On the production and properties of inhomogeneous thin films

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ON THE PRODUCTION AND PROPERTIES OF INHOMOGENEOUS THIN FILMS

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Résumé. — La fabrication des couches minces inhomogènes par évaporation thermique en employant le contrôle automatique est discutée et les types différents des profils d'indice résultants sont considérés. Les propriétés optiques sont calculées par l'intégration numérique de l'équation d'onde.

Abstract. — The production of inhomogeneous thin films by evaporation using feedback control is discussed and the different types of resulting refractive index profiles considered. The optical properties are calculated by numerical integration of the wave-equation.

1. **Introduction.** — Wave — propagation in plane — parallel, inhomogeneous layers has been studied by many authors and in many different connections. Some of the first investigations were made in the middle of the nineteenth century in order to explain the reflection of light from polished glass surfaces in terms of the so called transition layer. Later investigations were concerned with atmospheric reflection of light and radio-waves and many other problems and today the literature on the subject is very extensive.

Already in 1880 lord Rayleigh [1] observed the remarkable reflection-reducing properties of an inhomogeneous film with the index varying continuously from that of glass to that of air. Since such a film cannot be realized with known materials it is not of practical interest. However, by combining an inhomogeneous film with a homogeneous film as suggested by Nadeau [2] and Strong [3] efficient reflection-reducing coatings may be produced.

Quite apart from the eventual practical applications in anti-reflection layers and interference filters, it is of interest to know the optical properties of inhomogeneous films. In order to calculate e. g. the reflectance of a film at normal incidence and with refractive index $n(z)$, where z is the coordinate perpendicular to the film surface, one has to solve the one-dimensional wave-equation for the electric field strength U

$$\frac{\mathrm{d}^2 U}{\mathrm{d}z^2} + \left(\frac{2\pi}{\lambda} n(z)\right)^2 U = 0. \tag{1}$$

Simple, approximate solutions exist when the film thickness is small compared to the wavelenth λ or when the index changes slowly with z. If this is not the case, explicit solutions in terms of known functions may be found only for certain functions $n(z)$, and as a rule they are quite complicated. An approach which may sometimes be very useful is to replace eq. (1) by an approximate equation which can be solved exactly [9] but also this method often leads to complicated expressions. The starting point of previous calculations has been to select a particular $n(z)$ which gives as simple solutions as possible. Among the refractive index profiles studied are those which are hyperbolic [1, 4, 5], linear [6], and exponential [7, 8] functions of z. The expressions for $n(z)$ in these cases contain two parameters, their values beeing determined by the refractive index values at the boundaries.

We have adopted a somewhat different starting point by first considering the experimental possibilities of making the layers and then deriving an expression for $n(z)$ which is based on these considerations.

2. **Production of inhomogeneous layers.** — Inhomogeneous layers may be produced by varying the rates of evaporation from two sources which are operated simultaneously and contain substances with different indices. In order to be able to vary the rates in a prescribed way, a feedback system has to be used. We have built a system similar in principle to that described by Thun and al. [10], see (*fig.* 1). The reference voltage is taken from a special program unit and by chosing it in a suitable way the rates may be varied as desired. As rate sensors we use oscillating quartz crystals, which means that we have to transform the frequency change, which is proportional to the mass (or thickness if the density is constant) deposited on the crystal, to a voltage (or current). The derivative of this voltage is proportional to the rate of deposition on the crystal, and is compared with the reference voltage in the usual way to give an error signal to the amplifiers and source power supply. The evaporation sources used are ordinary alumina crucibles and carbon boats. If the

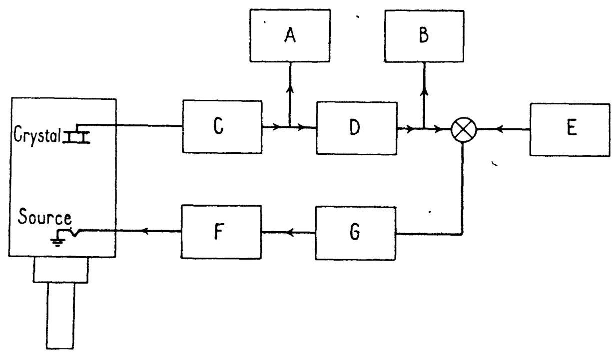

FIG. 1. — The block diagram of the control system. A, Recorder. — B, Recorder. — C, Frequency to voltage transformation. — D, Derivation. — E, Program unit. — F, Magnetic amplifiers. — G, Ampl. and stabilizing networks.

reference voltage is constant the deposition rate is kept constant by the control system. From the rate and thickness curves recorded by Varian model 10 G recorders it seems as if rates of 10 — 20 Å/s are kept constant within about 1 %. If the reference voltage is varied linearly with time, the thickness should be a parabolic function of time. Experimental curves for this case are shown in figure 2. The curves are direct copies from the recorder-paper but with another time-scale. The curve for the rate follows a straight line but the curve for the thickness differs a little from a parabola at low rates. (The crosses lie on a parabola through the end-points of the curve.) The deviations are probably due to the bad definition of the lower end-points of the curves. By using this method of recording it is not possible to study the linearity of the variation of the rate because small deviations from the average slope a straight line are very difficult to see. In order to make a reliable check of the accuracy of the control system it would be desirable to use a printing counter. A full account of the control system and its applications will be given elsewhere [11].

3. **A class of refractive index profiles.** — It was shown in a previous paper [12] that if the evaporation rates are varied linearly with time from $v_1(0)$ to $v_1(T)$ Å/s for source no. 1 and from $v_2(0)$

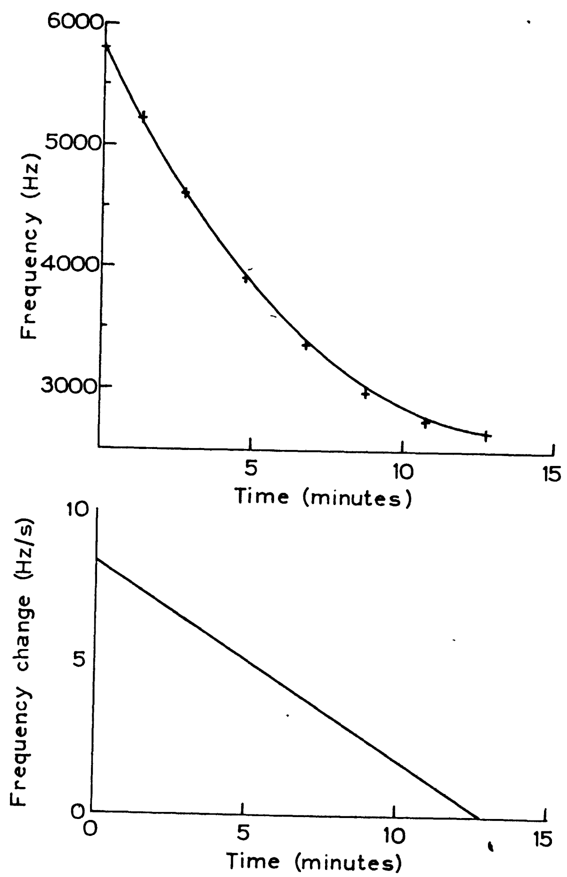

FIG. 2. — Curves showing the frequency and rate of change of the frequency with a linear variation of the reference signal.

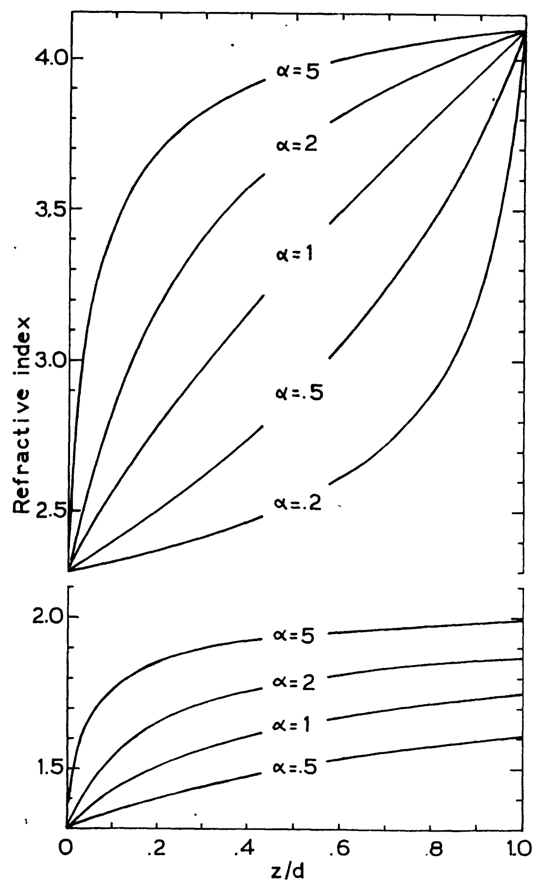

FIG. 3. — Refractive index profiles obtained by varying the evaporation rates linearly with time. Upper curves : both rates varied. Lower curves : one rate varied, the other constant.

to $v_2(T)$ Å/s for source no. 2 the following formula is valid under certain conditions :

$$n^2(z) = \frac{n^2(0) - \alpha' n^2(d)}{1 - \alpha'} + \frac{\alpha'[n^2(d) - n^2(0)]}{(1 - \alpha')\,[1 + (\alpha'^2 - 1)\,(z/d)]^{1/2}} \quad (2)$$

where d is the total film-thickness deposited during time T and

$$n^2(0) = (n_1^2 + \gamma n_2^2)/(1 + \gamma) \quad (3a)$$

$$n^2(d) = (n_1^2 + \beta n_2^2)/(1 + \beta) \quad (3b)$$

$$\gamma = v_2(T)/v_1(T), \qquad \beta = v_2(0)/v_1(0) \quad (4a)$$

$$\alpha' = \frac{v_1(0) + v_2(0)}{v_1(T) + v_2(T)} \quad (4b)$$

The refractive indices of the pure substances are n_1 and n_2. By using different values of the parameters, different refractive index profiles are obtained, see (*fig.* 3). Taking $v_1(0) = v_2(T) = 0$, $\alpha = \alpha'$ gives the curves in the upper part of the figure and $v_1(0) = v_1(T)$, $v_2(T) = 0$, $\alpha = \alpha' - 1$ those in the lower part.

4. **Calculation of the optical properties.** — The calculation of the optical properties of an inhomogeneous layer in general implies a great deal of numerical work, and without the capacity of the electronic digital computer it would be possible to perform but very incomplete investigations. Because of the numerical difficulties it is essential to represent the solutions of eq. (1) in a practical way. This may be done by the matrix theory of Abélès [13] which caracterises an inhomogeneous layer by a matrix the elements of which are solutions of the wave-equation (1) with certain boundary conditions. As is well known, the matrix elements of a homogeneous layer are sine or cosine functions of the variable $2\pi nd/\lambda$. We have calculated the matrix elements for various inhomogeneous layers. Figure 4 shows the results for the film with $\alpha = 2$ in the upper part of figure 3. The matrix is supposed to be

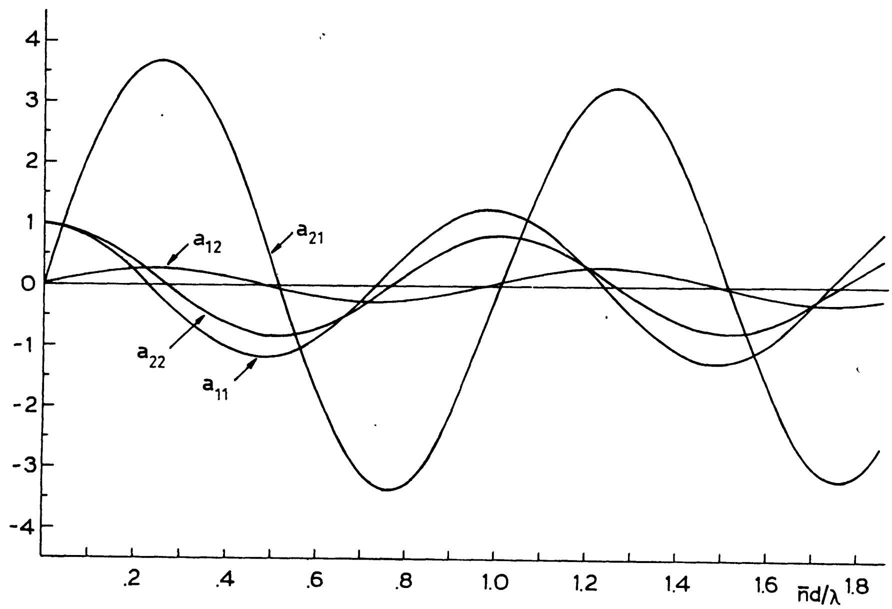

FIG. 4. — Matrix elements of a film with $n(0) = 2.3$, $n(d) = 4.1$, and $\alpha' = 2$.

$$M = \begin{bmatrix} a_{11} & ia_{12} \\ ia_{21} & a_{22} \end{bmatrix} \quad (5)$$

with a_{ik} real. The curves differ from those of a homogeneous film in that the extreme-value and zeros occur at other values of the optical thickness and that the heights of the extreme-values are not constant. Figure 5 shows the complex reflection coefficient of the same film, surrounded by media with index 1 and 1.5. The broken curve is for a homogeneous film with the same mean-index.

5. **Reflection-reducing coatings using inhomogeneous layers.** — As a simple application of inhomogeneous films we have studied a double layer consisting of one homogeneous $\lambda/4$ — film with index 1.52 and one inhomogeneous $\lambda/2$ — film with index varying from 2.31 to 1.52 which is the index of the substrate [2, 3], see (*fig.* 6).

This has also been done by Blaisse [14], who used a hyperbolic profile for the inhomogeneous film. We have calculated the reflectance for a few different profiles. Obviously, the profile influences the reflectance quite strongly. This is to be expected, partly because the mean-index of the inhomogeneous film depends on the profile. The broken curve shows a double layer with two homogeneous films of indices 1.52 and 2.05 ; this curve has a maximum equal to the reflectance of the bare substrate which is not the case when the high index film is inhomogeneous.

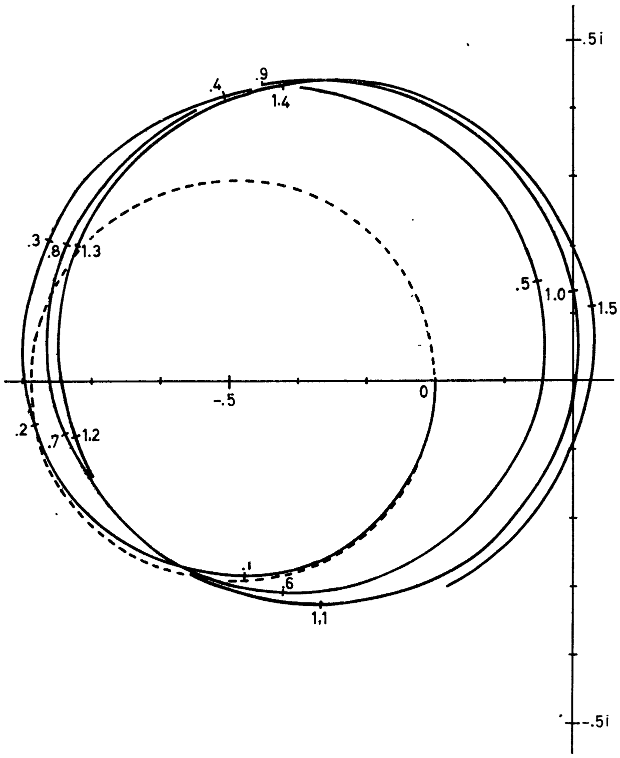

FIG. 5. — Complex reflection coefficient of inhomogeneous film with $n(0) = 2.3$, $n(d) = 4.1$, and $\alpha' = 2$ on glass with index 1.5. Broken curve : homogeneous film with index equal to the mean index of the inhomogeneous film.

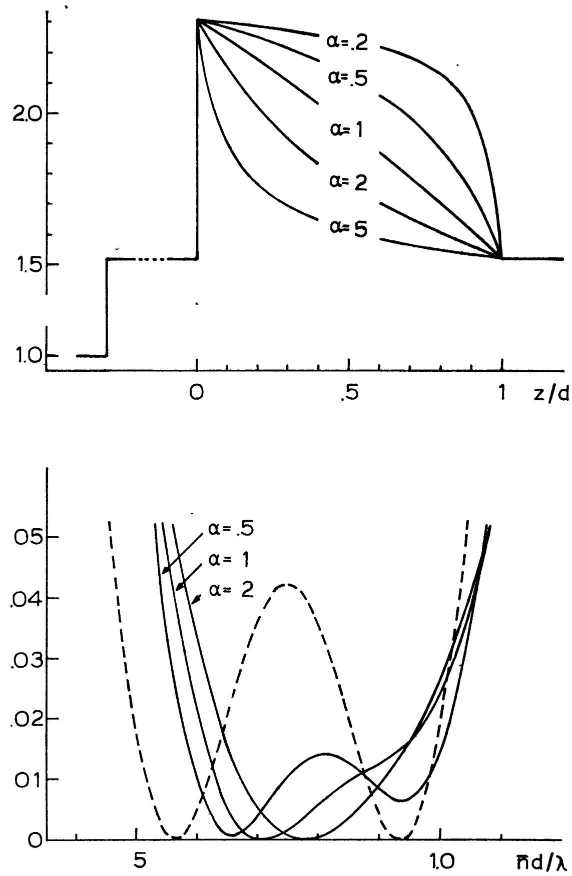

FIG. 6. — Refractive index and reflectance of double-layer coating with one film inhomogeneous. nd is the total optical thickness of the double layer.

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Discussion

M. HAHN. — 1) How do you control the thickness of films ?

2) Which materials were used for visible light ?

Réponse : 1) There is no direct control of the thickness of the inhomogeneous film during deposition. The thickness may be obtained from the specific functions $v_1(t)$ and $v_2(t)$ according to which the rates are varied. A check on the final thickness may be obtained by using a third crystal upon which the inhomogeneous layer is deposited.

2) We have used zinc sulfide and cryolithe but there are other materials which may give films with better mechanical properties, e.g. cerium oxide and cerium fluoride.

M. WEINBERGER. — What is the theoretical basis for the formulae offered for the optical constants of a non homogeneous films ? In his reply the speaker offered the formula

$$n^2 - 1 = AN_1 + BN_2.$$

However his definition of A and B which are constants for the materials at a particuliar wavelength were not clear to me. An elaboration and justification of the above formula would be appreciated.

Réponse : The formula for the refractive index profile was deduced under the following assumptions :

1) The rates $v_1(t)$ and $v_2(T)$ Å/s from the two sources are varied linearly with time.

2) The volume of a mixture of the two substances deposited is equal to the sum of the volumes of the two components deposited separately.

3) The refractive index as a function of composition of the mixture is described by the formula

$$n^2 - 1 = AN_1 + BN_2 \tag{1}$$

where A and B are constants, that is dispersion is neglected, N_1 and N_2 are the number of atoms per unit volume of the mixture. If n_1, ρ_1, and n_2, ρ_2 are the indices and densities of the pure substances and C_2 is the concentration of substance n° 2 in parts by weight

$$n^2(C_2) = \frac{\frac{n_1^2}{\rho_1}\left(\frac{1}{C_2} - 1\right) + \frac{n_2^2}{\rho_2}}{\frac{1}{\rho_1}\left(\frac{1}{C_2} - 1\right) + \frac{1}{\rho_2}}. \tag{2}$$

The full discussion of these questions may be found in the following references.

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