

Deposition of MgF₂ and TiO₂ Multilayer for Ar⁺Laser Front Mirror

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Abstract

Introduction: Laser anti-reflection coatings (LAR) are designed for the most effective anti-reflection results at a specifically defined wavelength. As such, they are ideal for applications involving lasers or other monochromatic light sources. Composite MgF₂ (low refractive index) and TiO₂ (high refractive index) films are fabricated by reactive ion-assisted coevaporation deposited on BK-7 glass substrate.

Aim: The purpose of the present paper is to produce a high reflection mirror for Ar⁺ laser, multilayers of MgF₂ (low refractive index) and TiO₂ (high refractive index) have been deposited on BK-7 glass substrate.

Material and method: A vacuum system capable of reaching pressures with of 10⁻⁷ mbarn(Balzers) and an optical thickness monitor(GSM-420 Balzers), as well as vibrating quartz crystal thickness monitor were mounted in this vacuum system to be able to check the obtained results. Substrate was circular glass with 20mm diameter and 1 mm thickness(BK-7 glass). Pure research grade MgF₂ and TiO₂ was used and both evaporated the from tantalium crucible separately, at pressures about 2-3×10⁻⁵ mbar. MgF₂ was coated with 1 nm/s deposition rate and for deposition of TiO₂ partial pressure of 5×10⁻⁵ mbar was used and coating rate was 0.5 nm/s.

Results: Substrate temperature play an important role on refraction index and packing density of thin films has affect on R value. After annealing although reflection power increased by 5%. Increasing of R for $\lambda=500nm$, by 5% but at other wavelength $\lambda < 400nm$ and $\lambda > 600nm$ (outside the spectral band width), also reflectivity rises 20 to 30 percent as well.

Conclusion: After eleven thin film layers of $\lambda/4$ (G/(HL)⁵/TiO₂/Air), mirror with 95% power of reflection over spectral band width of 160nm and nearly 5% of transmission was produced. These parameters are good enough for making front mirror of Ar⁺ laser.

Keywords: Thin films, MgF₂/TiO₂ Multilayer, Antireflection Coating, Refractive Index.

Introduction

To produce high reflective mirrors for laser and optical devices, metal coating might have a good and high reflection power over wide range of wavelength (Al,Ag,Au, Cu,...), but they have drawback as well. Transmission of these layers due to high reflecting and absorption of them is zero or it is very small and their optical characteristics are

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varying with time and conditions. The broadband multilayer coatings reflect or transmit over a broad range of wavelengths with performance exceeding that of single layer coatings. When compared at specific wavelengths, a broadband multilayer coating can outperform a broadband single layer coating by a factor of ten. MgF₂ is one of the most common materials with low refractive index and is used for anti-reflection coating. MgF₂ is used together with a substrate which has high reflection index like ZrO₂, SiO₂,..., specially with TiO₂. Thin films of TiO₂ with high reflectivity over wide range of spectrum (up to 10-12 μm if its film be thin enough), has made it suitable for making dielectric multilayer mirrors. For reflection coefficient of TiO₂ various values have been reported (between 1.9 to 2.6), which is due to deposition method and rate of it, as well as substrate temperature and partial pressure of oxygen. Especially TiO₂ shows different phases in various temperatures and also its crystallography direction of its growth varies the refractive index. For these reasons in this experimental work all quarter wave ($\lambda/4$, $\lambda = 550\text{nm}$) layers of MgF₂ and TiO₂ were deposited at optimum coating conditions which was obtained after various investigations on deposition rate, substrate temperature, pressure, ..., etc. For example, a magnesium fluoride (MgF₂) single layer anti reflective coating might exhibit reflectivity of about 2% at 550nm, whereas a multilayer coating designed for the same central wavelength of 550nm might exhibit 0.2% reflectivity.

Narrowband multilayer coatings can be designed to outperform broadband coatings at specific wavelengths. Narrowband AR coatings are often called V-coats. The terminology originates in the appearance of graphs that plot reflectivity against wavelength. A V-coat can perform with 0.1% reflectivity at a specific wavelength, but its reflectivity rises quickly for shorter and longer wavelengths. The graph of its performance looks like the letter "V." Hybrid coatings consist of dielectric layers deposited upon a metallic base layer. Water absorption by optical films causes swelling of the thin film, changes the refractive index, and overall shifts in the spectral transmittance. Water absorption causes the material to become more absorbing. It is not particularly hygroscopic, but it has voids. A void allows water vapor to penetrate low-density films and gap between the columnar grains by capillary action.^[1] Additionally, if the MgF₂ film contains crystallites in its structure, their grain boundaries can cause an absorption tail for the refractive index below the material's band gap. Because of the optimization done through the merit function used, TiO₂ or Cr had been initially chosen as the high reflective index (HR) materials.^[2,3] Antireflection from a multilayer structure requires a transitive material layer sandwiched between two reflective material layers. This amounts to a multilayer structure composed of a combination of alternating high and low index of refraction materials. Because the indices will alternate, reflection will occur at more than one boundary. Therefore, the superposition of all of the reflected waves needs to be taken into account. The multilayer structure a repeated high low refractive index thin film structure.^[4-6] So, exact knowledge about optical properties of MgF₂ thin film can help us for producing AR or HR thin films with good parameters.^[7,8] These reasons are caused to make a good reliable high reflecting mirror with low amount of transmittance for front mirror of lasers. For this purpose, dielectric multi-layers with two substances, with low and high refractive index are used. One of the major problem with thin films, in physical vapor deposition (PVD), specially for dielectric materials is the porosity, which usually form a columnar porosities with various diameters (12-20 nm). The residual air or water sorption in these porosities can change their optical and spectral as well as other properties. This problem can be overcome by high enough substrate temperature (depending on material and method of coating used) during and after deposition.

This article is organized as follows: In the next section, a brief description of the relevant theoretical methods is reported. Then procedure of experimental method will be

presented in section III. We discuss the errors, tabulation of the obtained reflection power, refractive index of MgF2 thin films and their variations due to substrate temperature in section IV. Finally, summary and conclusions is followed in section V.

Material and Method

Theoretical calculation of transmittance and reflectance

In optical coatings, two different kinds of graded index profiles can occur: (i) intentionally graded profile, when precisely controlled inhomogeneous optical coatings (for example, rugate filters) are produced;^[15] (ii) accidentally created in homogeneities that may arise from the instability of the deposition process. The latter may be due to different factors, including the change of microstructure of the film deposited, the contamination, the lack of control of the fabrication parameters, or the interaction of the growing film with the substrate.

In order to control the film quality it is, therefore, very important to calculate the refractive index depth profiles from in situ measurements. Bovard, inspired by the popular transmittance envelope method for the calculation of refractive index dispersion curves,^[19-22] developed an approximate method based on the transmittance envelopes of the curve transmittance-vs-time. The method used to determine the refractive index of the film at any time of the deposition process is based on the evaluation of the envelope curves connecting the reflectance minima and maxima. Following a WKBJ (Wentzel-Kramers-Brillouin-Jeffries) approximation,^[23] one can express the “optical transfer function” of an inhomogeneous thin film by the following characteristic matrix:^[24]

$$\mathcal{M} = \begin{bmatrix} \sqrt{\frac{n_{in}}{n_{out}}} \cos \delta & \frac{i}{\sqrt{n_{in} n_{out}}} \sin \delta \\ i\sqrt{n_{in} n_{out}} \sin \delta & \sqrt{\frac{n_{out}}{n_{in}}} \cos \delta \end{bmatrix} \quad (1)$$

with $\delta = (2\pi/\lambda) \int_0^d [n(z) - ik(z)] dz$ for normal incidence, where n_{in} and n_{out} are the film refractive index values near the film-substrate interface and near the surface, respectively, d is the thickness of the layer, z is the distance from the interface inside the layer, and $n(z)$ is the refractive index profile in the layer. From this characteristic matrix one can obtain expressions for the transmittance T and reflectance R of a film on a substrate:

$$T = \frac{n_s n_{out}}{n_0 n_{in}} \left| \frac{t_{in} t_{out} \exp(\delta)}{1 + r_{out} \check{r}_{in} \exp(2\delta)} \right|^2, \quad (2)$$

$$R = \left| \frac{r_{out} + \check{r}_{in} \exp(2\delta)}{1 + r_{out} \check{r}_{in} \exp(2\delta)} \right|^2. \quad (3)$$

here, $t_{out} = 2n_0/(n_{out} + n_0)$, $t_{in} = 2n_{in}/(n_{in} + n_s)$, $r_{out} = (n_0 - n_{out})/(n_0 + n_{out})$ and $\check{r}_{in} = (n_{in} - n_s + ik_s)/(n_{in} + n_s - ik_s)$. The sign over r_{in} indicates that this parameter can have both complex or real values, as one can consider the reflectance problem for both transparent and absorbing substrates. From the approximate expressions (2) and (3), one can obtain expressions for the minima and maxima of the transmittance or reflectance curves, $T(z)$ or $R(z)$. From the data available (monitored $R(z)$ or $T(z)$ values) one can then evaluate experimentally the envelopes and solve the envelopes expressions for $n(z)$ with these assumptions: (i) the value of the film extinction coefficient is low ($k \ll n$), so that only the k appearing in the exponential terms is significant; (ii) once a material is deposited, its

refractive index value is not altered by the following deposition on top of it; (iii) the substrate is considered semi-infinite, which means that the contribution to the reflectance from the back side is negligible.

Results and Discussion

Refractive index profile (Transmittance and Reflectance)

Bovard^[18] has developed a method for the transmittance which consists of solving the equations for the envelopes of the transmittance minima and maxima for n_{in} , n_{out} and the absorbance parameter $\mathcal{A} = \exp[-(4\pi/\lambda) \int_0^d k(z) dz]$. In the following we show a generalized solution, for low or high index substrates, based on his previous work^[18]. For $d = 0$, we have:

$$n_{in} = \left(N_t + \eta \sqrt{N_t^2 - n_0^2 n_s^2} \right)^{1/2},$$

$$N_t = \frac{n_0^2 + n_s^2}{2} - 2\epsilon n_0 n_s \left(\frac{T_{max} - T_{min}}{T_{max} + T_{min}} \right), \tag{4}$$

and for $d > 0$:

$$n_{out}(z) = \frac{2n_{in}n_s n_0 \epsilon \left(\frac{T_{max} - T_{min}}{T_{max} T_{min}} \right)}{n_s^2 - n_{in}^2} + n_0 \left[1 + \frac{4n_{in}^2 n_s^2 \left(\frac{T_{max} - T_{min}}{T_{max} T_{min}} \right)^2}{(n_{in}^2 - n_s^2)^2} \right]^{1/2}, \tag{5}$$

$$\mathcal{A}(z) = \frac{\epsilon}{r_{in} r_{out}} \frac{\sqrt{T_{max}/T_{min}} - 1}{\sqrt{T_{max}/T_{min}} + 1}, \tag{6}$$

with $\epsilon = -1$ for $n_{in} > |\check{n}_s|$, $\epsilon = +1$ for $n_{in} < |\check{n}_s|$, and $\eta = 1$ for $n_{in}^2 > n_0 |\check{n}_s|$, $\eta = -1$ for $n_{in}^2 < n_0 |\check{n}_s|$. The solution to the inverse problem, i.e. finding the variation of the optical parameters with depth from Eq. (3), consists of solving the following minima and maxima envelopes relations for three unknowns r_{in} , r_{out} and \mathcal{A} :

$$R_{min} = \frac{r_{out}^2 - 2\epsilon r_{out} \Re(\check{r}_{in}) \mathcal{A} + |\check{r}_{in}|^2 \mathcal{A}^2}{1 - 2\epsilon r_{out} \Re(\check{r}_{in}) \mathcal{A} + r_{out}^2 |\check{r}_{in}|^2 \mathcal{A}^2}, \tag{7}$$

$$R_{max} = \frac{r_{out}^2 + 2\epsilon r_{out} \Re(\check{r}_{in}) \mathcal{A} + |\check{r}_{in}|^2 \mathcal{A}^2}{1 + 2\epsilon r_{out} \Re(\check{r}_{in}) \mathcal{A} + r_{out}^2 |\check{r}_{in}|^2 \mathcal{A}^2}. \tag{8}$$

Here, $\Re(\check{r}_{in})$ is the real part of \check{r}_{in} . The system of Eqs. (7) and (8) offers only two equations. Therefore, to solve it we assume that at the first stage of the deposition ($d \approx 0$), the film is homogeneous ($n_{out} \rightarrow n_{in}$) and transparent ($\mathcal{A} \rightarrow 1$). Using this approximation, and solving for n_{in} , we find

$$n_{in} = \sqrt{n_0 n_s} \left(N_r + \eta \sqrt{N_r^2 - \frac{n_s^2 + k_s^2}{n_s^2}} \right)^{1/2}, \tag{9}$$

where

$$N_r = \frac{(\epsilon + 1)R_{min} - (\epsilon - 1)R_{max} + 2}{(\epsilon - 1)R_{max} - (\epsilon + 1)R_{min} + 2}. \tag{10}$$

Keeping the value of n_n constant, and solving for r_{out} and for A when $d > 0$, the calculated solutions are as follows:

$$r_{out}(z) = - \left[-\frac{1}{2} \left(\frac{B}{A} - \zeta C \right) - \frac{1}{2} \sqrt{-4 + \left(\frac{B}{A} - \zeta C \right)^2} \right]^{1/2}, \tag{11}$$

$$A(z) = 1/2 \frac{(1 - r_{out}^4)(R_{max} - R_{min})}{\epsilon r_{out} \Re(\check{r}_{in}) [r_{out}^2(2R_{min}R_{max} - R_{max} - R_{min}) + (2 - R_{min} - R_{max})]}; \tag{12}$$

where

$$\begin{aligned} A &= |\check{r}_{in}|^2 (R_{max} - R_{min})^2, \\ B &= 2\Re(\check{r}_{in})^2 (2 - R_{max} - R_{min})(2R_{min}R_{max} - R_{min} - R_{max}), \\ C &= \sqrt{2 + \frac{B^2}{A^2} - \frac{D}{A}}, \\ D &= -2|\check{r}_{in}|^2 (R_{max} - R_{min})^2 + 8\Re(\check{r}_{in})^2 [(R_{max} + R_{min})^2 + 2(1 - R_{max} - R_{min}) \\ &\quad + 2R_{max}^2 R_{min}^2 \left(\frac{1}{R_{min}} + \frac{1}{R_{max}} - 1 \right)], \end{aligned}$$

and

$$\zeta = \begin{cases} +1 & \text{if } \frac{1 + r_{out}^4}{r_{out}^2} > \frac{B}{A}, \\ -1 & \text{if } \frac{1 + r_{out}^4}{r_{out}^2} < \frac{B}{A}. \end{cases}$$

The refractive index profile $n_{out}(z)$ is then calculated from $n_{out} = n_0(1 - r_{out})/(1 + r_{out})$.

Deposition rate and physical position in the layer

When the deposition rate is constant during the deposition, the transformation of the results as a function of thickness is obvious. Unfortunately, it may not be the case for numerous experiments. If the deposition rate varies during the deposition, one can still use the fact that the optical thickness between two successive extreme is equal to a quarter waves.

Using the relation $m\lambda/4 = \int_0^d n(z)dz$ (m is an integer representing the interference order) and assuming that the deposition rate is constant between successive extreme, one obtains the following relation:

$$\frac{\lambda}{4} \approx \frac{z_2 - z_1}{t_2 - t_1} \int_{t_1}^{t_2} n(t)dt, \tag{12}$$

where z_1, z_2 and t_1, t_2 correspond to the positions of two successive extreme on the thickness (distance) and time scales, respectively. The relationship between the time and distance scales can be obtained from $t_1 = 0$ ($z_1 = 0$), where the first extremum occurs.

Error calculus

A good estimate of the error in determining the $n(z)$ and $A(z)$ values may be obtained from the calculation of the derivatives of Eqs. (4)–(6). In the case of the transmittance, assuming that n_s is known precisely and that the error of T_{max} and T_{min} are the same and not related, we have

$$\frac{\Delta n_{in}}{n_{in}} \approx \left| \frac{(n_{in}^2 - n_0^2)(n_s^2 - n_{in}^2)}{2(n_0 n_s + n_{in}^2)(n_0 n_s - n_{in}^2)} \right| \left(\frac{T_{max} + T_{min}}{T_{max} - T_{min}} \right) \frac{\Delta T}{T}, \quad (13)$$

$$\frac{\Delta n_{out}}{n_{out}} \approx \left(\frac{T_{max} + T_{min}}{T_{max} - T_{min}} \right) \frac{\Delta T}{T} + \frac{n_0 C_n}{\sqrt{1 + C_n^2}} \left[\left| \frac{n_{in}^2 + n_s^2}{n_{in}^2 - n_s^2} \right| \frac{\Delta n_{in}}{n_{in}} \right] \quad (14)$$

with

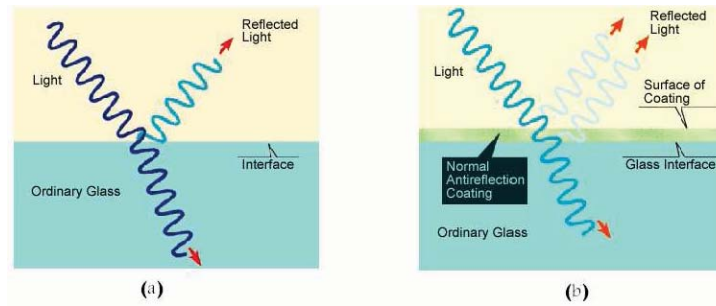


Fig. 1- (a) Roughly 4% of the incident light is reflected at a glass surface ($n = 1.52$) and (b) Reflection can be avoided by applying a coating of $n < 1.52$ (e.g. MgF₂, $n = 1.38$).

$$C_n = \left| \frac{2n_{in}n_s}{n_{in}^2 - n_s^2} \right| \left(\frac{T_{max} - T_{min}}{T_{max} T_{min}} \right). \quad (15)$$

similarly:

$$\frac{\Delta \mathcal{A}}{\mathcal{A}} \approx \frac{2\sqrt{T_{max} T_{min}}}{T_{max} - T_{min}} \frac{\Delta T}{T} + \frac{2n_0 n_{out}}{n_{out}^2 - n_0^2} \frac{\Delta n_{out}}{n_{out}} + \left| \frac{2n_s n_{in}}{n_{in}^2 - n_s^2} \right| \frac{\Delta n_{in}}{n_{in}}. \quad (16)$$

Expressions for the error in the envelope reflectance method can be found from the derivation of Eqs. (9) – (11) as functions of R and n_{in} . Optical coatings must be handled with care. The harder coatings, which are resistant to laser damage, tend to resist scratching and abrasion, but even they are softer than many glasses. The softer coatings will be marred by careless or vigorous rubbing. Varying humidity or temperature can alter the performance of a coating. Those containing water-absorbing layers exhibit sensitivity to changes in relative humidity because absorbed water changes the layer's refractive index. Temperature also affects refractive index and even thickness. In the vast majority of cases, a coating's sensitivity to the environment is small enough to be ignored. In critical or unusual applications, more sensitive coatings are placed on components that can be protected from the environment. For example, a multi-element lens system might feature hard, durable coatings on its outer elements but softer, more sensitive coatings on its internal elements. Part of the incident light is reflected by all transparent materials. This reflection is based on the abrupt change in the refractive index (n) at the interface between two media. Coating can reduce this reflection and, consequently, increase transmission. Transmission of an uncoated glass plate is approximately 92%. If both surfaces are coated with our novel films, 99% of the visible light is transmitted. These coatings are nano porous polymer films with pore sizes far below the wavelength of light. The degree of porosity easily allows the effective refractive index to be set in a range of 1.05 - 1.6. Films with such low refractive indices can be used to produce coatings with extremely high bandwidths and, at the same time, high transmission.

As with any thin film, performance depends on the incident light wavelength for two reasons. First, at other than the design wavelength, film thickness is no longer the ideal $1/4$ of λ . This is taken into account by all thin film design programs. A more subtle effect, which can be quite important, is caused by the change in refractive index of the coating and substrate with wavelength (i.e., dispersion). Only the most up-to-date computer design packages, such as those used by Melles-Griot, include this higher level of sophistication for multilayer coatings. For single-layer antireflection coatings, wavelength dependence of the coating

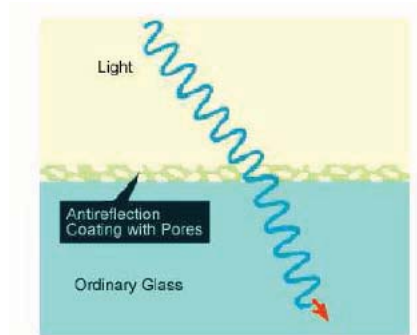


Fig. 2- Reflection (for one wavelength) can be suppressed completely only by a coating.

performance can be evaluated from analytical expressions. The first step in evaluating performance of a single-layer anti-reflection coating is to calculate the refractive index of the film and substrate at the wavelength of interest. For optical purposes, a thin film may be considered to be perfectly homogeneous. The refractive index of MgF₂, whether amorphous or crystalline, is connected to density with the Lorentz-Lorenz formula. The crystalline ordinary and extraordinary indices of refraction may be averaged for the amorphous phase. The formulas for crystalline MgF₂ are, respectively,

$$n_o = 1.36957 + \frac{3.5821 \times 10^{-3}}{\lambda - 0.14925} \quad (17)$$

and

$$n_e = 1.381 + \frac{3.7415 \times 10^{-3}}{\lambda - 0.14947} \quad (18)$$

for the ordinary and extraordinary rays, where λ is the wavelength in microns. For the average of the ordinary and extraordinary indices of refraction,

$$n = n(\lambda) + \frac{1}{2}(n_o + n_e) \quad (19)$$

The value 1.38 is the universally accepted amorphous film index for MgF₂ at a wavelength of 550 nanometers, which assumes a packing density of 100%. Real films, however, tend to be slightly porous. The refractive index of a real magnesium fluoride film is usually slightly lower than 1.38 because the packing density is rarely 100% in practice. Because it is a complex function of the manufacturing process, packing density varies slightly from batch to batch. Air and water vapor can also settle in the film and affect its refractive index. For Melles-Griot magnesium fluoride coatings, this will usually correspond to an effective reflection power between 97% and 100% of the 1.38 theoretical values. For coated surface reflectance at normal incidence, suppose that the coating is of quarter wave optical thickness for some wavelength λ . Let n_a denotes the refractive index of the external medium at this wavelength (1.0 for air or vacuum), and let n_f and n_s , respectively, denote the film and

substrate indices, as shown in the Fig(3a). For normal incidence at this wavelength, the single-pass irradiance reflectance of the coated surface can be shown to be Eq.(2), regardless of the polarization state of the incident radiation.

At oblique incidence, the situation is more complex. Let n_1 , n_2 , and n_3 , respectively, represent the wavelength-dependent refractive indices of the external medium (air or vacuum), coating film, and substrate as shown in the Fig(3b). Assume that the coating

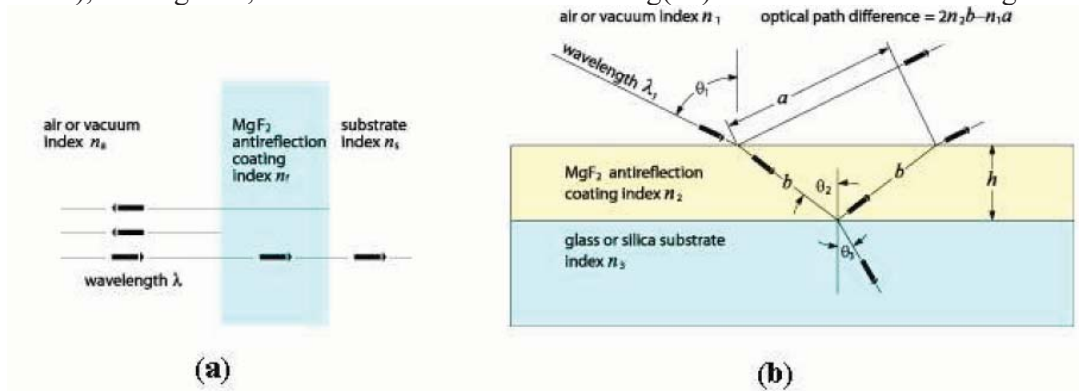


Fig. 3- (a) Reflectance at normal incidence and (b) Reflectance at oblique incidence.

exhibits a reflectance extremum of the first order for some wavelength λ_d and angle of incidence θ_{1d} in the external medium. The coating is completely specified when λ_d and θ_{1d} are known. One may then identify n_2 with the film index n_f (1.38 for MgF₂ at 550 nm). The extremum is a minimum if n_2 is less than n_3 and a maximum if n_2 exceeds n_3 . The same formulas apply in either case. Corresponding to the angle of incidence θ_{1d} is an angle of refraction in film:

$$\theta_{2d} = \arcsin\left(\frac{\theta_{1d}}{n_2(\lambda_d)}\right) \tag{20}$$

As θ_1 is reduced from θ_{1d} to zero, the reflectance extremum shifts in wavelength from λ_d to λ_n , where the subscript n denotes normal incidence. The wavelength is given by the equation

$$\lambda_n = \left(\frac{n_2(\lambda_n)}{n_2(\lambda_d)}\right)\left(\frac{\lambda_d}{\cos \theta_{2d}}\right) \tag{21}$$

Corresponding to the arbitrary angle of incidence θ_1 and arbitrary wavelength λ_1 , are angles of refraction in the coating and substrate, given by

$$\theta_2 = \arcsin\left(\frac{n_1(\lambda_1) \sin \theta_1}{n_2(\lambda_1)}\right) \tag{22}$$

and

$$\theta_3 = \arcsin\left(\frac{n_1(\lambda_1) \sin \theta_1}{n_3(\lambda_1)}\right) \tag{23}$$

Following are formulas for the single-interface amplitude reflectance for both the p- and s-polarizations:

$$\begin{aligned}
 r_{12p} &= \frac{n_2 \cos \theta_1 - n_1 \cos \theta_2}{n_2 \cos \theta_1 + n_1 \cos \theta_2} \\
 r_{23p} &= \frac{n_3 \cos \theta_2 - n_2 \cos \theta_3}{n_3 \cos \theta_2 + n_2 \cos \theta_3} \\
 r_{12s} &= \frac{n_1 \cos \theta_1 - n_2 \cos \theta_2}{n_1 \cos \theta_1 + n_2 \cos \theta_2} \\
 r_{23s} &= \frac{n_2 \cos \theta_2 - n_3 \cos \theta_3}{n_2 \cos \theta_2 + n_3 \cos \theta_3}
 \end{aligned}
 \tag{24}$$

The subscript "12p," for example, means that the formula gives the amplitude reflectance for the p-polarization at the interface between the first and second media. The corresponding irradiance reflectance for the coated surface, accounting for both interferences and the phase differences between the reflected waves, are given by

$$\begin{aligned}
 R_p &= \frac{r_{12p}^2 + r_{23p}^2 + 2r_{12p}r_{23p} \cos(2\beta)}{1 + r_{12p}^2 r_{23p}^2 + 2r_{12p}r_{23p} \cos(2\beta)} \\
 R_s &= \frac{r_{12s}^2 + r_{23s}^2 + 2r_{12s}r_{23s} \cos(2\beta)}{1 + r_{12s}^2 r_{23s}^2 + 2r_{12s}r_{23s} \cos(2\beta)}
 \end{aligned}
 \tag{25}$$

where β is the phase difference (in the external medium) between waves reflected from the first and second surfaces of the coating.

$$\beta = \frac{2\pi}{\lambda_1} n_2(\lambda_1) h \cos \theta_2
 \tag{26}$$

The cosines must be in radians. The average reflectance is given by

$$\bar{R} = \frac{1}{2}(R_p + R_s)
 \tag{27}$$

With these formulas, reflectance curves can be calculated as functions of either wavelength λ_1 or angle of incidence θ_1 . If we define, n_f , n_f^* , R and R^* refractive index of thin films in vacuum condition, after film being expose to air, reflection power of thin film in vacuum condition and after film being expose to air, respectively. We can obtain: ^[10]

$$n_f = [n_g \left(\frac{1 + \sqrt{R}}{1 - \sqrt{R}} \right)]^{1/2}
 \tag{28}$$

$$n_f^* = [n_g \left(\frac{1 + \sqrt{R^*}}{1 - \sqrt{R^*}} \right)]^{1/2}
 \tag{29}$$

where n_g is refractive index of substrate. Packing density of thin film which has porosity and sorption of water vapor residual air has occupied in it, can be calculated:

$$P = \frac{0.33 + n_f - n_f^*}{0.33}
 \tag{30}$$

Eq.(30) goes to unity when n_f and n_f^* are equal. Reflection power(R) for dielectric thin film, with quarter wave thickness $\lambda/4$ and refraction index n_1 , that coated on substrate with refractive index n_g and optical admittance($y = n_1^2/n_s$), is obtained:

$$R = \left| \frac{n_0 - y}{n_0 + y} \right|^2 \tag{31}$$

where n_0 is refractive index of incident medium(for vacuum condition and air $n \approx 1$).

For AR purposes($R \approx 0$)means finding a substance with refractive index $n_1 = \sqrt{n_0 n_g}$, so for glass substrate like BK-7 ($n_g = 1.52$), then n_1 should be 1.23. There is not such a material (dielectric) with this index and the nearest one to this value is MgF₂ with $n_1=1.38$. Therefore with one layer of this sort of material R would not be zero and even when $3\lambda/4$ is coated the amount of R might be a little higher than for $\lambda/4$ case, but again is not exactly AR($R \approx 0$).

$$n_0 = \frac{1.33n_f - n_f^*}{0.33 + n_f - n_f^*} \tag{32}$$

$$n = n_0(p) + n_p(1 - p) \tag{33}$$

where n is refractive index of thin film MgF₂ with it's content which have been absorbed, n_p is index of material which has filled the porosities in vacuum condition is equal to one and by putting this value in Eq. (33) would get $n = n_f$, which is exactly index of film in vacuum, if $n_p=1.33$ is used, then $n = n_f^*$ which is refractive index after being next to air or water vapor. Other factors that affect layer microstructure and its dependent properties are substrate surface condition. The preparation of the surface for deposition is different for glass, polymer, and semiconductor substrates. Surface conditioning consists of one or more of the following procedures: removing adsorbed molecular contamination that causes the surface to be chemically or electrically inert, generating reactive but incomplete chemical bonds in the substrate surface, increasing surface area, depositing a reactive layer that stimulates nucleation of the arriving add atoms, or establishing energetic physical bonding such as inter diffusion mixing (sometimes attributed to mutual solubility). The adhesion mechanism with most metals is the formation of an interfacial oxide bond. This is especially applicable to reactive metals such as aluminum, titanium, and chromium. As above mentioned, MgF₂ is not particularly hydroscopic, but it has voids. A void allows water vapor to penetrate low-density films and gap between the columnar grains by capillary action. Additionally, if the MgF₂ film contains crystallites in its structure, their grain boundaries can cause an absorption tail for the refractive index below the material's band gap. This absorption tail is known as the Urbach edge, and it obeys the relationship

$$\kappa = \kappa_0 \exp[-\sigma(E_g - E)] \tag{34}$$

where $E_g=12.3$ eV, E is the photon energy, κ_0 is a determined value for the absorption coefficient, and σ is determined through fitting the designed data[12-14]. This relationship states that the absorption coefficient rises exponentially as you approach the band gap. If the substrate is heated the Urbach edge of the MgF₂ film becomes steeper, making the material transparent. At energy of 10 eV, this would increase the transparency by about a factor of 10 more than an unheated material. In dielectric multilayer high reflecting mirrors, a substance with low refractive index (*i.e* MgF₂, $n_L \approx 1.35$)is used together with second material with

high index (TiO₂, $n_H \approx 2.3$). If thickness of quarter wavelength($\lambda/4$) for each layer is deposited, when the high refractive index substance thin film be the last(outer) layer, then reflection power of this multilayer coating is given by:

$$R = \left| \frac{1 - \left(\frac{n_H}{n_L}\right)^{2p} \left(\frac{n^2_H}{n^2_L}\right)}{1 + \left(\frac{n_H}{n_L}\right)^{2p} \left(\frac{n^2_H}{n^2_L}\right)} \right|^2 \quad (35)$$

where number of $\lambda/4$ thin films be $2p+1$ and if R is investigated in vacuum or air. It is obvious from Eq.(35) that if number of layer (p) of both materials increases, then reflection power(R) also rises as well. Often maximum amount of R is obtained when odd number of layer is coated and if the last $\lambda/4$ layer being the substance with high refractive index (n_H) and if $\left(\frac{n_H}{n_L}\right)^{2p} \left(\frac{n^2_H}{n^2_L}\right) > 1$ then

$$R \cong 1 - 4\left(\frac{n_H}{n_L}\right)^{2p} \left(\frac{n_g}{n^2_H}\right) \quad (36)$$

and

$$T \equiv 1 - R \cong 4\left(\frac{n_H}{n_L}\right)^{2p} \left(\frac{n_g}{n^2_H}\right) \quad (37)$$

where n_g is refractive index of substrate used ($n_g=1.52$ for BK-7 glass) and T is transmission power of multilayer set, supposing absorption(A) is either zero or has a very small amount. Wavelength range over which reflection power is high usually is determined by ratio of refractive index of materials used in multilayer system, $\left(\frac{n_H}{n_L}\right)$,

$$\delta g = \frac{2}{\pi} \sin^{-1} \left[\frac{n_H - n_L}{n_H + n_L} \right] \quad (38)$$

where δg is half of band width of spectral range and it is clear that, the higher is ratio of n_H/n_L , the wider is band width. For narrow and wide band optical filters usually this ratio play an important role. Increasing or reduction number of coating thin films with $\lambda/4$ thickness has very little effect on this parameter(half band width).

Experimental details

There are a couple of ways to fabricate thin film structures. Two of the most common methods used are evaporation and sputtering. The objective of these deposition processes is to controllably transfer atoms from a source to a substrate where film growth occurs atomistically[9]. Here, we describe the method of evaporation. We used the method of evaporation. In this method, the material which is desired to be used for film formation is thermally heated by conduction. This material is therefore allowed to reach high enough temperatures, causing atoms to leave the source deposit themselves onto a substrate held above the evaporating material. By evaporating material onto a substrate, the layer thickness can be monitored and controlled to the scale of angstroms. Therefore, the key variable influencing evaporation rates is the temperature.

To deposit and produce a multilayer dielectric coating for high reflecting mirror, a vacuum system capable of reaching pressures with of 10^{-7} mbarn(Balzers), was employed. An optical thickness monitor(GSM-420 Balzers), as well as vibrating quartz crystal thickness monitor were mounted in this vacuum system to be able to check the obtained results, also enable to measure optical thickness and geometrical thickness of thin films as well. Even to measure and investigate the reflection and transmission of produced film not only the GSM-420 optical system was used but also a double beam spectrophotometer (Shimadzu) with wavelength range of 200-300 nm was employed as well for double checking the results in various conditions. Light sources of GSM-420 optical monitor were halogen and deuterium lamp, a diffraction grating with 600 lines per mm and also suitable spectral filters were used

in its monochrometer(350-950 nm), for light detection(transmitted or reflected) a silicon photo diode was used and results of this system was calibrated and with spectrophotometer, good agreement was obtained. Pure research grade MgF₂ and TiO₂ was used and both evaporated therefrom tantalum crucible separately, at pressures about $2-3 \times 10^{-5}$ mbar. MgF₂ was coated with 1 nm/s deposition rate and for deposition of TiO₂ partial pressure of 5×10^{-5} mbar was used and coating rate was 0.5 nm/s. Substrate was circular glass with 20mm diameter and 1 mm thickness(BK-7 glass). Substrates just before being used, were cleaned ultrasonically in heated Aceton and then by isopropal Alcohol.

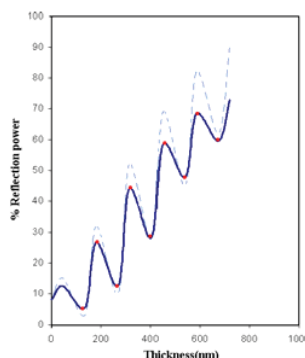


Fig. 4- Variation of reflection power of multilayer of TiO₂ and MgF₂ film with respect to thickness, while substrate temperature during deposition was at 280⁰C is shown. Dash line shows the theoretical results and experimental data are shown at extremum of solid curve.

Substrate temperature during and after deposition was at 280⁰C and this temperature were monitored by use of a thermocouple which was fixed on back of substrate holder. A shutter was used to cover the substrate and controlling the period of deposition exactly, which could be moved from outside of bell jar(work chamber). Substrate and other parts of work chamber could be cleaned(etched) by using argon glow discharge at 10⁻² mbar pressure, to make sure degassing, dose not cause problem for thin films, because out gassing in temperatures of nearly 300⁰C can be noticeable.

After finding the optimum condition for deposition of MgF₂ and TiO₂, (substrate temperature, deposition rates, distance between evaporation source and glass substrate), to start with TiO₂ coating with thickness of quarter wave($\lambda=4$), reflection power increased, then after deposition of MgF₂ reduction of R occurred. For each layer R_{max}, R_{min} were investigated.

Table 1- Results for R_{min}(%), R_{max}(%) and R(%) corresponding to the deposition of different layers.

Number	Glass(G)/TiO ₂ (T)/MgF ₂ (M), Air(A)	R _{min} (%)	R _{max} (%)	R (%)
1	G/T/A	-	13.6	12.42
2	G/T/M/A	4.3	-	5.38
3	G/T/M/T/A	-	28	26.67
4	G/(T/m) ² /A	14	-	12.62
5	G/(T/m) ² /T/A	-	52	44.1
6	G/(T/m) ³ /A	32	-	28.32
7	G/(T/m) ³ /T/A	-	65	58.54
8	G/(T/m) ⁴ /A	48	-	45.74
9	G/(T/m) ⁴ /T/A	-	78	68.44
10	G/(T/m) ⁵ /A	65	-	59.68
11	G/(T/m) ⁵ /T/A	-	86	73.86

Fig.(4) shows how this variation of R with respect to film thickness happens. Maximums points are when TiO₂ is the last layer and minimums points happen when MgF₂ is the last(outer) coating. As it can be seen there is a little difference between two curves (calculated and experimental), which is due to differences of refractive index which is calculated with what exactly is deposited, experimentally.

After eleven layers of $\lambda=4$ thin films of TiO₂ and MgF₂ (G/(TiO₂/MgF₂)₅/TiO₂/Air),

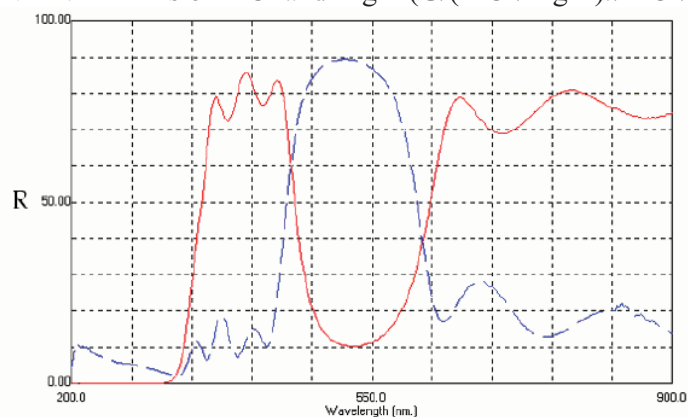


Fig. 5- Reflection and transmission power against wavelength for multilayer sample produced. Dash line shows the reflection power and the curve shows the transmission power of multilayer.

reflection power was investigated against wavelength (200-900 nm), both by GSM-420 optical monitor and as well as by spectrophotometer.

Fig.(5) give the variation of R vs, maximum amount of R was nearly 90%, transmission of this multilayer mirror was also investigated for the same range of wavelength. Results of Table(I) show that, Results for $R_{min}(\%)$, $R_{max}(\%)$ and $R(\%)$ corresponding to the deposition of different layers, see Fig.(4).

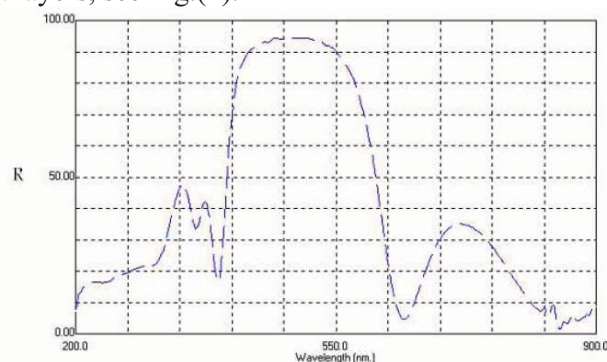


Fig. 6- Against wavelength (200-900 nm) after annealing the produced mirror at 300⁰C for 60 min.

The variation of reflection power for the produced sample after annealing at 300⁰C for 60 min is given in fig. (6). The reflection power has increased 5% after annealing for $400 < \lambda < 600$ nm, and about 30% for λ outside of this range.

Conclusion

After observing the obtained results, we get conclusion as follow:

i) Substrate temperature play an important role on refraction index and packing density of thin films has affect on R value. In high substrate temperature (T, 2800C)there is no porosity or very little.

ii) Deposition rate specially for TiO₂ layers has marked effect on refractive index of thinfilms and therefore has effect on R amount as well.

iii) Partial pressure of oxygen during coating TiO₂ is required and its amount also is important.

vi) Annealing at suitable temperature (T'3200C)and period of time, also can vary the obtained results and caused a more reproducible results. After annealing although reflection power increased by 5%, but spectral width which over that R was high became wider about 20nm as well.

vii)Comparison of the results before and after annealing shows, increasing of R for $\lambda = 500nm$, by 5% but at other wavelength $\lambda < 400nm$ and $\lambda > 600nm$ (outside the spectral band width), also reflectivity rises 20 to 30 percent as well. Adding amount of r and T for spectral wavelength range used(200-900 nm), nearly absorption is zero and can be ignored.

Acknowledgments

This work carried out with the support and instruments of optical industry of Isfahan(thin film group) and University of Arak. The authors are grateful for these help and support.

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