Physical model of optical constants of SiO_2 thin films

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Abstract: SiO_2 thin films were deposited by ion beam sputtering (IBS) and electron beam evaporation (EB) technologies. The optical constants of SiO_2 thin films were fitted by nonlinear least square algorithm. 8 group experiments were designed based on L_8 (2^2) orthogonal array. The results show that intermix is the most important factor for the IBS SiO_2 thin film while Proe model for the EB SiO_2 thin film. The values of MSE evaluation function for IBS SiO_2 thin film and EB SiO_2 thin film decline 35% and 38% respectively, which shows that the physical models are reasonable and the physical meaning is clear. The method to estimate the effect of different factors was offered, which is meaningful for the analysis of optical constants of thin films.

Key words: SiO_2 thin films; optical constants; intermix; graded index; pore; overlayer

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SiO_2 薄膜光学常数物理模型

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摘 要： SiO_2 薄膜是光学薄膜领域内常用的重要低折射率材料之一。在文中研究中，通过测量薄膜的椭偏参数，使用非线性最小二乘法反演计算获得薄膜的光学常数。采用离子束溅射和电子束蒸发两种方法制备了 SiO_2 薄膜，在拟合过程中，基于 $L_8(2^2)$ 正交表设计了 8 组反演计算实验，以评价函数 MSE 为考核指标。实验结果表明：对 IBS SiO_2 薄膜拟合 MSE 函数影响最大的为界面层模型，对 EB SiO_2 薄膜拟合 MSE 函数影响最大的为 Pore 模型。同时确定了不同物理模型对拟合 MSE 函数的影响大小和反演计算过程模型选择的次序，按照确定的模型选择次序拟合，两种薄膜反演计算的 MSE 函数相对初始 MSE 可下降 35%和 38%，表明拟合过程模型选择合理物理意义明确。文中提供了一种判断薄膜物理模型中各因素对于薄膜光学常数分析作用大小的途径，对于薄膜光学常数分析具有指导意义。

关键词： SiO_2 薄膜； 光学常数； 界面层； 折射率梯度； 孔隙； 表面层

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0 Introduction

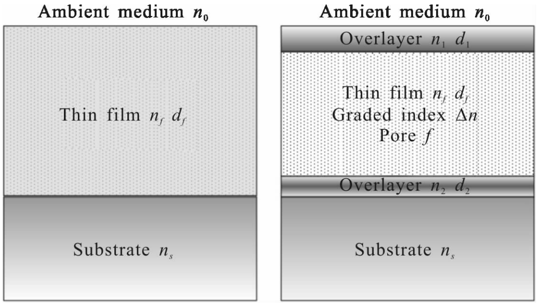
SiO_2 thin films is one of the most important low refractive index materials in the field of optical thin film, having low absorption coefficient, low scattering, high thermal stability and corrosion resistance, are widely used in various types of optical element of multilayer thin films, such as antireflective coatings, high-reflection coatings, beam splitters, optical filters, etc. The preparation of SiO_2 thin films are thermal evaporation, electron beam evaporation, ion assisted deposition, ion beam sputtering, magnetron sputtering, sol-gel, plasma enhanced chemical vapor deposition (PECVD), atomic layer deposition and thermal oxidation, etc[1-2].

The optical constants of SiO_2 films is the key basic data to the optical multilayer design, because optical constants of SiO_2 films is directly related to the preparation technology, so the precise acquisition optical constants is a priority in the multilayer applications. In recent years, the full spectrum inversion fitting calculation of optical constants of thin film has been widely used, a lot of literatures reported inversion fitting calculation method based on the reflectance/transmittance and the ellipsometric parameters in the wide optical spectrum from ultraviolet to infrared. In the process of the inversion calculation of optical constants, the physical model of film-substrate system directly determine the precision of inversion results[3-4].

In a lot of literature[5-12], it is generally considered that the physical model of film-substrate system are mainly interface layer (intermix), porosity (pore), refractive index gradient (graded index) and surface layer (overlayer), but the research on the rationality of physical model is less reported. In this paper, the SiO_2 films are prepared by the ion beam sputtering deposition (IBS) and electron beam evaporation deposition (EB) respectively. Ellipsometric parameters was used in the inversion calculation of the optical constants of SiO_2 films in visible range. Based on the orthogonal experiment design method, the physical model rationality of SiO_2 film-substrate system was studied, the results show that the method in the thin film optical constants of inversion on the computational physics model rationality evaluation has extensive application value. The result shows that this method in inversion of thin film optical constants for rationality evaluation of the physical model can be widely used.

1 Basic theory

According to the basic theory of optical thin film, ideal film-substrate system have two interfaces, the first interface is between air and film, the second interface is between film and substrate. With the improvement of the precision of measuring of optical thin film, the physical model of film-substrate system was modified. So the calculation accuracy of optical constants of thin films can be improved. The calculation results will have a physical meaning more clearly. Tikhonravov [6] gives theory and method of the modified physical model of ideal film-substrate system, as shown in Fig.1. In the modified model, graded index, overlayer, intermix and pore are all included, so the ideal film-substrate system is equivalent to a multilayer system.



(a) Ideal thin film-substrate system (b) Modified multilayer thin film-substrate system

Fig.1 Thin film-substrate system

Reflected ellipsometry measures the change in polarization state of light reflected from the surface of the thin film. The measured values are expressed as Ψ and Δ[13-14]. These values are related to the ratio of

Fresnel reflection coefficients r_p and r_s for $p-$ and $s-$polarized light, respectively.

$$\rho=\frac{r_p}{r_s}=\tan\psi\exp(i\Delta) \tag{1}$$

ψ and Δ are ellipsometric parameters, they are expressed as follows.

$$\psi=\mathrm{actan}\left(\frac{R_p}{R_s}\right) \tag{2}$$

$$\Delta=\delta_p-\delta_s \tag{3}$$

R_p and δ_p are reflectivity and reflected phase for $p-$polarized light, respectively. R_s and δ_s are reflectivity and reflected phase for $s-$polarized light, respectively.

The ellipsometric parameters ψ and Δ are functions of the angle of incidence θ, the film characteristics (thickness d_f, the refractive index $n(\lambda)$, the extinction coefficient $k(\lambda)$, the refractive index inhomogeneity Δn, porosity f), the surface layer (refractive index n_1, physical thickness d_1), the interface layer (refractive index n_2, physical thickness d_2). Therefore, with ψ and Δ values, using nonlinear constrained optimization algorithm, the optimal iterative solution of the above-mentioned variables are gradually achieved. In the iterative process, the evaluation function is defined as follows[15]:

$$MSE=\left\{\frac{1}{2N-M}\sum_{i=1}^{N}\left[\left(\frac{\Psi_i^{\mathrm{mod}}-\Psi_i^{\mathrm{exp}}}{\sigma_{\Psi,i}^{\mathrm{exp}}}\right)^2+\left(\frac{\Delta_i^{\mathrm{mod}}-\Delta_i^{\mathrm{exp}}}{\sigma_{\Delta,i}^{\mathrm{exp}}}\right)^2\right]\right\}^{\frac{1}{2}}=\left\{\frac{1}{2N-M}\chi^2\right\}^{\frac{1}{2}} \tag{4}$$

Where N is the number of (Ψ, Δ) pairs, M is the number of variable parameters in the model, and σ is the standard deviations of the experimental data points. The smaller MSE is the fit is better.

Since the variables are all functions of wavelengths, the dispersion model of the optical constants is needed. The dispersion model are fitted instead of the refractive index $n(\lambda)$ and the extinction coefficient $k(\lambda)$. In this article the extinction coefficient was set as zero (in the visible band of the order of 10^{-9}–10^{-6}), and the Cauchy model was used:

$$n(\lambda)=A+\frac{B}{\lambda^2}+\frac{C}{\lambda^4} \tag{5}$$

Where A, B, and C are constants, for extrapolating the index values estimated in the transparent region of the spectrum to weakly absorbing region.

2 Experiment and measurement

The first SiO_2 films were prepared using an ion-beam sputtering (IBS) apparatus. A 16 cm RF ion source was used to obtain the sputtering ion beam (Ar^+). The sputtering ion beam voltage and ion beam current were 1 250 eV and 600 mA. The target used was high purity UV fused silica disk (purity greater than 99.995%) of diameter 365 mm. O_2 gas was supplied to the target as atmospheric gas, the flow rate was 25 sccm. The background pressure during deposition was 1×10^{-4} torr, and the deposition time is 6 000 s. The second SiO_2 films were deposited using an electron beam evaporation (EB) of a high purity UV fused silica source (purity greater than 99.995%). The substrate temperature was 120℃. During deposition, the deposition rate was 0.3 nm/s, and the pressure in the chamber was 1×10^{-4} Pa. For the EB deposition of SiO_2 films with a thickness 1 500 nm, an IC5 quartz crystal monitor was utilized to measure film thickness. All SiO_2 films were deposited on silicon substrates, and the substrate are 40 mm in diameter, 0.32 mm in thickness and surface roughness greater than 0.3 nm. The visible ellipsometric parameters Ψ and Δ were measured at an angle of incidence of 55° and 65° on a J.A. Woollam ellipsometer between 400 and 900 nm. Figure 2 and Fig.3 show the spectra of the Ψ and Δ of IBS SiO_2 films and EB SiO_2 films.

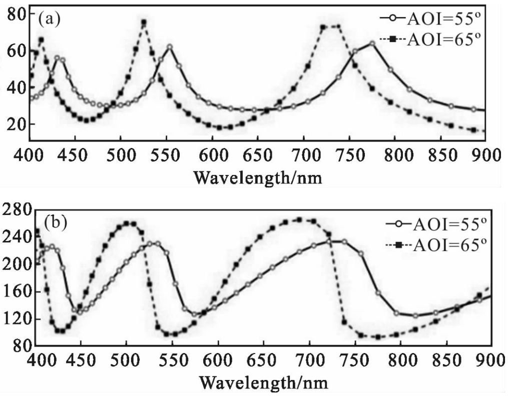


Fig.2 Ellipsometric parameters for IBS SiO_2 thin film on Si

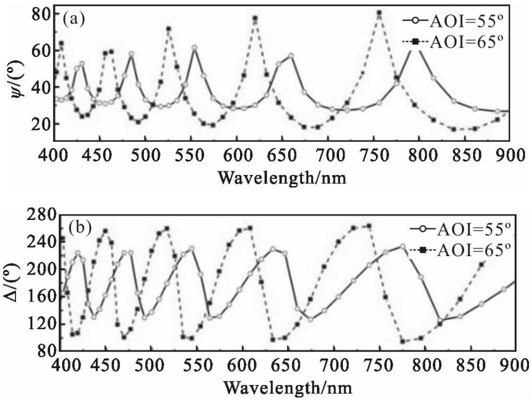


Fig.3 Ellipsometric parameters for EB SiO_2 thin film on Si

3 Results and discussion

In this paper, the modified physical model of SiO_2 films is selected as main calculation model. In this model, the film-substrate includes the interface layer between substrate and film, pore, graded index and overlayer. The Cauchy model was used as film refractive index dispersion equation. In the process of inversion calculation, the orthogonal experimental design method to determine the different model of MSE function contribution. The *A*, *B*, *C*, and *D* represent the intermix, pore, graded index and overlayer in the modified physical model (1 means using the model, 0 means it is not applicable model). Two factors and two levels were analyzed in the orthogonal experiment. The L_8 (2^2) orthogonal table is used for the design of the inversion calculation experiment of SiO_2 films optical constants, the design results are shown in Tab.1. According to the experiments sequence in Tab.1, the optical constants of SiO_2 films were calculated respectively. Table 1 presents the MSE values of each experiment.

The range analysis of the orthogonal experiment was carried out on the table[16]. The range through the use of poor can judge a model′s contribution to the MSE. The size of the range can illustrate the model′s contribution to the MSE. Figure 4 and Fig.5 show the evaluation function range of IBS SiO_2 films and EB SiO_2 films, respectively. For the IBS SiO_2 thin films, they are intermix, graded index, pore and overlayer by order of the contribution to the MSE. For the EB SiO_2 films, they are Pore, Intermix, Overlayer and Graded index. In order to make MSE decline orderly in the fitting process, the model should be added in according to the determined order.

Tab.1 Results of orthogonal experiments of the SiO_2 thin film optical constants

Number	Experi-ments	Inter-mix	Pore	Graded index	Over-layer	MSE (SiO_2 film)	
						IBS	EB
1	*ABCD*	1	1	1	1	5.79	7.759
2	*ABC*	1	1	1	0	5.786	7.534
3	*AD*	1	0	0	1	5.851	7.061
4	*A*	1	0	0	0	5.851	7.109
5	*BD*	0	1	0	1	7.289	7.065
6	*B*	0	1	0	0	8.232	7.807
7	*CD*	0	0	1	1	6.622	6.694
8	*C*	0	0	1	0	6.651	6.672

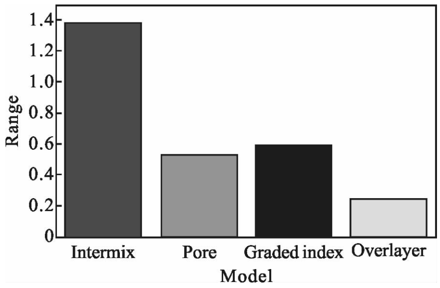


Fig.4 MSE range analysis of IBS SiO_2 thin film fitting

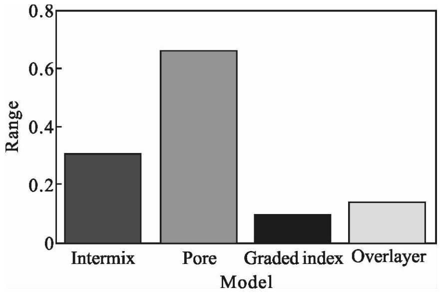


Fig.5 MSE range analysis of EB SiO_2 thin film fitting

The range analysis of MSE is to obtain the weight of different models contribution to the MSE. Through analysis of variance to Tab.1, confidence probability of the impact of different models on MSE can be obtained. The confidence probability analysis of the impact of different models on MSE is shown in Fig.6 and Fig.7. For IBS SiO_2 thin film, the film-substrate interface mixed layer (intermix) is the most important for MSE, the confidence probability is 99.03%, indicating that intermix is a key factor. For EB SiO_2 thin film, the pore model is the most important factor, whose confidence probability is 94.78% , indicating that electron beam evaporation film is a porous structure. These results have been consensus on properties of the SiO_2 films by the two deposition technologies.

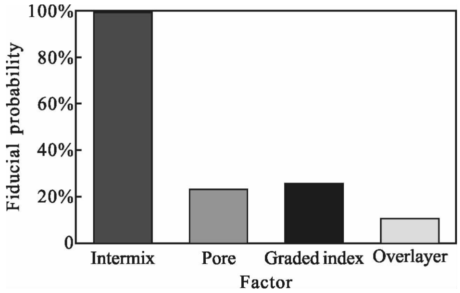


Fig.6 Confidence probability analysis of IBS SiO_2 thin film

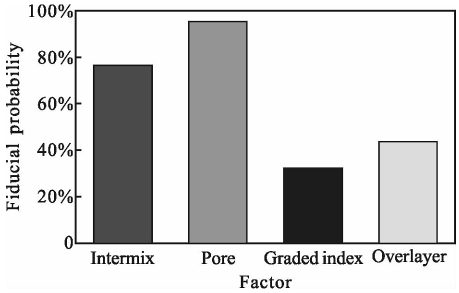


Fig.7 Confidence probability analysis of EB SiO_2 thin film

With the analysis results, for IBS SiO_2 thin film, after the fitting with the Cauchy model, the intermix, graded index, pore and overlayer were added orderly, fitting MSE result shown in Fig.8. MSE eventually fell 35% relative to the initial value. For EB SiO_2 thin film, with the model added by order of pore, intermix, overlayer and graded index, the final MSE value declined 38% shown in Fig.9. Table 2 shows the optical constants fitting results of the two SiO_2 films. Figure 10 shows the refractive index and refractive index inhomogeneity of the SiO_2 films.

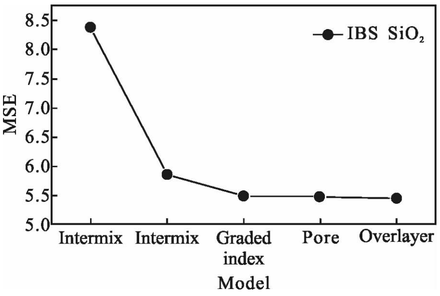


Fig.8 Fitting MSE variation of IBS SiO_2 thin film

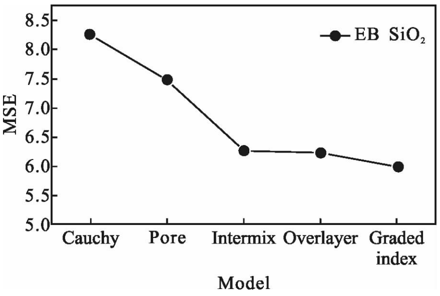
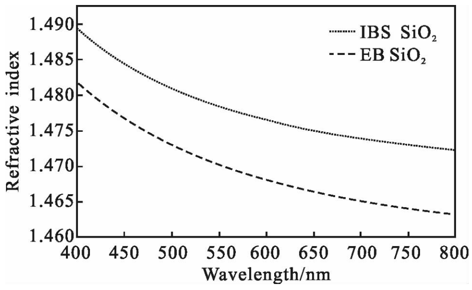


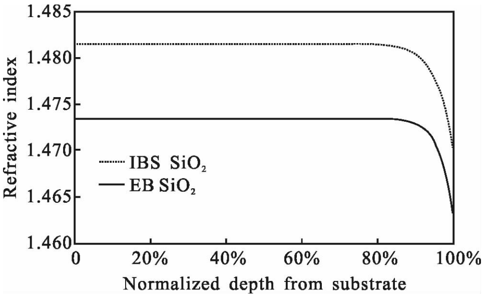
Fig.9 Fitting MSE variation of EB SiO_2 thin film

Tab.2 Calculated optical constants of the SiO_2 thin films

Properties		IBS SiO_2	EB SiO_2
Thickness	d_f/nm	789.8±0.8	1479.8±0.2
Intermix	d_2/nm	3.4±0.7	2.1±0.5
Overlayer	d_1/nm	0.42±0.03	1.297±0.78
Cauchy equation coefficient	A	1.466 9±0.001 0	1.459 6±0.000 9
	B	0.003 4±0.000 3	0.003 0±0.000 4
	C	5.1e−5±0.3e−5	1.2e−4±0.6e−5
Inhomogeneity of refractive index	Δn	−0.007±0.004	−0.024±0.008
Pore	Porosity/%	0.029±0.00	0.466±0.332
MSE		5.451	5.957



(a) Refractive index



(b) Inhomogeneity of refractive index

Fig.10 Refractive index and refractive index inhomogeneity of the SiO_2 films

4 Conclusion

In this work, the contribution to MSE function of the single thin film-substrate system model was evaluated by the orthogonal array experiments, this method could be applied to all types of thin films. After the contribution of the models was determined, the models were added orderly, the MSE declined 35% and 38% for IBS SiO_2 thin film and EB SiO_2 thin film. For both the films, MSE declined monotonically, indicating that the fitting process was reasonable. At the same time, the experimental results show that the interface effect between the IBS SiO_2 film and substrate is significant, while the EB SiO_2 film is porous structure. There are no unified models for film optical constants; the accurate optical constants could be obtained only with the specialties of deposition technologies and features of film structure synthetically considered.

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