Accuracy design of ultra-low residual reflection coatings for laser optics*

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Refractive index inhomogeneity is one of the important characteristics of optical coating material, which is one of the key factors to produce loss to the ultra-low residual reflection coatings except using the refractive index inhomogeneity to obtain gradient-index coating. In the normal structure of antireflection coatings for center wavelength at 532 nm, the physical thicknesses of layer H and layer L are 22.18 nm and 118.86 nm, respectively. The residual reflectance caused by refractive index inhomogeneity (the degree of inhomogeneous is between −0.2 and 0.2) is about 200 ppm, and the minimum reflectivity wavelength is between 528.2 nm and 535.2 nm. A new numerical method adding the refractive index inhomogeneity to the spectra calculation was proposed to design the laser antireflection coatings, which can achieve the design of antireflection coatings with ppm residual reflection by adjusting physical thickness of the couple layers. When the degree of refractive index inhomogeneity of the layer H and layer L is −0.08 and 0.05 respectively, the residual reflectance increase from zero to 0.0769% at 532 nm. According to the above accuracy numerical method, if layer H physical thickness increases by 1.30 nm and layer L decrease by 4.50 nm, residual reflectance of thin film will achieve to 2.06 ppm. When the degree of refractive index inhomogeneity of the layer H and layer L is 0.08 and −0.05 respectively, the residual reflectance increase from zero to 0.0784% at 532 nm. The residual reflectance of designed thin film can be reduced to 0.8 ppm by decreasing the layer H of 1.55 nm while increasing the layer L of 4.94 nm.

Keywords: ultra-low residual reflectance, antireflection coatings for laser optics, refractive index inhomogeneity, accuracy design

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1. Introduction

Antireflection film is one of the most important optical coatings for laser optics. As the development of the optical coatings technology, it is not difficult to obtain the coatings with the residual reflectance below 0.5%.[1] In recent years, with the development of the technology in many applications, such as high-precision ring laser gyroscope (RLG), high power laser, and extreme ultraviolet lithography (EUVL) laser, the transmittance or reflectance of laser optical coatings reaching 99.999% or higher was required.[2–5] Therefore, the study on ultra-low loss laser optical coatings is still one of important frontier topics in optical coatings.

In the research field of low loss laser, the definite factors affecting the loss of laser optical coatings are scattering[6] (volume scattering and surface scattering) and absorption[7,8] (bulk absorption and surface absorption). As the development of the substrate surface processing technology and ion beam sputtering deposition technology, great progress in control of scattering and absorption had been made. For ultra-low loss antireflective coatings, the residual reflectance is one of the important loss sources except the scattering and absorption. Currently, the control of residual reflectance of optical coatings could be realized by accurately matching the coating design and preparation technology. Therefore, any characteristic deviation from the design and fabrication will lead to the increase of the residual reflection loss of the coatings. The homogeneity of refractive index, extinction coefficient, and physical thickness are the basic premises of the laser antireflective coatings design. However, the growth of thin film, which undergoes non-equilibrium physical process in reality, always results in the inhomogeneity in refractive index. The refractive index is not only a function of the wavelength, but also a function of the film layer thickness.[9] Therefore, refractive index inhomogeneity is a new kind of loss mechanism to the ultra-low residual reflection coatings for laser optics.

The studies of refractive index inhomogeneity are mainly focused on two aspects. One aspect is the effect of refractive index inhomogeneity on its optical properties. Jacobsson and Tikhonravov discussed the effect of coatings inhomogeneity on the spectral properties.[10,11] The other aspect is to achieve

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the optical modulation of the coatings by controlling the refractive index inhomogeneity. The primary research included the theoretical design of the coatings, the preparation principle, the fabrication, the performance measurement, and so on, which has formed an important research direction and become a significant branch in optical coatings.[12–15] In this paper, the refractive index inhomogeneity of ultra-low residual reflection coatings was studied. Moreover, the ideal design of the coatings was revised by numerical design method, of which research results will have a great significance for the loss control of ultra-low residual reflection laser coatings.

2. Fundamental design

The typical design of antireflection coatings for laser optics is usually V-type coating stack, which only uses two kinds of materials with different refractive index. To control the residual reflectance, we can adjust its physical thickness to obtain the minimum reflectance at single wavelength point. Figure 1 present the physical model of the V-type coating stack of antireflective coatings for laser optics. In the case of normal incidence, without considering the extinction coefficient of the layer material, in order to decrease the residual reflectance of laser wavelength λ_0 to zero, the optical thicknesses of two layers must satisfy:[1]

$$\tan^2\delta_{\mathrm{L}} = \frac{(n_{\mathrm{s}}-n_0)\left(n_{\mathrm{H}}^2-n_0n_{\mathrm{s}}\right)n_{\mathrm{L}}^2}{\left(n_{\mathrm{L}}^2n_{\mathrm{s}}-n_0n_{\mathrm{H}}^2\right)\left(n_0n_{\mathrm{s}}-n_{\mathrm{L}}^2\right)}, \tag{1}$$

$$\tan^2\delta_{\mathrm{H}} = \frac{(n_{\mathrm{s}}-n_0)\left(n_0n_{\mathrm{s}}-n_{\mathrm{L}}^2\right)n_{\mathrm{H}}^2}{\left(n_{\mathrm{L}}^2n_{\mathrm{s}}-n_0n_{\mathrm{H}}^2\right)\left(n_{\mathrm{H}}^2-n_0n_{\mathrm{s}}\right)}, \tag{2}$$

where $\delta_{\mathrm{H}} = 2\pi n_{\mathrm{H}}d_{\mathrm{H}}/\lambda_0$ and $\delta_{\mathrm{L}} = 2\pi n_{\mathrm{L}}d_{\mathrm{L}}/\lambda_0$ are the phase thicknesses of high refractive index layer and low refractive index layer, respectively. n_0, n_{s}, n_{H}, and n_{L} are the refractive indices of the incident medium, substrate, high refractive material, and low refractive material, respectively. d_{H} and d_{L} are the physical thicknesses of high refractive index layer and low refractive index layer, respectively.

In Eqs. (1) and (2), both $\tan\delta_{\mathrm{H}}$ and $\tan\delta_{\mathrm{L}}$ has two values. According the matching condition, $\tan\delta_{\mathrm{H}}$ and $\tan\delta_{\mathrm{L}}$ must be the values that have opposite algebraic symbol. So there are two conditions as follows:

i) When $\tan\delta_{\mathrm{L}} > 0$, $\tan\delta_{\mathrm{H}} < 0$, $90^\circ > \delta_{\mathrm{L}} > 0^\circ$, $n_{\mathrm{L}}d_{\mathrm{L}} < \lambda_0/4$, $180^\circ > \delta_{\mathrm{H}} > 90^\circ$, $n_{\mathrm{H}}d_{\mathrm{H}} > \lambda_0/4$ (the high refractive index layer is thicker than the low refractive index layer), the solution is called thick solution.

ii) When $\tan\delta_{\mathrm{H}} > 0$, $\tan\delta_{\mathrm{L}} < 0$, $90^\circ > \delta_{\mathrm{H}} > 0^\circ$, $n_{\mathrm{H}}d_{\mathrm{H}} < \lambda_0/4$, $180^\circ > \delta_{\mathrm{L}} > 90^\circ$, $n_{\mathrm{L}}d_{\mathrm{L}} > \lambda_0/4$ (the low refractive index layer is thicker than the high refractive index layer), the solution is called thin solution.

In the first solution, the high refractive index layer is thicker than the low refractive index layer. The second solution is just contrary. Therefore, if the value of n_{s}, n_{H}, and n_{L} at the design wavelength are given, the coating structure can be gotten according the above design. Taking the 532 nm antireflective coatings for example, the second solution is chosen as

$$\mathrm{Sub}/\alpha\mathrm{H}\beta\mathrm{L}/\mathrm{Air}\ \ (\lambda = 532\ \mathrm{nm},\ \ \mathrm{AOI} = 0^\circ), \tag{3}$$

where H and L indicate the $\lambda_0/4$ optical thickness of high and low refractive index respectively, and Sub indicates the substrate. The refractive index of the substrate is 1.461. The high refractive index layer and low refractive index layer are 2.12 and 1.48, respectively. Thus the structure of the designed coatings is

$$\mathrm{Sub}/0.3535\mathrm{H}1.3226\mathrm{L}/\mathrm{Air}\ \ (\lambda = 532\ \mathrm{nm},\ \ \mathrm{AOI} = 0^\circ). \tag{4}$$

The physical thicknesses of the layer H and layer L are 22.18 nm and 118.86 nm, respectively.

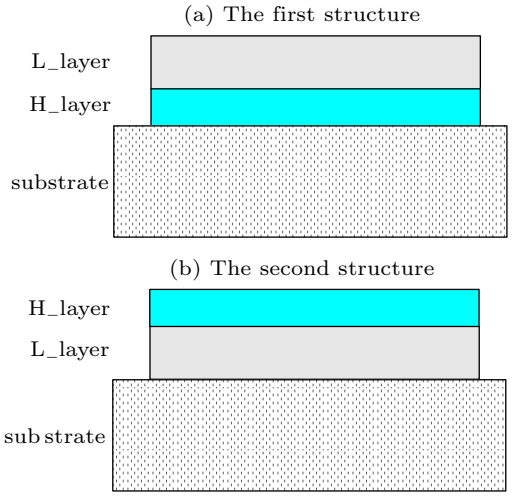


Fig. 1. (color online) The physical models of the V-type coating stack of the antireflective film. (a) The first structure and (b) the second structure.

3. The effect of the refractive index inhomogeneity on antireflective coatings

3.1. The physical model of antireflection coatings

Figures 2 and 3 show the physical models of refractive index inhomogeneity of single layer and antireflection coatings for laser optics, respectively.

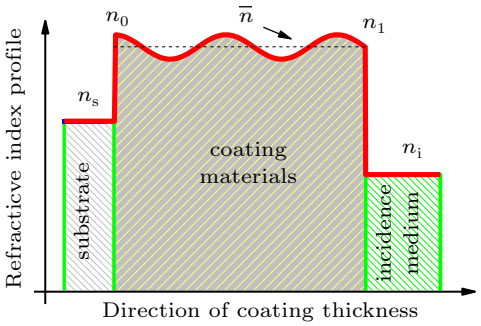


Fig. 2. (color online) The refractive index inhomogeneity model of single layer.

In Fig. 2, the refractive index of the substrate is n_s. The refractive indices of the substrate and near incident medium are n_0 and n_1, respectively. The physical thickness of the film is d_f. The coatings average refractive index is $\bar{n}_f = (n_1 + n_0)/2$. So, the refractive index inhomogeneous degree (Δn) is

$$\Delta n = (n_1 - n_0)/\bar{n}_f. \tag{5}$$

The distribution of refractive index can be described by mathematical function in the layers. X direction is defined as the direction from coatings to the incident medium. The refractive index distribution is described as

$$n_f(x) = ax^T + b. \tag{6}$$

Equation (6) is substituted into Eq. (5), and

$$n_1 = n_f(d_f) = ad_f^T + b, \tag{7}$$

$$n_0 = n_f(0) = b. \tag{8}$$

If the coatings average refractive index $\bar{n}_f$ and inhomogeneity δ were known, the coefficient can be gotten as

$$a = \bar{n}_f\delta/d_f^T, \quad b = (2-\delta)\bar{n}_f/2. \tag{9}$$

At last, the refractive index inhomogeneous distribution can be deduced as

$$n_f(x) = \bar{n}_f\left[\left(\frac{x}{d_f}\right)^T + \frac{(2-\delta)}{2}\right]. \tag{10}$$

Therefore, only the average refractive index and inhomogeneity degree need to be discussed in the following research of refractive index inhomogeneity.

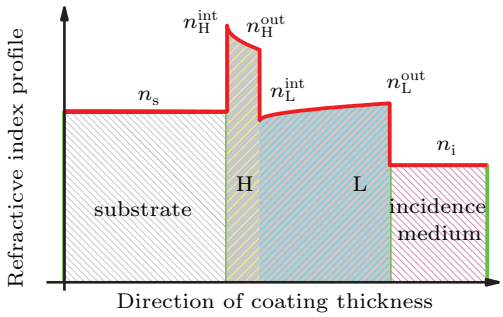


Fig. 3. (color online) The refractive index inhomogeneity model of double layers.

3.2. The effects of refractive index inhomogeneity on antireflection coatings

The normal design of antireflection coatings for laser optics is given in Section 2 without considering the inhomogeneity of refractive index. If the ranges of refractive index inhomogeneity degree of layer H and layer L vary from −0.02 to 0.02, the residual reflection is shown in Fig. 4. The relationship between residual reflectance at 532 nm and refractive index inhomogeneity was obtained. As shown in Fig. 4, the residual reflectance increased from 0 to 200 ppm, which is due to optical thickness deviation caused by the refractive index inhomogeneity. The result of numerical calculation shows that the residual reflectance could reduce to the lowest when the refractive index inhomogeneity of the two layers could match well in a certain range. For example, if the refractive index inhomogeneity degree of layer H ranges from −0.015 to 0.015 and the refractive index inhomogeneity degree of layer L ranges from −0.005 to 0.005, the residual reflectance could be controlled below 10 ppm.

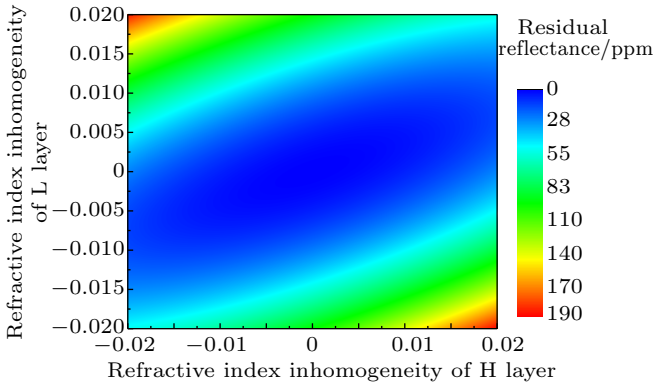


Fig. 4. (color online) The effect of refractive index inhomogeneity on residual reflectance.

Actually, the effect of refractive index inhomogeneity on residual reflectance is caused by the variation of optical thickness which results in the shift of the wavelength with zero-reflection. If the physical thickness of the double layers is decided by normal design and the refractive index inhomogeneity degree of the two layers ranges from −0.02 to 0.02, the effect of inhomogeneity on the residual reflectance could be obtained as shown in Fig. 5. The residual reflectance at central wavelength could reduce to the lowest by matching the refractive index inhomogeneity of the two layers. However, the lowest residual reflectance might not reach to zero as normal design.

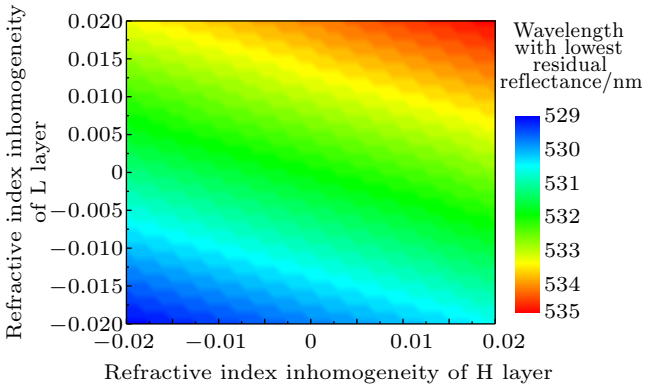


Fig. 5. (color online) The effect of refractive index inhomogeneity on wavelength with lowest residual reflectance.

4. Revised design

4.1. Method

The design method of antireflection coatings suggested by Jacobsson only can be applied if the refractive index inhomogeneity of coatings is not obvious. However, it can be solved by numerical calculation when the refractive index inhomogeneity is rather large. In this paper, a calculation process to design antireflection coatings with rather large refractive inhomogeneity is presented. The calculation flowchart is shown in Fig. 6.

The calculation procedure is as follows. (i) The average refractive indices of high and low refractive index layer are given firstly, and the physical thicknesses $d_{\rm H}$ and $d_{\rm L}$ are obtained under the assumption that the refractive indices are homogeneous. (ii) The refractive index inhomogeneity degrees of two kinds films are denoted by $\Delta n_{\rm H}$ and $\Delta n_{\rm L}$. The physics thicknesses of high and low refractive index layers are in the range $[(1-\delta_{\rm H})\sim(1+\delta_{\rm H})]d_{\rm H}$ and $[(1-\delta_{\rm L})\sim(1+\delta_{\rm L})]d_{\rm L}$, respectively. The range of the variable thickness is as the same order of magnitude as the refractive index inhomogeneity of films. (iii) Each layer is equally divided into several sub-layers, and then the refractive indices for each sub-layer are discretization. (iv) Calculate the residual reflectance with any thickness combination of high and low refractive index layers, with the step length of thickness variation 0.05 nm, which is related to the accuracy of thickness control during deposition process. (v) Find out the lowest residual reflection and its corresponding combination of physics thickness. The design precision of this method depends on the accuracy of selected step length.

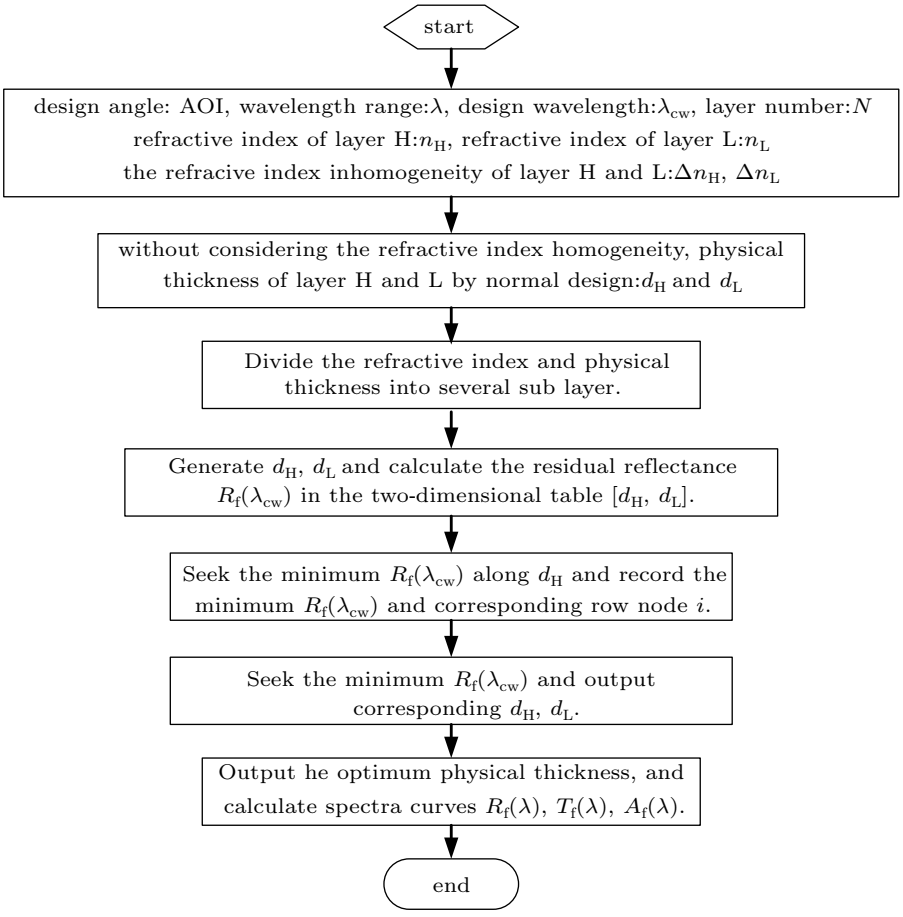


Fig. 6. The accuracy design flowchart for antireflection coatings.

4.2. Results and discussion

Taking a normal design for example, the parameters were set as follows: the refractive index inhomogeneity degree of layer H:$\Delta n_{\rm H}=-0.08$, the refractive index inhomogeneity degree of layer L $\Delta n_{\rm L}=0.05$, the design wavelength $\lambda_{\rm cw}=532$ nm, the physical thickness of high refractive index layer ranges from 19 nm to 26 nm, and the physical thickness of low refractive index layer ranges from 111 nm to 126 nm. The relationship between residual reflectance at 532 nm and physical thicknesses of layer H and layer L is presented in Fig. 7(a). The results show that the largest residual reflectance is 14955 ppm and the lowest residual reflectance is 2.06 ppm. The reflectance spectra are shown in Fig. 7(b). There are three spectra: the black curve is normal design without considering the inhomogeneity of refractive index, the red curve is cal-

culation result considering the effect of the inhomogeneity of refractive index, and the blue one is redesign result in the case of taking account into refractive index inhomogeneity. Compared with normal design, the central wavelength shifts toward longer wavelength and the residual reflectance at 532 nm increases from 0 to 0.0769%, which are caused by the refractive index inhomogeneity. The lowest residual reflectance can be achieved through seeking the physical thicknesses of layer H and layer L in the thickness range referred above. The step length is 0.05 nm which could affect the accuracy. The result is that the physical thicknesses of layer H and layer L are 23.48 nm and 114.36 nm, respectively. Compared with normal design, layer H increases by 1.30 nm and layer L reduces by 4.50 nm. The residual reflectance at 532 nm could reach 2.06 ppm by the revised design.

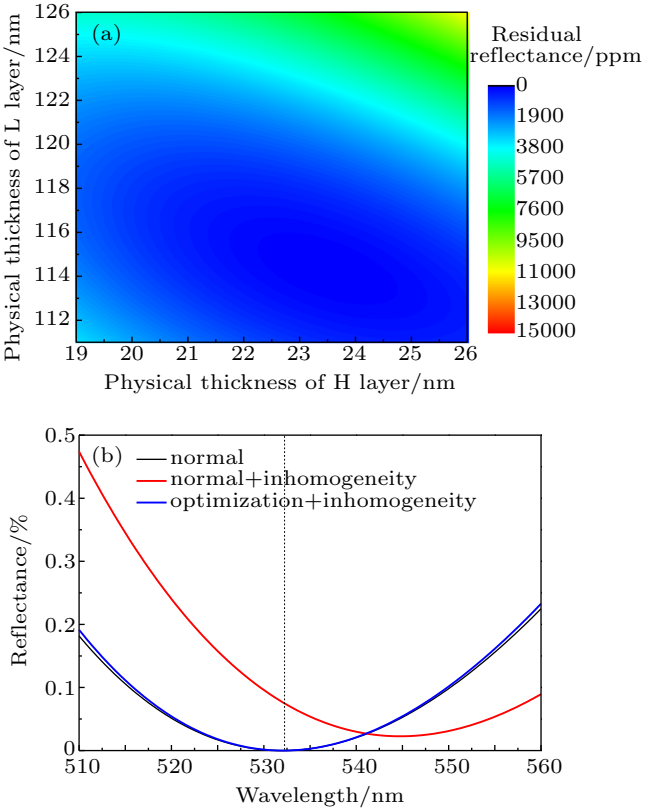


Fig. 7. (color online) Optimized design result with taking account into the refractive index inhomogeneity ($\delta_H = -0.08$, $\delta_L = 0.05$). (a) Relationship between residual reflectance and physical thicknesses of layer H and layer L. (b) Reflectance spectral curves with different design methods.

If the inhomogeneity of high refractive index layer was 0.08 ($\Delta n_H = 0.08$), the inhomogeneity of low refractive index layer was -0.05 ($\Delta n_L = -0.05$), and the design wavelength and the physical thicknesses of two materials were set as the same as above, the relationship between residual reflectance at 532 nm and physical thicknesses of layer H and layer L was given in Fig. 8(a). The largest residual reflectance is 9915 ppm, and the lowest residual reflectance is 0.8 ppm. The reflectance spectral curves are shown in Fig. 8(b). Compared with normal design, the central wavelength shifts toward short wavelength and the residual reflectance at 532 nm increases from 0 to 0.0784%, which are caused by the inhomogeneity of refractive index. The lowest residual reflectance can be achieved through seeking the physical thicknesses of layer H and layer L in the thickness range referred above. The step length is also 0.05 nm. The result is that the physical thicknesses of layer H and layer L are 20.63 nm and 123.81 nm, respectively. Compared with normal design, layer H reduces by 1.55 nm, and layer L increases by 4.95 nm. The residual reflectance at 532 nm could reach 0.8 ppm by the revised design.

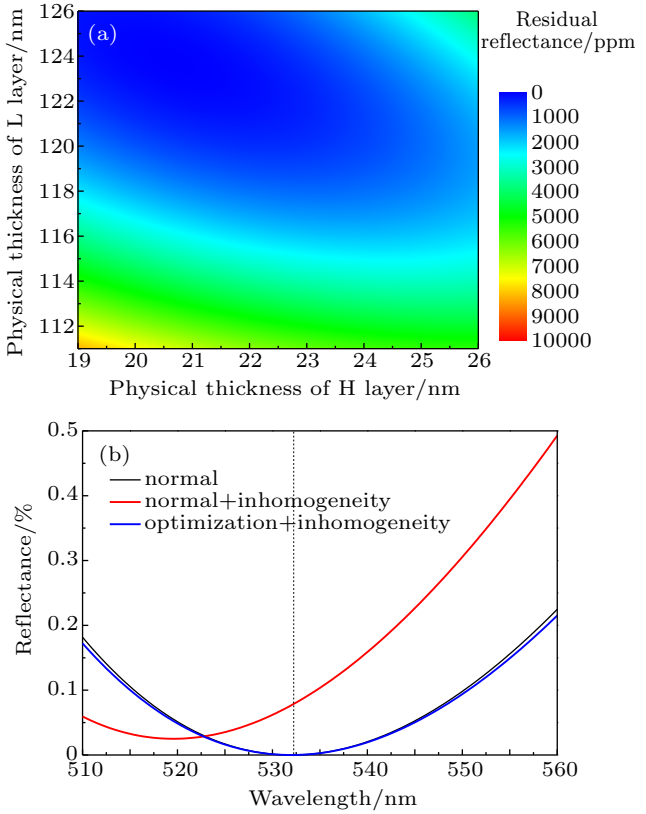


Fig. 8. (color online) Optimized design result with taking account into the refractive index inhomogeneity ($\delta_H = 0.08$, $\delta_L = -0.05$). (a) Relationship between residual reflectance and physical thicknesses of layer H and layer L. (b) Reflectance spectral curves with different design methods.

The reasons for the refractive index inhomogeneity of the film are inferred as following. (i) The physical thickness of the films varied from dozens of nanometers to several microns. Therefore, any fluctuation of technological parameters, such as substrate temperature, gas partial pressure, deposition rate, and so on, would cause the inhomogeneity. (ii) The substrate surface characteristics, such as subsurface damage and surface roughness, would transfer to films during the film growth, and then it may cause the inhomogeneity. (iii) The stress of the film was changing during the film growth, which would cause the microstructure difference as the increment of film thickness. Hence, the microstructure of film maybe one reason to induce the refractive index inhomogeneity. (iv) The mutual

infiltration existing between layer H and layer L during the deposition may cause the refractive index inhomogeneity.

From the above two design examples, it can be concluded that the general design method should be revised because of the inhomogeneity of refractive index. In order to obtain the ultra-low residual reflection coatings for laser optics, the accuracy of thickness control should be analyzed and be cited to the referred above optimized calculation design method.

5. Conclusion

The effect of the refractive index inhomogeneity on the ultra-low residual reflection coatings for laser optics was studied in this paper. For the ultra-low reflectance antireflection coatings working at 532 nm, if the film structure is obtained by normal design without considering the refractive index inhomogeneity, there must be an error in actual spectrum. Because the refractive index could not be a constant value along the film thickness increasing direction in the practical deposition process. When the refractive index inhomogeneity degrees of high and low refractive index layers range from -0.02 to 0.02, the residual reflectance at 532 nm is 200 ppm. In addition, the wavelength of lowest reflectance is between 528.2 nm and 535.2 nm. In this paper, the film structure is designed with taking the inhomogeneity of refractive index into account by numerical calculation. The result shows that the residual reflectance could reduce to ppm level by adjusting the physical thicknesses of the two layers. Since the refractive index inhomogeneity of thin film cannot be avoided during the coating deposition procedure, the revised design method has great influence on the fabrication of ultra-low reflectance antireflection coatings for laser optics.

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