Refractive index inhomogeneity of LaF_3 film at deep ultraviolet wavelength

Dawei Lin (林大伟)[1], Chun Guo (郭 春)[1,2], and Bincheng Li (李斌成)[1*]

[1] *Institude of Optics and Electrics, Chinese Academy of Sciences, Chengdu 610209, China*

[2] *University of the Chinese Academy of Sciences, Beijing 100039, China*

[*] *Corresponding author: bcli@ioe.ac.cn*

Received November 30, 2012; accepted January 4, 2013; posted online May 21, 2013

It is well known that the optical property of an optical thin film can be influenced by even small inhomogeneity of refractive index (RI). In order to investigate the RI inhomogeneity of LaF_3 single layer in deep ultraviolet (DUV) range, single-layer LaF_3 samples deposited on fused silica and CaF_2 substrates are prepared by resistive heating evaporation at different deposition temperatures. The reflectance and transmittance spectra of LaF_3 film samples are measured with a spectrophotometer, and used to calculate the RI inhomogeneity. The experimental results show that no RI inhomogeneity of LaF_3 film is observed when deposited on CaF_2 substrate, while negative RI inhomogeneity is presented when deposited on fused silica substrate. The level of inhomogeneity is affected by the substrate temperature, which decreases with the increasing substrate temperature from 250 to 400 °C.

OCIS codes: 310.6188, 310.6860.

doi: 10.3788/COL201311.S10602.

It is well known that the transmittance and reflectance spectra of an optical thin film can be influenced by even small inhomogeneity or variation in its complex refractive index (RI) profile[1]. Inhomogeneous layers can be either helpful or harmful to the optical performance of thin film. If the RI of an inhomogeneous film changes smoothly from the substrate to the ambient medium, it will be good for antireflection coatings. While in many multilayers, inhomogeneous film will be a problem to performance optimization and accurate thickness control[2]. An existing inhomogeneous model developed by Arndt *et al.*[3] was used to determine the optical constants from reflectance and transmittance measurements. In these studies the envelops of the reflectance and transmittance spectra of thin films were used to calculate the optical constants[4]. For optical layers with large thicknesses, RI inhomogeneities can be readily determined from spectral data[5].

LaF_3 is an important high-RI material used in deep ultraviolet (DUV) wavelength region. In the past the properties of LaF_3 films have been the focus of many research papers. Different deposition methods[6] were employed to prepare LaF_3 films. The relation between the crystalline structure and the substrate temperature[7], the micro-structure and laser-induced damage threshold (LIDT) characteristics of the LaF_3 films have been investigated. Several research papers also reported RI inhomogeneity of LaF_3 films. Liu *et al.*[8] accused the RI inhomogeneous behavior to the loose structure of LaF_3 film.

Experimentally, RI inhomogeneity, which is important to coating design and manufacturing, is a function of various deposition parameters. In this letter, LaF_3 films are prepared by thermal evaporation at different substrate temperatures. Fused silica and CaF_2 are used as the substrates. Transmittance and reflectance spectra are measured with a Perkin Elmer Lambda 1050 spectrophotometer. An envelope method[3,9−11] is applied to calculate the RI inhomogeneities of LaF_3 films deposited on fused silica with different substrate temperatures. RI inhomogeneities of LaF_3 films on fused silica and CaF_2 substrates are compared.

LaF_3 films were deposited by molybdenum boat evaporation. The base pressure of the coating machine chamber was pumped down to less than 2×10^{-4} Pa using a rotary pump and a cryo pump. The substrate temperatures ranged from 250 to 400 °C with a step of 50 °C. Deposition rate of thin film was 0.1 nm/s, controlled by a quartz oscillator controller. When the substrate temperature was 350 °C, fused silica and CaF_2 were both used as the substrates in one process. The optical thickness of all the films was set to six quarter waves at 193 nm. Before coating, the substrates were cleaned with a UV photo cleaner at two main working wavelengths, 185 and 254 nm, to remove hydrocarbon contamination. Spectra of the films were measured with a Lambda 1050 spectrophotometer whose short wavelength limit is 180 nm. Before the measurement, the film samples were cleaned with UV photo cleaner for 40 min.

Assume a weakly inhomogeneous film as shown in Fig. 1(a), where $n_{\rm in}$ is the RI of the film close to the substrate and $n_{\rm out}$ is that close to air. The average RI, defined by $n_{\rm av} = (n_{\rm in} + n_{\rm out})/2$, represents the RI of the film, and Δn, defined by $\Delta n = n_{\rm out} - n_{\rm in}$, represents the difference of RIs close to the substrate and close to air, respectively. A negative Δn indicates a negative RI inhomogeneity of the film.

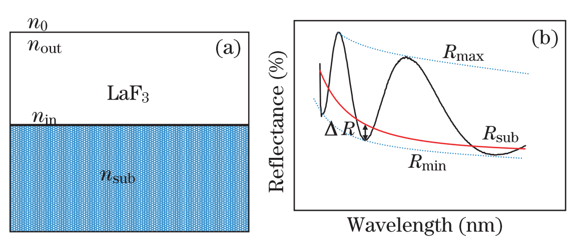

Fig. 1. (Color online) (a) Schematic diagram of inhomogeneous RI model and (b) reflectance of a negatively inhomogeneous film ($n_{\rm out} < n_{\rm in}$). $R_{\rm min}$ is below $R_{\rm sub}$.

In Fig. 1(b), the reflectance (R) of a negatively inhomogeneous film was schematically shown. Assuming $n_{\mathrm{out}} < n_{\mathrm{in}}$ and $n_{\mathrm{sub}} < n_{\mathrm{av}}$, the minimum reflectance ($R_{\min}$) of the film is smaller than the reflectance of the substrate (R_{sub}). Using a characteristic matrix[4],

$$\mathbf{M} = \begin{bmatrix} \left(\frac{n_{\mathrm{in}}}{n_{\mathrm{out}}}\right)^{\frac{1}{2}} \cos\delta & \frac{\mathrm{i}\sin\delta}{(n_{\mathrm{out}} n_{\mathrm{in}})^{\frac{1}{2}}} \\ \mathrm{i}\left(\frac{n_{\mathrm{in}}}{n_{\mathrm{out}}}\right)^{\frac{1}{2}} \sin\delta & \left(\frac{n_{\mathrm{in}}}{n_{\mathrm{out}}}\right)^{\frac{1}{2}} \cos\delta \end{bmatrix},$$

where δ is the phase thickness, the spectrum of the negatively inhomogeneous film can be calculated. The RI inhomogeneity, $\Delta n/n_{\mathrm{av}}$, can be express by

$$\frac{\Delta n}{n_{\mathrm{av}}} = \frac{n_{\mathrm{sub}}^2 - n_0^2}{4 n_0 n_{\mathrm{sub}}} \frac{\Delta R}{R_{\mathrm{sub}}},$$

where ΔR is the difference between R_{sub} and $R_{\min}$, and n_0 and n_{sub} are the RIs of the incident medium and the substrate, respectively. The RIs, n_{in} and n_{out}, can be calculated by an envelope method from the spectrophotometric measurements at extreme wavelengths[9].

LaF_3 films deposited on fused silica substrates at different substrate temperatures showed a RI inhomogeneity, as shown in Fig. 2. All LaF_3 films showed a negative RI inhomogeneity. The RI decreased as the substrate temperature increased. Then the maximum of transmittance spectrum was higher than that of the substrate due to antireflection effect.

The RIs of the LaF3 thin films were calculated by the envelope method using the maxima and minima of R and T at quarter and half wavelengths. RIs, n_{out}, and n_{in} of the films deposited at different substrate temperatures at 193 nm were summarized in Table 1.

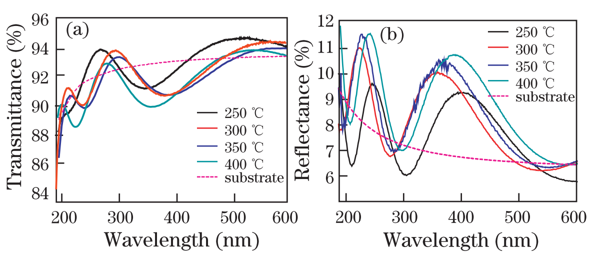

Fig. 2. (Color online) (a) Transmittance and (b) reflectance spectra of the bare fused silica substrate and LaF_3 films prepared at 250, 300, 350, and 400 °C, respectively.

Table 1. RIs, n_{out}, n_{in}, and Inhomogeneity of LaF_3 Films Prepared at Different Substrate Temperatures at 193 nm

Temperature (°C)	n_{out}	n_{in}	Δn	$\Delta n/n_{\mathrm{av}}$ (%)	d (nm)
250	1.5200	1.6888	0.1688	12.85	193.9
300	1.6066	1.7057	0.0991	5.98	171.2
350	1.6628	1.7215	0.0587	3.47	174.3
400	1.6906	1.7272	0.0366	2.14	170.1

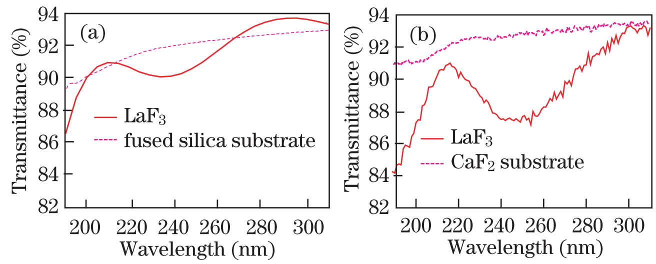

Fig. 3. (Color online) Transmittance spectra of LaF_3 films prepared at 350 °C on (a) fused silica and (b) CaF_2 substrate.

From Table 1 we can see that both n_{out} and n_{in} increase as the substrate temperature increases, indicating the overall RI of the film increases with the increasing substrate temperature. At the same time Δn, the difference between n_{out} and n_{in}, decreases with the increasing substrate temperature. Therefore, the RI inhomogeneity, $\Delta n/n_{\mathrm{av}}$, decreases as the substrate temperature increases. This is because as the substrate temperature increases, more thermal energy causes faster molecular movement, results in less voids in the film. Such a dense structure results in a high packing density, and consequently a decrease in the RI inhomogeneity behavior.

Figure 3 shows the transmittance spectra of LaF_3 films deposited on fused silica and CaF_2 substrates at substrate temperature of 350 °C. Clearly, LaF_3 film deposited on CaF_2 substrate presented negligible RI inhomogeneity, as compared to that deposited on fused silica substrate. This is due to the fact that fused silica has an amorphous structure, while CaF_2 has a cubic-crystalline structure. As LaF_3 material has the same crystalline structure as CaF_2, it could grow on CaF_2 substrate following homoepitaxy which had little dislocation in the film[12]. In this case the film grows homogeneously.

In conclusion, RI inhomogeneities of LaF_3 films prepared by molybdenum boat evaporation at different substrate temperatures and on different substrates are investigated. The experimental results reveal that LaF_3 films deposited on fused silica substrates present strong negative inhomogeneity of RI, with the inhomogeneity decreased as the substrate temperature increasing. On the other hand, LaF_3 films deposited on CaF_2 substrates show negligible RI inhomogeneity.

References

1. A. V. Tikhonravov, M. K. Trubetskov, B. T. Sullivan, and J. A. Dobrowolski, Appl. Opt. **36,** 7188 (1997).
2. J. A. Dobrowolski and P. G. Verly, Proc. SPIE **2046,** (1993).
3. D. P. Arndt, R. M. A. Azzam, J. M. Bennett, J. P. Borgogno, C. K. Carniglia, W. E. Case, J. A. Dobrowolski, U. J. Gibson, T. T. Hart, F. C. Ho, V. A. Hodgkin, W. P. Klapp, H. A. Macleod, E. Pelletier, M. K. Purvis, D. M. Quinn, D. H. Strome, R. Swenson, P. A. Temple, and T. F. Thonn, Appl. Opt. **23,** 3571 (1984).
4. B. Bovard, F. J. Van Milligen, M. J. Messerly, S. G. Saxe, and H. A. Macleod, Appl. Opt. **12,** 1803 (1985).
5. J. P. Borgogno, F. Flory, P. Roche, B. Schmitt, G. Albrand, E. Pelletier, and H. A. Macleod, Appl. Opt. **23,** 3567 (1984).

6. Y. Taki, Vacuum **74,** 431 (2004).
7. M. Vijayakumar, S. Selvasekarapandian, T. Gnanasekarah, S. Fujihara, and S. Koji, J. Fluorine Chem. **125,** 1119 (2004).
8. M. C. Liu, C. C. Lee, M. Kaneko, K. Nakahira, and Y. Takano, Opt. Eng. **45,** 083801 (2006).
9. J. H. Lee and C. K. Hwangbo, Surf. Coating. Tech. **128,** 280 (2000).
10. S. Humphrey, Appl. Opt. **21,** 4660 (2007).
11. N. K. Sahoo and A. P. Shapiro, Appl. Opt. **4,** 698 (1998).
12. Z. Wu and B. Wu, *The Growth of Thin Film* (in Chinese) (Science Press, Beijing, 2001) pp. 181-187.