第13卷　第4期
2005年8月

光学 精密工程
Optics and Precision Engineering

Vol. 13　No. 4
Aug. 2005

Article ID　1004-924X(2005)04-0505-07

Fabrication and characterization of rugate structures composed of SiO_2 and Nb_2O_5

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Abstract: Gradient index layers and rugate structures were fabricated on a *Leybold Syrus pro* deposition system by plasma-assisted coevaporation of the low index material silica and the high index material niobium pentoxide. To obtain information about the compositional profiles of the produced layers, cross sectional transmission electron microscopy was used in assistance to deposition rate data recorded by two independent crystal monitors during the film preparation. The depth dependent concentration profiles were transformed to refractive index gradients by means of effective medium approximation. Based on the refractive index gradients the corresponding samples' transmission and reflection spectra could be calculated by utilizing matrix formalism. The relevance of the established refractive index profiles could be verified by comparison of the calculated spectra with the measured ones.

Key words: physical vapor deposition; effective medium approximation; rugate structure

1　Introduction

Optical coating designs based on layers, which are inhomogeneous along the stack axis, have optical and mechanical properties that differ from those of conventional high-low-index stacks. Particularly, the wide accessible angular range and the low optical scatter level make them superior to traditional stacks with respect to selected purposes such as omni-directional devices or notch filters[1-4]. Manufacturing such systems in practice requires the calculation, deposition, monitoring and characterization of optical coatings with a well defined continuous refractive index profile perpendicular to the layer surface. Chemical vapor deposition processes have frequently been used to prepare such samples[5-7]. Ion assisted electron beam evaporation technology may also be used for such tasks when utilizing coevaporation processes from two sources. Thus the desired refractive index profile can be prepared by varying precisely the evaporation rates of both materials.

The coevaporation process results in the deposition of mixture layers with a refractive index which is determined at least by the refractive indices of the constituents of the mixture and their concentrations. For that reason, modeling of the refractive index profile starts from the investigation of the compositional profile. This strategy has been applied previously in applica-

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Received date: 2005-06-06; **Revised date:** 2005-06-16.

tion to other coating materials[8-11], and it will be the purpose of this work to apply it to reverse engineering of rugate structures built from niobia and silica. We emphasize that the choice of Nb_2O_5 (traditionally a sputtering material[12-13]) has been stimulated by recent success to deposit high quality Nb_2O_5 layers by means of ion assisted electron beam evaporation with the advanced plasma source (APS)[14].

The study pursues reverse engineering tasks in rugate structures. More concrete, the task is to determine the refractive index profile $n(\lambda, z)$ of an experimentally prepared rugate. This can not be done in an unambiguous manner from transmission and reflection spectra of the deposited film alone, because of the multiplicity of solutions of reverse engineering from spectrophotometric data of homogeneous layer systems[15] and even more of heterogeneous coatings[16]. Instead, it is very helpful to utilize a priori information on the refractive index profile as obtained from non-optical characterization techniques. We show the merit in refractive index profile determination based on deposition rate recordings during the film preparation and cross sectional TEM investigations of the deposited film.

2 Theoretical background

We are dealing with a material mixture composed from two consistents, numbered by j. Each of the pure materials has its wavelength-dependent refractive index $n_j(\lambda)$ and occupies a thickness depending volume fraction $V_j(z)$ of the full volume V of the mixture. Thus a volume filling factor $p_j(z)$ can be defined as a function of film thickness z:

$$p_j(z) = \frac{V_j(z)}{V}; \quad \sum_{j=1}^{2} p_j(z) = 1 . \qquad (1)$$

The refractive index $n_m(\lambda, z)$ of the composition may be calculated by means of effective medium theory drawn by Bruggeman[17]:

$$0 = \sum_{j=1}^{2} p_j(z) \frac{n_j^2(\lambda) - n_m^2(\lambda, z)}{n_j^2(\lambda) + 2n_m^2(\lambda, z)} , \qquad (2)$$

This approximation is suitable for mixtures, where the constituents can not be classified into inclusions and host[18]. In terms of Eq. (2), the dispersion of the refractive index of the mixture follows from that of the pure materials. Moreover, if $p_j(z)$ varies continuously, Eq. (3) automatically defines the spatial refractive index profile $n_m(\lambda, z)$ of the rugate. Hence, knowledge on the compositional profile (represented by $p_j(z)$) allows the calculation of the refractive index profile. The refractive indices of the individual film constituents SiO_2 and Nb_2O_5 were calculated from transmission and reflection spectra of homogeneous pure silica and niobia films. In the regarded wavelength range, absorption losses are negligible, so only purely real indices of refraction were used for these calculations.

3 Fabrication

The rugate structures built of silica and niobium pentoxide were prepared by APS assisted electron beam evaporation in a *Leybold Syrus* pro deposition system. The films were deposited on fused silica substrates for optical analysis and on silicon wafers for the cross sectional TEM examinations. Fig. 1 shows a sketch of the deposition principle. The deposition rates (and particular their dependence on time) are recorded automatically by the system by means of quartz crystal monitors. The base pressure was 6.3×10^{-4} Pa, while the plasma working pressure accounts for 4.5×10^{-2} Pa at a bias voltage of 140 V. The variable oxygen flow is controlled by its partial pressure. The deposition rate for SiO_2 was changed in the range 0.5～0.9 nm/s, while the rate for Nb_2O_5 varied alternating from 0.2 nm/s to 0.9 nm/s. It took 1 066 s for a ten period rugate structure.

Transmission and reflection spectra of the

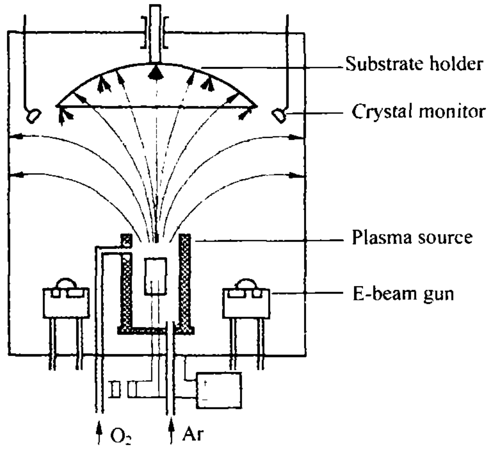


Fig. 1 Ion assisted coevaporation process

samples on fused silica have been recorded after deposition by means of a Perkin Elmer Lambda900 spectrophotometer. The samples on silicon have been utilized for TEM cross sectional investigations to estimate the composition of the samples from the TEM contrast.

4 Results

Fig. 2 shows the result of the deposition rate recording during preparation of the rugate.

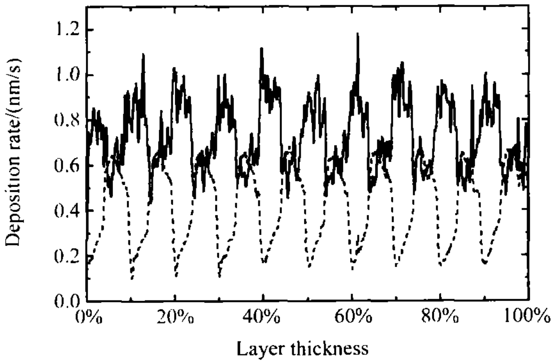


Fig. 2 Deposition rate vs. layer thickness (SiO_2 solid, Nb_2O_5 dashed)

The relation between the niobia and silica deposition rates r allows to calculate the compositional profile (Fig. 3) of the rugate structure in terms of the volume filling factors p of niobia and silica:

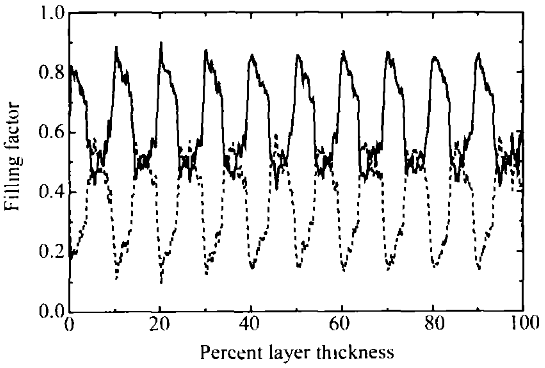


Fig. 3 Volume filling factors vs. layer thickness (SiO_2 solid, Nb_2O_5 dashed)

$$p(Nb_2O_5) = \frac{r(Nb_2O_5)}{r(Nb_2O_5) + r(SiO_2)}$$

$$p(SiO_2) = \frac{r(SiO_2)}{r(SiO_2) + r(Nb_2O_5)}, \qquad (3)$$

It is now straightforward to calculate the corresponding refractive index profile for any wavelength of interest by means of filling factors obtained from Eq. (3) and effective medium approximation Eq. (2). The profile shown in Fig. 4 is obtained for $\lambda = 550$ nm.

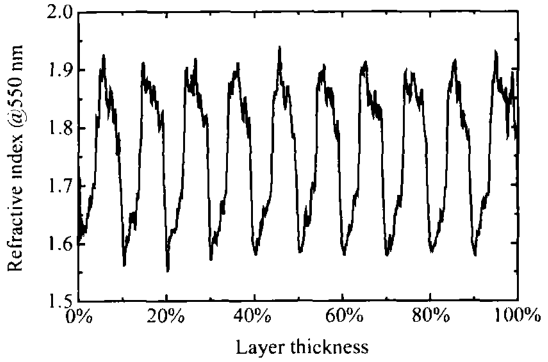


Fig. 4 Refractive index profile of the rugate structure obtained from rate recordings

The presented philosophy gives direct access to the refractive index profile. But one must keep in mind that the deposition rates result from an indirect monitoring procedure[19]. They are not recorded at the position of the growing film (which is held at a rotating substrate holder), but at fixed positions in the deposition chamber (see Fig. 1) and then corrected by means of tooling factors. Therefore, a cross-check of our results was performed, utilizing compositional profiles obtained from TEM investigations of the samples.

A cross sectional transmission electron picture of the rugate sample can be seen in Fig. 5. This image reveals a dark to bright contrast that corresponds to the compositional profile of the film. Particularly, the effect of the substrate holder rotation and the position of the e-beam gun appear as a fine substructure in the TEM image. However, the modulation period caused by the substrate holder rotation is in the region of 3.3 nm and thus too small to affect the optical film spectra at normal incidence and has not been taken into account for the further calculations.

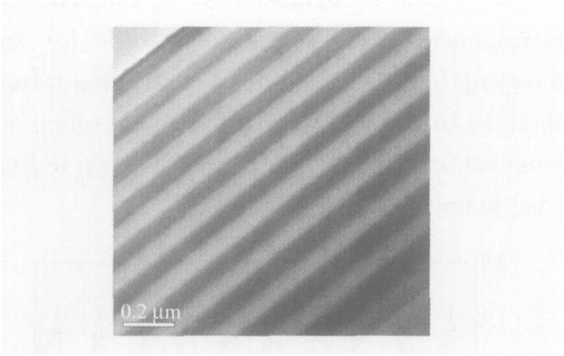


Fig. 5 Cross sectional TEM image of the rugate structure, courtesy of Ute Kaiser, University of Ulm, Germany

The shape of the TEM intensity profile is related to the compositional profile of the deposited materials. In order to relate this intensity profile to the material concentrations in a quantitative way, the curve has been calibrated with respect to the deposition rates at the beginning and the end of the deposition. We then obtain the material concentrations and the corresponding refractive index profile according to Eq. (3), which is plotted in Fig. 6 and differs in its shape from that determined from the rate recordings.

Having calculated the refractive index profiles corresponding to rate recordings and TEM images, the spectra (transmission or reflection) may be calculated by any thin film calculation software. The result of the transmission calculation is shown in Fig. 7 together with the measured spectra of the rugate. The full coating thickness has been tuned to reproduce the correct stopband position, which leads to a coating thickness of 1 484 nm for the rate-recording-based calculation, and 1 402 nm for the TEM-based calculation.

Both calculations are in a good qualitative agreement to the experimental spectrum and allow reproducing the main spectral features. But the fine details of the measured spectrum are not reproduced yet. The obtained refractive index profiles may therefore be regarded as an initial approximation to the real refractive index profile.

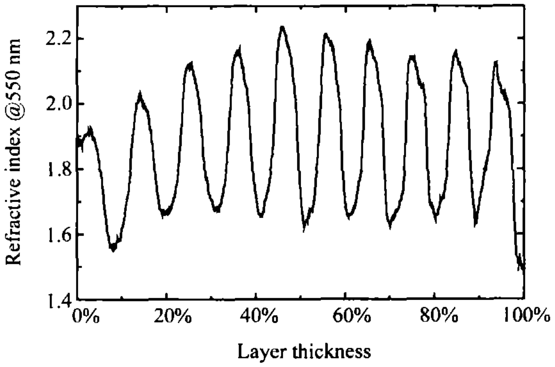


Fig. 6 Refractive index profile of the rugate structure obtained from rate recordings

5 Discussion

We demonstrated that non-optical data on the compositional profile can be utilized to obtain a first approximation of the refractive index profile for computation of its spectral properties. Analogous results have been achieved from deposition rate recordings as well as TEM investigations.

When discussing the merit of spectra reproduction, one must keep in mind, that the calcu-

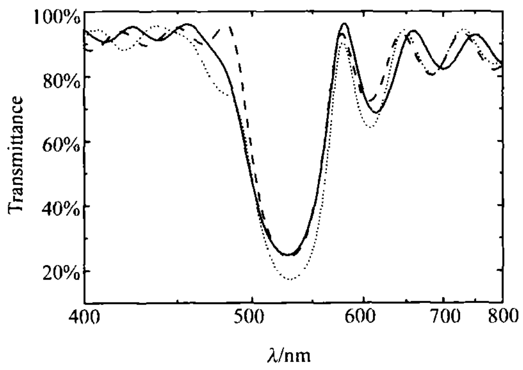


Fig. 7 Transmittance spectra of the rugate structure on fused silica (measured spectra (solid), calculated from deposition rate (dashed), calculated from TEM linescan (dotted)

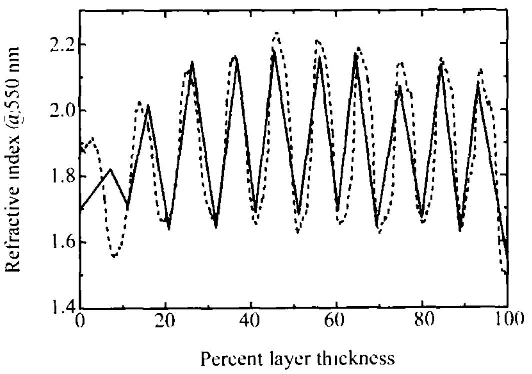
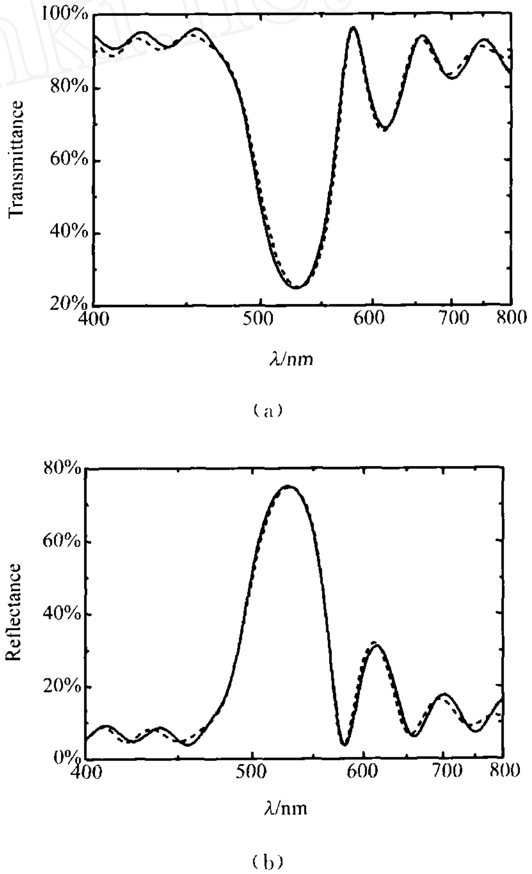


Fig. 8 Refractive index profile of the rugate from reverse engineering (solid) by means of TEMinvestigation as starting design (dashed)



(a)

(b)

Fig. 9 Comparison of transmittance (a) and reflectance (b) spectra from measurement (solid) and reverse engineering (dashed) by means of TEM data as initial design

lations of the transmission spectra from Fig. 7 do not represent spectra fits. Furthermore both methods essentially represent indirect characterization methods. The samples used for TEM investigation are not identical with those used for optical analysis, because they have been deposited onto different substrates. Rate recordings, on the other hand, are accomplished at fixed positions in the deposition chamber (see Fig. 1), and are corrected by means of simple tooling factors. Moreover, cross-correlations between the rate recordings for niobia and silica may occur due to insufficient screening of the crystal monitors from each other. All these problems lead to systematic errors, so that the merit of reproducing the experimental spectra is rather astonishing.

Practically, the established refractive index profiles are extremely helpful for reverse engineering, because they may be used as initial approximation for local search methods. This is exemplified for the rugate system (see Figs. 8 and 9). From Fig. 6 one may postulate that the rugate structure may be approximated by a sequence of positive and negative linear refractive index ramps (at 550 nm wavelength, as shown in Fig. 8). In terms of this assumption, the experimental spectrum from Fig. 7 may be fitted, varying only the corner points of the mentioned triangular shaped refractive index profile. As a result of the fit, we obtain a theoretical transmittance and reflectance as shown in Fig. 9, while the final parameters of the refractive index profile at 550 nm are given in Fig. 8. The fit quality is excellent.

Comparing the relative value of the rate recording and TEM data, is should be emphasized, that rate recording data is available in real time during deposition, and may be used for fast reverse engineering and even for rapid prototyping[20] of rugates. On the contrary, TEM cross sectional investigations are time-consuming and expansive. Hence, we favor the utilization of the rate recordings for rapid (in-situ) reverse engineering of rugates. Moreover, these data may be combined with in-situ broadband optical spectroscopy data of the growing film[21], to obtain a consistent and complete picture of the growth process of any coating. Similar information may be drawn from TEM, but only with serious time delay.

We come to the result, that the sophisticated evaluation of rate recordings and TEM images may be extremely helpful for reverse engineering of inhomogeneous optical coatings. The results are mutually consistent.

6 Summary

The refractive index profile of rugate structures built from SiO_2 and Nb_2O_5 may be estimated utilizing a combination of optical and non-optical characterization techniques. TEM cross sectional investigations delivered high quality profiles which are consistent with the main features of the measured sample transmission and could be utilized as starting approximation for the spectra fit. The same kind of information can be drawn from deposition rate recordings. On the other hand, TEM investigations cannot be used for online re-engineering during deposition, or rapid prototyping. For real time reverse engineering, the goal is to combine the fast rate-recording technique with an in-situ optical broadband monitoring during film deposition[22].

7 Acknowledgments

The authors thank to Heidi Haase for sample preparation, Prof. Ute Kaiser (University of Ulm, Germany) for TEM-performance and the BMWA for financial support. Vesna Janicki wishes to thank to the Fraunhofer Society in Germany for a Fraunhofer Fellowship at IOF in Jena.

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Brief professional biography of the author:

Robert Leitel has studied physics at the Friedrich Schiller University, Jena. His major field of research concerns inhomogeneous optical thin films. Actually, he works on his PhD at the Institute for Applied Physics, focusing on omnidirectional antireflection coatings.