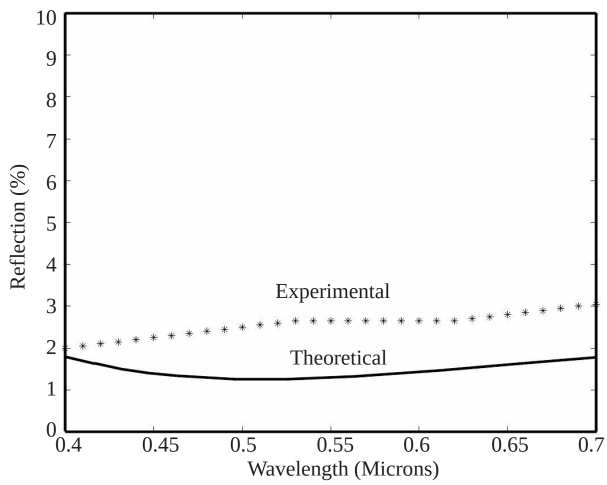

Figure 5. Reflection profile of single layer MgF_2 coating on glass substrate.

Same deposition conditions were used to evaporate Al_2O_3as for Configuration 1. The single layer of MgF_2 on glass shows a typical reflection curve with reflection values averaging around 2.5% in the given spectrum (see Figure 5). The two-layer configuration (MgF_2/Al_2O_3/glass) exhibits a sharp dip in reflection near design wavelength. However, the reflection tends to increase considerably on either side of the design wavelength, as shown in Figure 6. It appears that two minima, very close to each other, have appeared in this configuration near design wavelength. High reflections as exhibited in the two layer structure have been considerably reduced in three layer (air/MgF_2/Al_2O_3/MgF_2) structure. As shown in Figure 7, major portion of the theoretical and experimental curve is in close agreement with each other. There is a deviation in the two curves on the two extremes of the spectrum. However, experimental curve exhibits a far better reflection compared to the theoretical curve. Maximum reflection value as observed in this case is 1.6% at 0.420 μm. The measured average reflection for this configuration is less than 1% over the entire spectral region.

Configuration 3. The third configuration employs three different materials. A high index material ZrO_2 is used as a central layer surrounded by MgF_2 and Al_2O_3in the sequence air/MgF_2/ZrO_2/Al_2O_3/substrate. Design data for this configuration is given in Table 3. Higher index layer (ZrO_2) is made thicker compared to the other layers to obtain consistent properties. Further, the ZrO_2 layer is deposited at high substrate temperature to obtain a void-free and durable film [23]. The power is varied slowly for even melting of the material and to avoid spitting. The film was deposited at high currents once the evaporation started. Same deposition conditions as used in previous configurations for MgF_2 and $Al_2O_3$are employed. The deposition conditions are shown in Table 6.

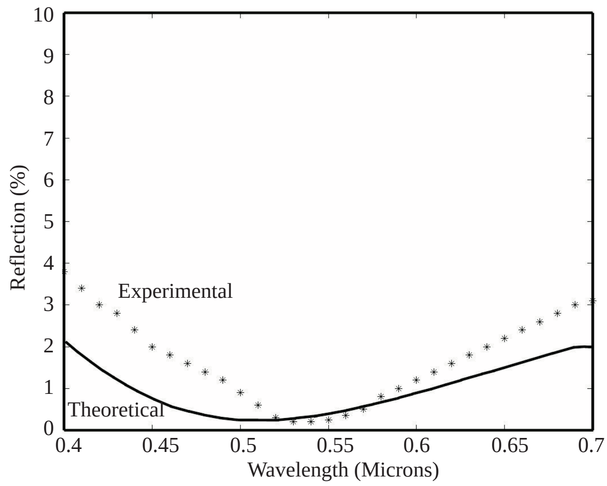

Figure 6. Reflection profile of layer MgF_2/Al_2O_3 coating on glass substrate.

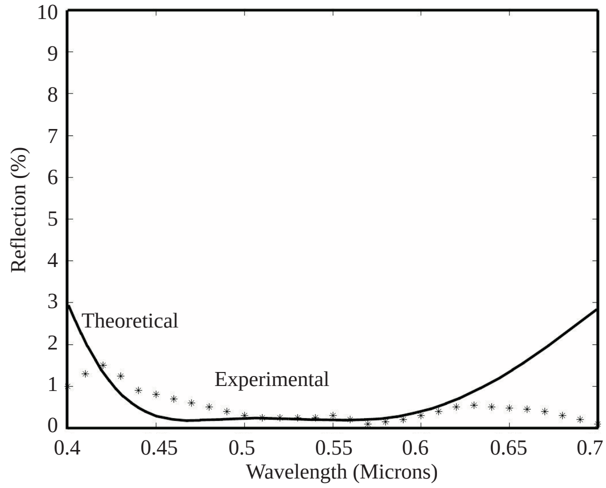

Figure 7. Reflection profile of three layer MgF_2/Al_2O_3/MgF_2 coating on glass substrate.

In the previous two configurations low and medium index materials were used to prepare antireflection coatings. In this configuration, low, medium and high index materials are used to form three-layer structure on a glass substrate. The refractive index of the materials are varied from low to high and then to medium, starting from air. High refractive index material is used, which is the thicker of the three, to act as a curve widening layer, using the concept of absentee layer [24]. Figure 8 shows the reflection profile of theoretical and experimental curves. It can be seen that the two curves are in close agreement with each other in the central portion of the spectrum, but are deviating from each other in the short and long wavelength regions. However, the experimental curve is following the same trend as that of the theoretical curve.

In all the three configurations, it should be noted that reflection values for the experimental curves are below 1% , especially at the design wavelength. Configuration 2 has exhibited a very good performance in terms of reducing reflection from the surface of the substrate over the entire spectrum. Higher values of reflection at the short wavelength portion of the spectrum for the configuration based on cryolite is attributed to the slight variation of refractive index and layer thickness along with the substrate properties. An in-depth study is planned to analyze structural and interface properties of the three configurations through x-ray diffraction (XRD) and scanning electron microscopy (SEM) techniques [25].

Table 6. The deposition conditions used in the formation of films for Configuration 3.

	Condition	Value
Glow Discharge	Chamber pressure during glow discharge	8×10^{-6} mbar
	Time for glow discharge	8 min
MgF_2 Deposition	Chamber pressure during deposition	5×10^{-6} mbar
	Evaporation time	2 min 13 s
	Substrate temperature	275 °C
ZrO_2 Deposition	Chamber pressure during deposition	9×10^{-6} mbar
	Evaporation time	12 min
	Substrate temperature	290 °C
Al_2O_3 Deposition	Chamber pressure during deposition	6×10^{-6} mbar
	Evaporation time	1 min 3 s
	Substrate temperature	260 °C

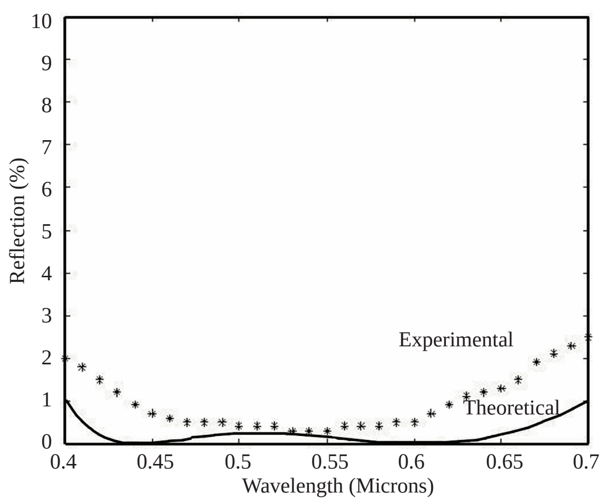

Figure 8. Reflection profile of three layer $MgF_2/ZrO_2/Al_2O_3$ coating on glass substrate.

6. Conclusions

From this research work we conclude that usable antireflection coatings can be formed by using conventional dielectric materials as film layers. Three configurations have been deposited after optimizing the physical parameters and deposition conditions. Configurations 2 and 3 have given highly promising results which support their use as ARC for optical components. Configuration 1, however, has shown higher reflection values at short wavelengths, which are appreciably low compared to bare glass reflection.

Structural studies employing XRD and SEM would give a better insight of the structure and interface being formed. This would help us in further optimizing configuration 1 and would lead us to develop a better understanding of the three configurations at micro level. Nevertheless, these three configurations can be applied to optical components to get wide transmission bands in the given spectral region.

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