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On-site determination of optical constants for thin films

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Abstract: The optical constants (refractive index and extinction coefficient) accuracy of thin films directly affects the properties of designed and fabricated optical devices. Most of the determination methods of optical constants are complex and cannot be applied during the film depositing process. In this paper, an optical constants determination method of thin films on-site is proposed. By monitoring the transmittance of depositing materials, this method can rapidly and accurately determine the optical constants on-site. For demonstration, the near-infrared optical constants of high-absorption material Si, low-absorption material Ta_2O_5 and ultra-low-absorption material SiO_2 are obtained as n=3. 22, k=4. 6×10^{-3}, n=2. 06, k=1. 3×10^{-3} and n= 1. 46, k=6. 6×10^{-5} respectively by this method. It reveals that this method is suitable for determining both strong and weak absorption materials' optical constants. It provides an effective way for precisely determining optical constants on-site, which is meaningful for the design and fabrication of high-quality optical devices.

Key words: optical constants, near-infrared, thin film, on-site, determination

薄膜光学常数的原位测定

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摘要：薄膜的光学常数（折射率和消光系数）精度直接影响设计和制造的光学器件的性能。大多数光学常数的测定方法较为复杂，不能直接应用在镀膜过程中。提出了一种薄膜光学常数原位实时测量的方法，通过监测沉积材料的透射率可以快速准确地测量光学常数。测量了高吸收材料Si、低吸收材料Ta_2O_5和超低吸收材

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料 SiO_2 的近红外光学常数,用这种方法测得光学常数分别为 n=3.22, k=4.6×10^{-3}, n=2.06, k=1.3×10^{-3} 和 n=1.46, k=6.6×10^{-5}。该方法适用于强吸收材料和弱吸收材料光学常数的测定,为在线精确测量薄膜的光学常数提供了一种有效的方法,对设计和制造高质量的光学器件具有重要意义。

关 键 词:光学常数;近红外;薄膜;原位;测定

Introduction

Optical thin films have broad applications in the fields of astronomy [1-3], energy-saving building [4-6], laser detection [7-9], optical communication systems [10], and sensors[11, 12], *etc*. The determination of optical constants is essential for the design and preparation of high-performance optical thin film devices, such as ultra-narrow bandpass filters, which heavily depends on the accuracy of optical constants. However, the optical constants of thin films are affected by many factors, including the deposition methods, deposition conditions and other parameters [13-15]. They might change after being exposed to air. To date, numerous approaches of optical constants measurements have been reported, such as Brewster angle method [16], prism-film coupling [17], ellipsometry [18], photometric method [19], envelope method [20], *etc*. Most of these methods require not only complex equipment but also work off-site. For example, although the ellipsometry method is highly sensitive to optical constants, it requires complicated instruments and data analysis [21]. The Prism-film coupling also needs complicated test equipment and requires enough thickness of samples which is difficult to be prepared [17]. For optical constants determination, it is critical to improve the measurement accuracy and reduce experimental error. However, the optical constants of deposited film always have some deviation between the optical monitor of deposition system and off-site measuring setup, due to their difference in principle, method and composition, *etc*. This will result in the difficulty of control precision and deviation of optical property from the designed ones. To avoid such a problem, it is ideal to determine the optical constants of films directly from the deposition system without using other measurement setups. Determining the optical constants of films on-site is of great significance for evaluating the quality of thin film devices and improving their yield.

In this paper, an on-site determination method for optical constants of thin films is proposed by employing the optical monitor of deposition system itself, which can eliminate the error between the deposition system and measurement system and get the result in real time. It is valid for both high-absorption and low-absorption materials with very high precision.

1 Experimental details

The Leybold ARES1110 high-vacuum evaporation coating device is selected for this experimental demonstration, which is equipped with a monochromatic optical monitoring system, is shown in Fig. 1. During the coating process, the material of evaporator is deposited onto the glass substrate under the action of electron beam heating, and plasma is introduced into the chamber by plasma source to improve the quality of the coating. Optical coating thickness measurement unit (a dual-beam photometer) measures the optical transmission of the coating through the centrally substrate mounted on holder. The physical thickness is obtained by the system's quartz crystal film thickness monitor. Sample signal is transmitted to the optical monitoring system through the optical fiber, and the monochromator controlled by the stepping motor is integrated into the standard device, allowing precise measuring of entire refractive or transmission spectra. This information is synchronized with the computer terminal, the optical information of the current coating can be obtained through nonlinear fitting of the monitoring curve. This method can be applied to coating systems equipped with optical film thickness detectors, and also applicable to broadband optical monitoring systems.

2 Results and discussion

2.1 Calculation of refractive index and extinction coefficient

Since the film transmittance mainly depends on the refractive index n and physical thickness d. The refractive index of a transparent dielectric film can be accurately determined in real time by the change trend of transmittance with thickness, which is obtained by the optical monitor of deposition system during depositing. And the extinction coefficient k can be accurately determined by the change of peak transmittance with depositing thickness simultaneously. Therefore, the optical constants of transparent dielectric films can be obtained by on-site fitting the spectral shape with nonlinear optimization methods.

For optical thin films deposited on a substrate, an ideal equivalent interface can replace the two interfaces of a monolayer film and all media are assumed to be homogeneous and nonmagnetic. Where the overall reflection and transmission are the superpositions of the multiple reflection and transmission at the two interfaces. Due to the thickness of substrate is much larger than the wavelength of light, the interference effect can be ignored. Moreover, the absorption at near-infrared wavelengths is negligible when glass is used as substrate. The transmission coefficient of the film and substrate is expressed as:

$$t_{total} = \frac{t_{12}t_{23}}{1 - r_1 r_2} \quad . \tag{1}$$

In Eq. (1), t_{12} and t_{23} are the transmission coefficient of thin film and substrate, respectively. r_1 is the reflection coefficient of interface between thin film and substrate, r_2 is the reflection coefficient of substrate. For simplicity, glass is assumed as substrate in the following description.

The feature matrix of thin film is described as:

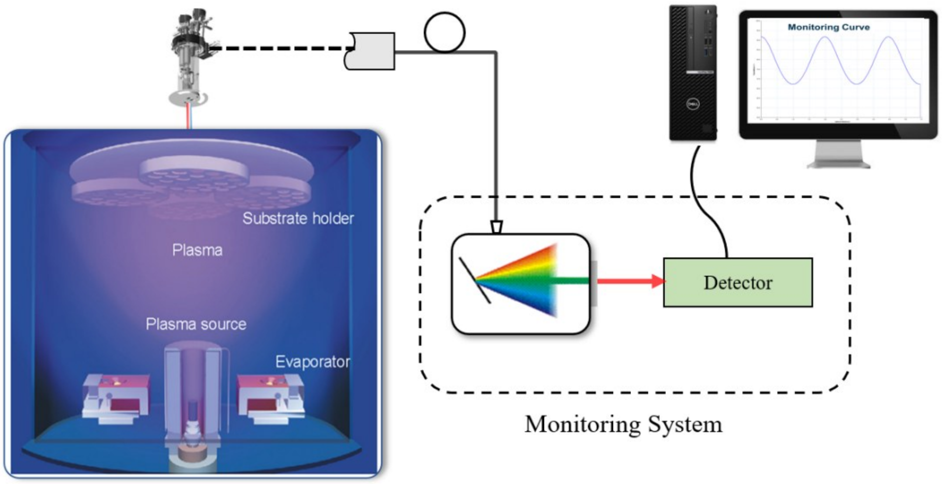

Fig. 1 Schematic diagram of on-site determination for the deposition system
图1 沉积系统原位测量的示意图

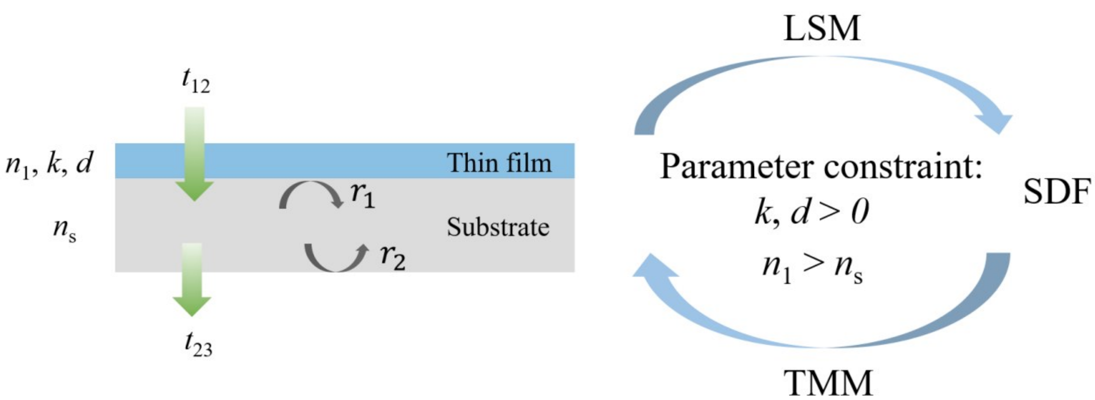

Fig. 2 Calculation flow of the dielectric thin film optical constants, which equivalent interface is composed of thin film material deposited on a transparent substrate, with the parameter constraints imposed by the absorption loss of film
图2 介质薄膜光学常数的计算流程,其等效界面由薄膜材料沉积在透明基板上构成,参数约束为薄膜的吸收损耗

$$\begin{bmatrix} B \\ C \end{bmatrix} = \begin{bmatrix} \cos\delta_1 & i\sin\delta_1/\eta_1 \\ i\eta_1\sin\delta_1 & \cos\delta_1 \end{bmatrix} \begin{bmatrix} 1 \\ \eta_2 \end{bmatrix} \quad . \quad (2)$$

Herein, the $\delta_1 = \frac{2\pi n_1 d}{\lambda} cos\theta$ is the phase thickness of thin film and n_1 represents the optical constants of thin film. d is the physical thickness of thin film, and the angle of incident light θ is fixed to 0. Here, $\eta_1=n_1$, $\eta_2=n_2$, η_1 and η_2 are the optical admittance of thin film and substrate, respectively. The optical admittance of the combination of thin film and substrate is $Y = \frac{C}{B}$.

According to Fresnel's formula and transmission matrix method (TMM), the reflection of a thin film can be defined as: (Here, η_0 is incident medium admittance)

$$R = \left(\frac{\eta_0 - Y}{\eta_0 + Y} \right)^2 \quad . \quad (3)$$

From Eq. (2), a series of extreme values appear at the reference wavelength when the effective optical thickness of the film is integral multiple. And the reflectance has a maximum value when the optical thickness of the film is odd multiple. In the spectral transmission curve of the sample, the extreme reflectance can be calculated in terms of the transmittance corresponding to the odd multiple optical thicknesses. The refractive index of the film n can be obtained by Eq. (3).

The influence of the absorption of thin film material on transmittance is further considered. The film's absorption will directly affect the transmittance of thin film, even if it is a slight difference. From Equation. (1), we can obtain the total transmittance of the film and substrate:

$$T_{total} = \frac{4\eta_0\eta_2 t_{23}^{\ 2}}{\left(\eta_0 B + C\right)\left(\eta_0 B + C\right)^* \left(1 - r_{21} r_{23}\right)^2} \quad . \quad (4)$$

The incident light is unpolarized and chosen as λ, where incident medium admittance $\eta_0=n_0=1$. The transmittance can be described as $T_{total} = f\left(n, k, d, n_s, n_0, \lambda\right)$. The relationship is established between the film transmittance and its optical constants n, k, d. Since the film transmittance T mainly depends on the refractive index n and physical thickness d, the refractive index n of the film under the corresponding process conditions can be accurately obtained through the relationship between the film transmittance T and physical thickness d. Due to the absorption of thin film materials, the transmission peaks in spectra are different. The extinction coefficient of thin film material can be obtained by fitting the transmission spectral curve of thin film.

Fig. 2 schematically shows the flow of the calculation process of optical constants. The fitting method we chose is the widely used least squares method (LSM), the goal of the optimization is to minimize the spectral discrepancy function (SDF) such that the thickness-dependent transmission of the dielectric thin film can approach the monitoring curve. We define the spectral discrepancy function of the optimization as [22]

$$SDF = \sqrt{\frac{1}{N}\sum_{n=1}^{N}\left\{T_{fit}\left[d,n\left(\lambda_n\right),k\left(\lambda_n\right)\right] - T_{exp}\left(\lambda_n\right)\right\}^2}, \quad (5)$$

where T_{fit} and T_{exp} represent the optical transmission of optimally fitting and monitoring, respectively. Here, absorption loss and monitoring sensitivity of the film is considered, the lower limit of film thickness and extinction coefficient is set to 0, and the substrate refractive index is less than the thin film refractive index. In addition, the error value of on-line detection method of optical constants of materials is 0.05%, which is mainly determined by the square root of measurement error of coating optical monitoring system provided by the manufacturer.

2.2 On-line detection of optical constants of high absorption materials

Taking absorptive material of Si as an example, it is deposited on glass with the thickness of 6H, where H is the abbreviation of 1/4 wavelength optical thickness of high refractive index material at monitor wavelength. The change of transmittance with Si depositing thickness at monitor wavelength of 1300 nm is shown in Fig. 3(a). The transmission curve can be fitted in real time to obtain the refractive index of Si as n =3.22. In addition, the extinction coefficient of thin films can be obtained by the reduction of transmission peaks as well. For high absorption materials, the extinction coefficient is easy to be accurately obtained, since the transmission decreases obviously with the increase of deposition thickness, as shown in Fig. 3(b). The extinction coefficient k of Si can also be accurately obtained as 4.6×10^{-3} by rapidly fitting the peaks of the transmission curve as shown in Fig. 3(b).

We demonstrate that the optical constants of samples can be obtained by nonlinear fitting of monitoring curves during direct deposition. In order to further evaluate the quality of this method, SDF can be used to describe. As discussed above, monitoring curve peaks are

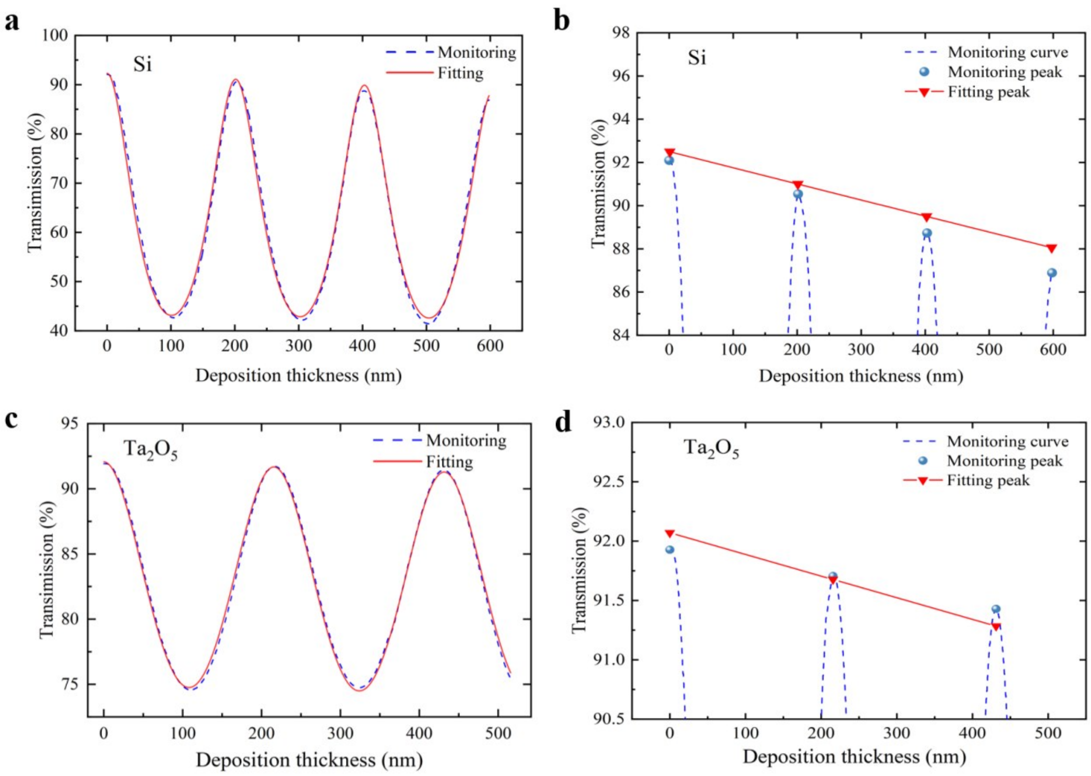

Fig. 3 (a) The transmission curve changed with the deposition thickness of Si film (monitor wavelength of 1300 nm) and (b) shows a zoom-in view of the Si transmission curve; (c) The transmission curve changed with the deposition thickness of Ta_2O_5 film (monitor wavelength of 890 nm) and (d) shows a zoom-in view of the Ta_2O_5 transmission curve; Blue dotted line, monitoring transmission curve; red solid line, fitting curve by LSM; cyan solid circles, monitoring transmission peaks; red inverted triangle, fitting curve peaks

图 3 (a)透射曲线随 Si 膜沉积厚度的变化(监控波长为 1300 nm)和(b)Si 的透射曲线放大视图;(c)透射曲线随 Ta_2O_5 薄膜的沉积厚度的变化(监控波长为 890 nm),(d)Ta_2O_5 透射曲线的放大图;蓝色虚线,监控的透射曲线;红色实线,LSM 的拟合曲线;青色实心圆,监控的透射曲线峰值;红色倒三角形,拟合曲线的峰值

closely related to the optical constants in the deposition process. The transmittance measurements and fitting curve peaks of Si film are verified in Fig. 3, and the spectral discrepancy function (SDF=0.78%) shows the reliability of our method.

2.3 On-line detection of optical constants of low absorption materials

This method is not only suitable for high absorption materials like Si, but also valid for low absorption film materials. The change of transmittance with the depositing Ta_2O_5 thickness at monitor wavelength of 890 nm is shown in Fig. 3(c). The transmission curve can be fitted in real time to obtain the refractive index of Ta_2O_5 as n = 2.06. Even though Ta_2O_5 has weak absorption, the extinction coefficient of thin films can also be obtained by fitting the reduction of transmission peaks as shown in Fig. 3(d). We can obtain the extinction coefficient of Ta_2O_5 k=1.3×10^{-3} (SDF=0.12%) at the film thickness d=500.1nm.

2.4 On-line detection of optical constants of materials with refractive index close to monitor substrate

Although this method is valid to determine the optical constants on-site for both low and high absorption materials, it is invalid when there is a small refractive index difference between deposited material and substrate lead to the weak signal changes. In order to determine the optical constants of SiO_2 with glass substrate on-site, the substrate can be coated with high refractive index material or film stack to solve this problem. A film stack of (HL)^4 (H: an optical thickness of Ta_2O_5 layer, L: an optical thickness of SiO_2 layer) has been coated on the glass substrate to act as an equivalent substrate, as shown in the inset of Fig. 4. Then SiO_2 film can be deposited onto this corresponding substrate to determine its refractive index and extinction coefficient. Benefits from the coated substrate, the transmittance change becomes much more apparent than the bare glass substrate. The change of transmittance with the depositing SiO_2 thickness at monitor wavelength of 1064 nm is shown in Fig. 4. The transmission curve can be fitted in real time to obtain the refractive index of SiO_2 as n =1.46.

For absorption material coated in an equivalent substrate, the refractive index and extinction coefficient of thin films can be obtained by monitoring the intensity of light and reduction of transmission peaks, respectively. The extinction coefficient k of SiO_2 films can also be obtained as 6.6×10^{-5} by simply fitting the curve of the transmittance T with the extreme point, as shown in Fig. 4. Such a small extinction coefficient (SDF=0.02%) extracted by our method reveals that the high accuracy of this method.

As discussed above, this method is not only suitable for high absorption materials, but also suitable for low absorption materials. What is more, it is also applicable to the film material with refractive index close to the monitoring substrate by depositing a high refractive index difference film or film stack. The extracted optical constants of the above three kinds materials are listed below in Table 1.

Table 1 The optical constants of film materials at monitoring wavelengths
表1 监控波长下的薄膜材料光学常数

Sample	Monitoring wavelength (nm)	d (nm)	n	k	SDF
Si	1300	598.1	3.22	4.6×10^{-3}	0.78%
Ta_2O_5	890	500.1	2.06	1.3×10^{-3}	0.12%
SiO_2	1064	1404	1.46	6.6×10^{-5}	0.02%

3 Conclusion

In this paper, we have proposed an on-site method to accurately determine the optical constants of thin films and eliminate the systematic error, by employing the optical monitor of the coating system itself. The relationship between the transmission and depositing thickness of thin films is employed to determine the optical constants on-

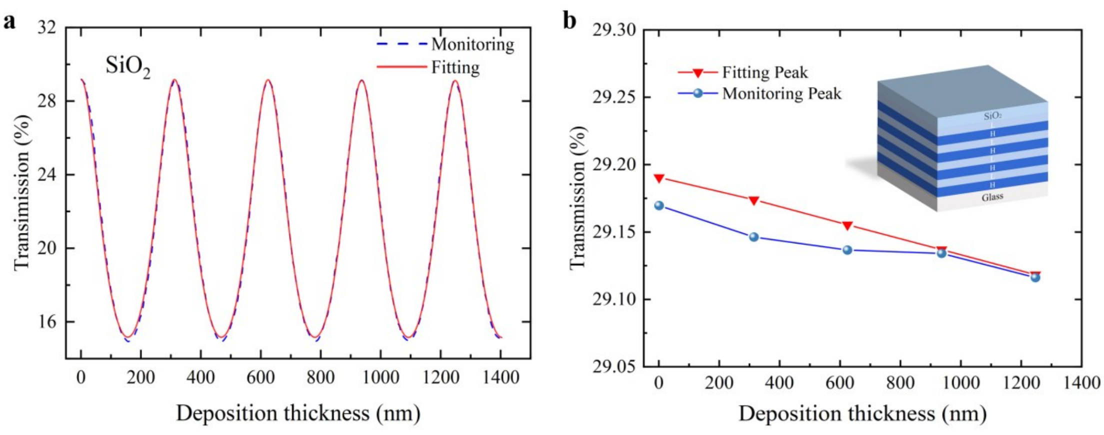

Fig. 4 (a) transmission curve changed with the deposition thickness of SiO_2 film (monitor wavelength of 1064 nm) and (b) shows a zoom-in view of the SiO_2 transmission curve; The inset represents the glass substrate coating with a film stack of (HL)^4
图4 (a)透射曲线随 SiO_2 薄膜的沉积厚度(监控波长为1064 nm)而变化,(b) SiO_2 透射曲线的放大图;插图表示沉积有(HL)^4膜堆的玻璃基板

site. The results show that the on-site determination method can be applied to both high absorption materials and low absorption materials. The extinction coefficient of thin films can be obtained by the decrease of transmission peaks and spectrum shape fitting. The near-infrared optical constants n and k of high absorption materials Si are precisely obtained as 3.22 and 4.6×10^{-3} (SDF=0.78%) respectively at monitor wavelength of 1300 nm. The n and k of low absorption material Ta_2O_5 is also got to be 2.06 and 1.3×10^{-3} (SDF=0.12%) respectively at monitor wavelength of 890 nm. Meanwhile, this method is also valid for the film materials with refractive index close to the monitoring substrate by depositing a large refractive index difference film or film stack. The optical constants of SiO_2 are obtained as n=1.46 and k=6.6×10^{-5} (SDF=0.02%) at monitor wavelength of 1064 nm for demonstration. The results show that this method is sensitive enough for low-absorption materials, which is remarkably used for the design and fabrication of high-performance photonic devices.

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