Apparatus for the controlled deposition of multilayer films

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APPARATUS FOR THE CONTROLLED DEPOSITION OF MULTILAYER FILMS

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Résumé. — On décrit un appareillage simple et stable pour contrôler la préparation de couches multiples sous vide. De façon à comparer les facteurs de transmission ou de réflexion d'une couche multiple pour deux longueurs d'onde, on utilise un faisceau lumineux modulé, deux filtres interférentiels, deux photomultiplicateurs avec amplificateurs et un amplificateur différentiel. On a utilisé l'appareil dans un domaine de longueur d'onde s'étendant de l'ultraviolet au proche infrarouge, et on a adapté de manière convenable la source lumineuse et le photomultiplicateur. La reproductibilité des dépôts était de $\pm\ \lambda_0/65$ pour le filtre à bande étroite d'un film à 13 couches.

Abstract. — A simple and stable apparatus for the controlled deposition of multilayer films in vacuo is described. In order to compare transmittance or reflectance of a multilayer film at two wavelength bands, the apparatus is equipped with a modulated light beam, two interference filters, two photo-tubes with amplifiers and a differential amplifier. The apparatus has been used in a wavelength range from the ultraviolet to the near infrared with suitable adaptation of the light source and the photo-tubes. Reproducibility of the deposition was $\pm\ \lambda_0/65$ for the narrow band-pass filter of a thirteen-layer film.

1. **Introduction.** — In the field of applied optics a great deal of interest has recently been shown in the properties of multilayer interference films for use as high-reflecting mirrors, band pass filters, etc. To meet this requirement, precise control of film thickness in multi-layer films is very important. For the controlled deposition of multilayer films the photometric method seems to be most convenient, though various methods for the controlled deposition of thin films in vacuo have been described by many authors [1] a [6]. An apparatus for the controlled deposition of multilayer films constructed by the present authors is based on the comparison of transmittances or reflectances of a multi-layer film for two beams of different wavelength during the deposition [7]. The apparatus can be used in the near infrared after a simple replacement of photo-tubes by photoconductive cells, and can also be used in the ultraviolet by applying optical materials transparent to ultraviolet light.

2. **Design considerations and construction of the apparatus.** — The transmittance or reflectance curve of a stack of quarter-wave films, alternately of high and low index, plotted as a function of wave number is symmetrical with respect to the central wave number [1]. When the optical thickness of the top layer of the multi-layer films is increasing from zero to a quarter wavelength, transmittance or reflectance of the resultant film varies with the thickness in a regular way as will be shown in a later chapter. Therefore, the apparatus was designed in such a way it could measure the difference of transmittances or reflectances of the resultant film at two wavelengths, each of which is situated at a suitable and equal wave number distance from the central wavelength. During the deposition, the thickness of each individula layer can be controlled by observing the indicating meter, which shows subsequent zero differences at those moments when the optical thick-

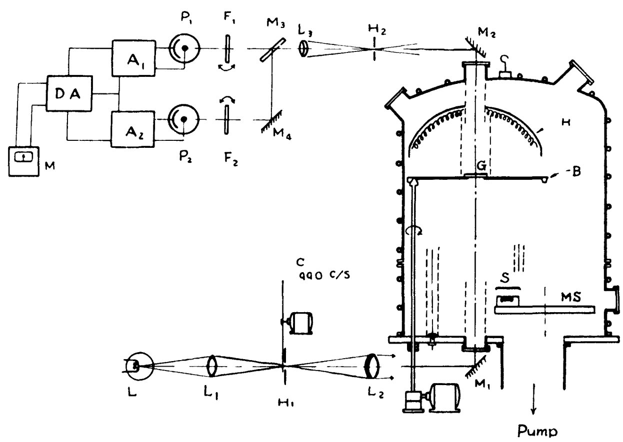

FIG. 1.

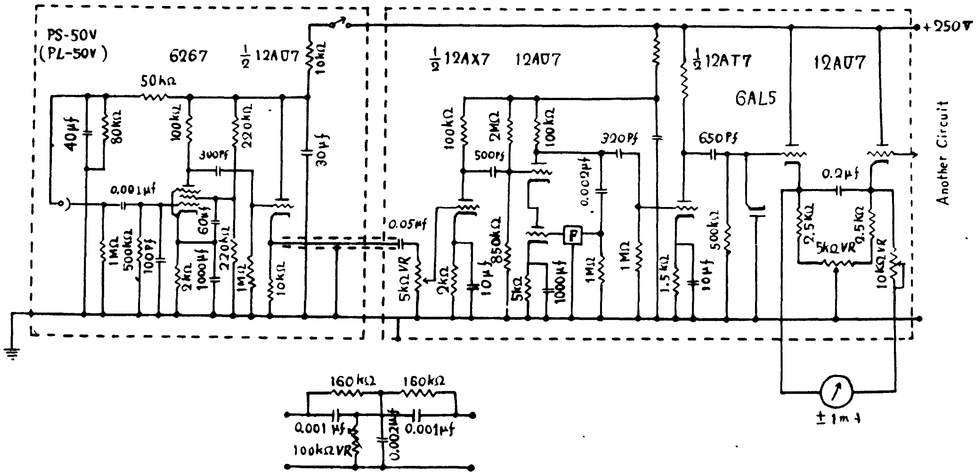

FIG. 2.

TABLE 1

PERFORMANCE OF THE PHOTOMETER AMPLIFIER

Peaked frequency	990 c/s on 60 c/s mains 1 000 c/s on 50 c/s mains
Bandwidth	about 100 c/s
Noise in the amplifier with the maximum gain	less than 0.5 % of the maximum meter reading
Dark current noise for Sb-Cs photo-tube	less than 0.5 % »
» Ag-Cs photo-tube	about 0.5 % »
» Pbs photo-cell	about 0.5 % »
Fluctuation for light beam giving the maximum meter reading with the maximum gain with	
Sb-Cs photo-tube	about 1 % »
Ag-Cs photo-tube	about 1.5 % »
Pbs photo-cell	about 1.5 % »

nesses have just become a quarter wavelength.

The optical system combined with an evaporation chamber is schematically shown in figure 1. The light source L is a car lamp, and the diameter of the small aperture H_1 is about 0.5 mm. The electric current to the lamp is supplied through a ballast tube from a transformer which is connected to a stabilized a. c. voltage supply. To eliminate any influence of light from other sources, the light beam is modulated with a chopper disk at a frequency of about 1 000 c/s.

After passing the multi-layer film to be deposited, the light beam is devided into two beams by a beam splitter of an evaporated Al film. After passing ghrough conventional interference filters of metal films and dielectric films and if necessary further passing through glass filters, each beam falls on to a photo-tube. Each filter system can be tilted with respect to the optical axis by rotating the holding stand, so that the pass-band may be slightly shifted. Adoption of this photometer system enables the apparatus to be simple and stable, and to be used in a wider wavelength range than that of one photo-tube system with rotating mirrors or filters [3].

The signal from each photo-tube is amplified by a preamplifier and by a frequency-selective amplifier containing a twin-*T* network which is tuned to the modulation frequency of the beam [8]. Then outputs from two amplifiers are compared with each other in a differental amplifier. In order to obtain a high signal-to-noise ratio with a high input resistance and a narrow band-width, it is desirable to select a rather low frequency, for which 1 000 c/s was chosen [4].

The circuit of the photometer amplifier is shown in figure 2. Tubes of pre-amplifiers are heated by a d. c. voltage supply. Gain-control rheostats in the main amplifiers serve for zero adjustment of deflection of the indicating meter before the deposition of film. Performance ot the photometer is shown in Table 1.

3. **Choice of observation wavelengths for the deposition control.** — Let us suppose that the deposition of the top or the *k*-th layer on a stack

of quarter-wave films, alternately of high and low index, is controlled by a transmitted light beam of normal incidence [7]. Writing δ_k for the change in phase of light of wavelength λ on traversing the k-th film of optical thickness $(nt)_k$, we kave

$$\delta_k = 2\pi(nt)_k/\lambda. \tag{1}$$

With the apparatus the transmittances (T_k) in two wavelength bands are compared with each other. They become equal when the k-th layer just becomes a quarter of the central wavelength λ_0. Writing λ_1 and λ_2, for the central wavelengths of two observation bands, we obtain for the sensitivity S_T of the deposition control the following expression :

$$S_T = \left| T_k \times \frac{\partial T_k}{\partial (nt)_k} \right|_{(\lambda=\lambda_1)} + \left| T_k \times \frac{\partial T_k}{\partial (nt)_k} \right|_{(\lambda=\lambda_2)}$$

$$= \frac{2\pi}{\lambda_0} \times T_k \times \left| \frac{\partial T_k}{\partial \delta_k} \right| \times \left\{ \frac{\lambda_0}{\lambda_1} + \frac{\lambda_0}{\lambda_2} \right\} \tag{2}$$

$$= \frac{4\pi}{\lambda_0} \times T_k \times \left| \frac{\partial T_k}{\partial \delta_k} \right|. \tag{3}$$

On each side of the so-called pass-band of the resultant k-layer film, there are a principal maximum and $(k - 1)$ secondary maxima of $\partial T_k/\partial\delta_k$. However, variation of transmittance around secondary maxima of $\partial T_k/\partial\delta_k$ is so complicated that it can not be used for the deposition control, as is apparent from (*fig.* 3), which shows the change of

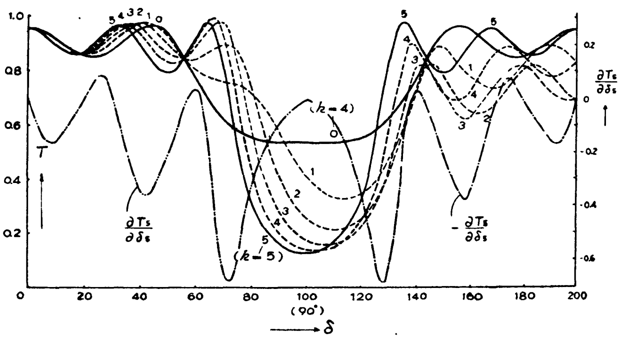

FIG. 3.

the calculated transmittance with a stepped increases of $\lambda_0/20$ of the top-layer's thickness of five-layer film of quarter-wave films, alternately of ZnS $(n = 2.35)$ and MgF_2 $(n = 1.40)$. In figure 4, spectral transmittance of the k-layer film and spectral sensitivity of the deposition control are shown. Although different filters for each individual layer are theoretically required, it is more practical to choose the observation filter for the last layer and use this throughout the deposition of the subsequent layers, since exchange of filters to the most fitting ones for these layers during the deposition is very difficult.

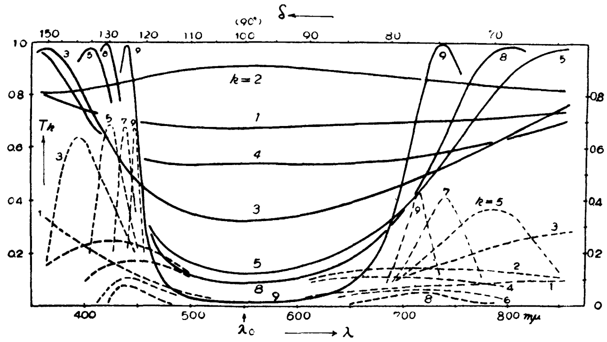

FIG. 4.

4. **Application of the apparatus.** — It was found that the deposition of a stack of quarter-wave and three-quarter-wave films, alternately of high and low index, could be controlled with the apparatus in a similar way as the deposition of a stack of quarter-wave films. In order to control the deposition of a narrow band-pass filter containing a spacer layer ot a half-wave film with the apparatus, a monitoring substrate had to be exchanged to a new one before and after, or in the middle of the process of depositing the spacer layer. This seems to be based on the fact that after deposition of the space layer a narrow pass-band appears around the central wavelength and useful regularity of variation of transmittance for the deposition control appears only around the narrow pass-band. Reproducibility was within $\pm$ $\lambda_0/65$ for the deposition of the first-order narrow band-pass filter of a thirteen-layer film of ZnS and MgF_2 [7].

The deposition of heat reflecting films [9] could be controlled satisfactorily by means of replacing one of the photo-tube with a PbS photo-conductive

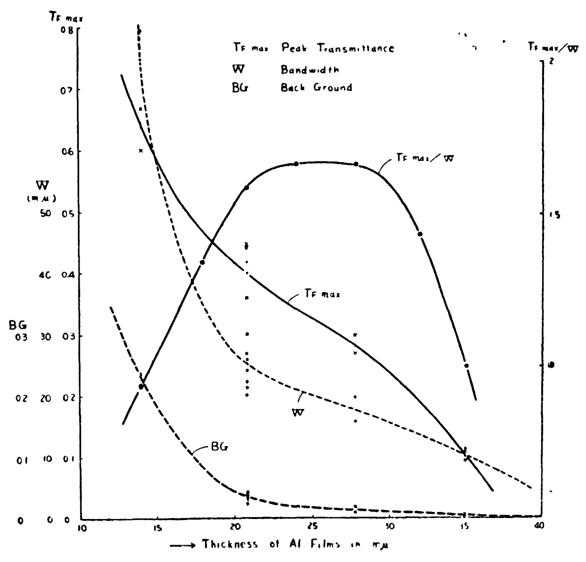

FIG. 5.

cell which is more suitable for the longer wavelengths. The deposition of high-reflecting films for the near infrared, such as films for gas lasers [10], could also be controlled with exchanging two phototubes for two photo-conductive cells. For these purposes the authors employed CeO_2 for high index films and MgF_2 for low index films.

The apparatus has also been applied for the deposition of multi-layer films for the ultraviolet by means of replacing the white light lamp with a hydrogen discharge tube, and optical glass with fused quartz. Interference filters for observation bands in the ultraviolet were made of Al films and MgF_2 films. Measured characteristics around 300 mμ of the second-order filters are shown in figure 5 as a function of the thickness of Al films. On the basis of this preliminary experiment, thicknesses of Al films were chosen to be about 25 mμ.

High reflecting films for the Fabry-Perot interferometer in a wavelength range from 300 mμ to 450 mμ were made with Sb_2O_3 films and MgF_2 films. A non-absorbing film of Sb_2O_3 is deposited with a metal boat source covered with alumina by means of flame-spraying [11]. Some of the results are shown in figure 6.

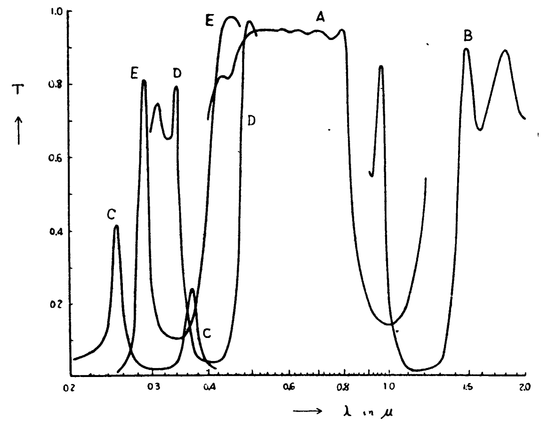

FIG. 6. — A : 8-layer film of CeO_2 (λ/8) — CeO_2 (λ/4) — MgF_2 (λ/4)..... for heat reflecting filter, B : 11-layer film of CeO_2 (λ/4) — MgF_2 (λ/4)..... for high-reflecting film for gas laser. C : Interference filter of Al — MgF_2 — Al. D : 11-layer film of Sb_2O_3 (λ/4) — MgF_2(λ/4)..... for Etalon. E : 7-layer film of Sb_2O_3(λ/4) MgF_2λ/4)..... for a narrow band filter.

5. Conclusions.

— Design and construction of apparatus for the controlled deposition of multi-layer films are described together with the results. Adoption of the photometer system with two photo-tubes enabled the apparatus to be used in a wavelength range from the ultraviolet to the near infrared. Adoption of inteference filters for observation bands and of the method of gain-control of the amplifiers for the zero adjustment of the deposition also made the apparatus simple and stable.

However, for a narrow band-pass filter the reproducibility of $\pm \lambda_0/65$ is not so satisfactory as that of more complicated apparatuses [4]. This can however be considerably improved with improvements of stabilized voltage supply for the light source and of the signal-to-noise ratio of the photometer amplifier. A further study based on this consideration is now in progress.

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