ZrO_2 films with variable refractive index by glancing angle deposition

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When optical film is deposited at oblique angle incidences, the film becomes porously, and the film effective refractive index will decrease. Six porous ZrO_2 films are fabricated by the glancing angle deposition (GLAD) technique with the different incidence angles. By changing the substrate tilt angle from 0° to 85°, the film refractive index varies from 1.92 to 1.27.

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Material refractive index plays an important role in optical films. It influences the film optical characteristics, such as transmission spectrum, reflection spectrum etc.. In traditional films, the film refractive index is close to the bulk material refractive index, which is fixed. So, once the film material is determined, the film refractive index is unvariable. Porous film is a film mixing air and film material into the thin film, the porous structure serves to reduce the film effective refractive index. By changing the rate of the air and the film material, porous films with different refractive indices can be deposited.

Today, porous low refractive index films are mainly fabricated by electrochemical etching[1]. Here we use glancing angle deposition (GLAD) technique to fabricate porous low refractive index films. GLAD is a new way to fabricate porous films, and develops quickly in these years[2,3]. When films are deposited at oblique angle incidences, self-shadowing becomes the dominant growth mechanism. With the films becoming porous, the film effective refractive indices become lower than the bulk materials' refractive indices[4]. By changing the incidence angles, we can get different refractive indices.

The GLAD process of fabricating porous low refractive index films is shown in Fig. 1. When the film material

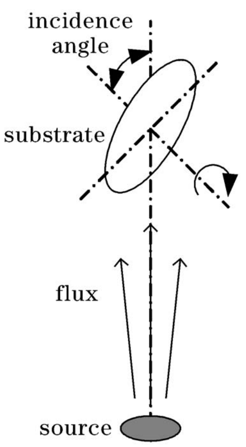

Fig. 1. Schematic diagram of the GLAD technique for physical vapour deposition onto rotating substrates.

vapoUr is non-normally incident onto the substrate, previously deposited material prevents subsequent deposition in its geometrical shadow, furthermore, the limited surface mobility of atoms prevents film growth in shadowed regions[5]. So the film becomes a mixture of air and film material. As the incidence angle increasing, the volume fraction of air increases, the film becomes more porous, and the effective refractive index of the mixed film becomes further lower.

The refractive index of the mixed film can be described by the effective medium theory model because the film column diameters are much smaller than the wavelength of interest[6,7]. The Bruggeman effective-medium approximation (EMA) applied to the particles A embedded in a continuous matrix of particles B is used here,

$$p_{\mathrm{A}}\frac{\varepsilon_{\mathrm{A}}-\varepsilon}{\varepsilon_{\mathrm{A}}+2\varepsilon}+p_{\mathrm{B}}\frac{\varepsilon_{\mathrm{B}}-\varepsilon}{\varepsilon_{\mathrm{B}}+2\varepsilon}=0, \quad (1)$$

where ε, ε_{A}, and ε_{B} are the dielectric functions of the effective medium, material A, and material B; P_{A} and P_{B} represent volume fractions of material A and B in the total volume, respectively. From Eq. (1) we can obtain the relationship of effective index of the mixed film and porosity.

Here, ZrO_2 was chosen to fabricate the porous low refractive index films by GLAD. ZrO_2 is transparent in the visible and near infrared frequency range, and is widely used as a high refractive index material in optical films. We fabricated 6 ZrO_2 samples with different incidence angles, and obtained their disperse spectra. The parameters of samples are listed in Table 1. The ZrO_2 volume fractions of the samples were obtained according to Eq. (1).

In our experiment, ZrO_2 was deposited by electron-beam deposition (Nanguang ZZX550). The films were deposited onto K9 glass substrates with different incidence angles varying from 0° to 85°, the working pressure was 3×10^{-3} Pa, and the working temperature was 40 °C. The incidence angles were controlled by a stepper which was controlled by a computer. Spectrometer Lambda 900 (PE Company) was used to measure reflection spectra of samples. Their spectra are shown in Fig. 2. For a single layer thin film, according to Fresnel formula[8],

Table 1. Incidence Angles, Refractive Indices at 1053 nm, and ZrO_2 Volume Fractions

Sample	Incidence Angle	Refractive Index (1053 nm)	ZrO_2 Volume Fraction
1	0	1.92	1
2	30	1.89	0.971
3	60	1.74	0.833
4	70	1.56	0.659
5	80	1.36	0.442
6	85	1.27	0.350

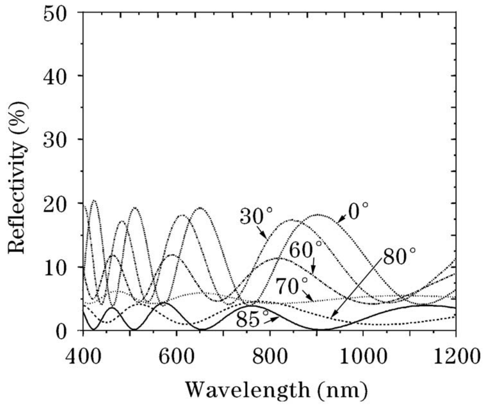

Fig. 2. Reflection spectra of different incidence angle deposited films.

the film refractive index can be calculated from the film reflectivity

$$R(\lambda) = \left(\frac{n_0 - n_f(\lambda)}{n_0 + n_f(\lambda)} - \frac{n_f(\lambda) - n_s}{n_f(\lambda) + n_s} \right)^2, \qquad (2)$$

where n_0 is the refractive index of air (equal to 1), $n_f(\lambda)$ is the refractive index of film material at wavelength λ, n_s is the refractive index of substrate (equal to 1.52). When n_f is higher than n_s, $R(\lambda)$ is at the peak value of reflection spectrum at wavelength λ; and when n_f is lower than n_s, $R(\lambda)$ is at the valley value of reflection spectrum at wavelength λ.

We calculated each sample's effective refractive indices at peak (valley) wavelengths, and then simulated the disperse spectra for each sample (see Fig. 3) according to Cauchy's disperse formula[8]

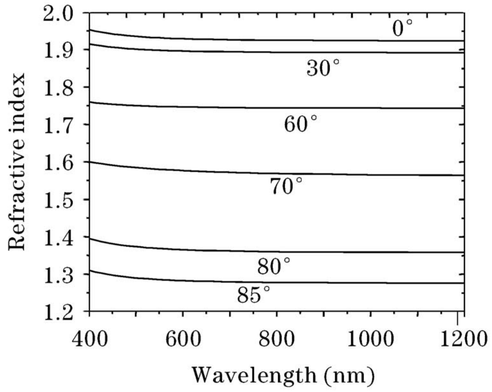

Fig. 3. Disperse spectra of different incidence angle deposited films.

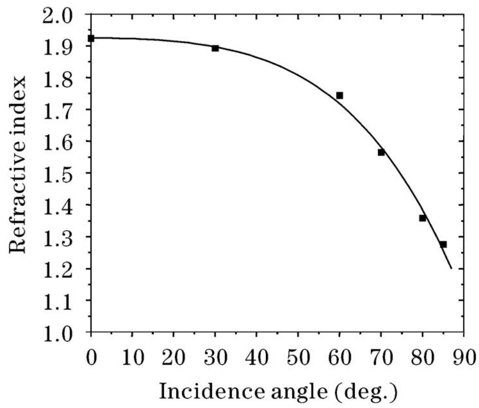

Fig. 4. Variation of effective refractive index with incidence angle at 1053 nm.

$$n(\lambda) = n + \frac{C}{\lambda^2} + \frac{D}{\lambda^4}, \qquad (3)$$

where the parameters n, C and D can be simulated from the refractive indices at peak (valley) wavelengths. It is found that as the incidence angle increases, the effective refractive index of the film will decrease. Figure 4 illustrates the variation of effective index with incidence angle at 1053 nm, with the incidence angle varying from 0° to 85°, the effective index varies from 1.92 to 1.27. As the incidence increases (see Table 1), the ZrO_2 volume fraction decreases, and the films become more porous. The relationship between the effective refractive of the mixed film and the substrate tilt angle is shown in Fig. 4. Any effective refractive index between 1.27 and 1.92 can be obtained by changing the substrate tilt angle[9].

In conclusion, we have shown an effective medium model describing the refractive indices of the thin films fabricated by GLAD. Six samples with different incidence angles changing from 0° to 85° and their effective refractive indices varying from 1.92 to 1.27 at 1053 nm are fabricated. For ZrO_2, by changing the incidence angle, we can get any refractive indices from 1.27 to 1.92, and this technology can be used to fabricate some new films, such as graded-index films, wide-band antireflection films.

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