



HAL
open science

A STATISTICAL THEORY FOR THE DIELECTRIC PROPERTIES OF THIN ISLAND FILMS, APPLICATION AND COMPARISON WITH EXPERIMENTAL RESULTS

D. Bedeaux, J. Vlieger

► **To cite this version:**

D. Bedeaux, J. Vlieger. A STATISTICAL THEORY FOR THE DIELECTRIC PROPERTIES OF THIN ISLAND FILMS, APPLICATION AND COMPARISON WITH EXPERIMENTAL RESULTS. Journal de Physique Colloques, 1983, 44 (C10), pp.C10-379-C10-382. 10.1051/jphyscol:19831077 . jpa-00223535

HAL Id: jpa-00223535

<https://hal.science/jpa-00223535>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

A STATISTICAL THEORY FOR THE DIELECTRIC PROPERTIES OF THIN ISLAND FILMS, APPLICATION AND COMPARISON WITH EXPERIMENTAL RESULTS

D. Bedeaux and J. Vlieger*

Institutt for Teoretisk Fysikk, Universitetet i Trondheim, Norges Tekniske Høgskole, N 7034 Trondheim-NTH, Norway

**Instituut-Lorentz voor Theoretische Natuurkunde, Rijksuniversiteit te Leiden, Nieuwsteeg 18, 2311 SB Leiden, The Netherlands*

Résumé - A partir de photographies de microscopie électronique de films minces et discontinus d'or, étudiés expérimentalement par Norrman et collaborateurs, on calcule la transmittivité à l'aide d'une théorie statistique présentée précédemment. Les résultats sont en assez bon accord avec les valeurs expérimentales.

Abstract - Using the electron micrographs of thin discontinuous goldfilms, studied experimentally by Norrman et al., we calculated the transmittance on the basis of a statistical theory given in a previous paper. The results agree rather well with the experimental values.

The statistical theory¹⁾ for the dielectric susceptibilities of a thin island film is used to calculate the transmittance of normally incident light through gold films on a glass substrate²⁾. Taking local fields into account, the dielectric susceptibility parallel to the substrate is found to have the following general form

$$\gamma = \alpha_{\parallel}^s \left(1 - \frac{1}{3} \kappa_{\parallel} \alpha_{\parallel}^s\right)^{-1} \tag{1}$$

where α_{\parallel}^s is the average polarizability of the islands parallel to the surface of the substrate per unit of surface area. Furthermore, κ_{\parallel} has the dimensionality of an inverse length and accounts for local field effects parallel to the surface. An explicit expression for κ_{\parallel} will be given below. The transmittance of normally incident light, divided by the Fresnel value, is given in terms of γ by³⁾

$$T = \left[1 + \frac{4\pi}{\lambda} \frac{Im\gamma}{\lambda(1+n_s)} + \left(\frac{2\pi|\gamma|}{\lambda(1+n_s)}\right)^2\right]^{-1} \tag{2}$$

where λ is the wavelength of the incident light, c the velocity of light and n_s the refractive index of the substrate. For the film under consideration²⁾ the islands are in reasonable approximation prolate spheroids with the long axis parallel to the surface of the substrate. The polarizability of such a spheroid is given by⁴⁾

$$\alpha^{(k)}(e, v) = v \left[(\epsilon - \epsilon_m)^{-1} + \frac{1}{\epsilon_m} L^{(k)}(e) \right]^{-1} \tag{3}$$

where V and e are the volume and the eccentricity of the spheroid. Furthermore, ϵ is the dielectric constant of the island material (gold in this case) and ϵ_m is the, as yet unspecified, dielectric constant of a medium in which the island is thought to be embedded, which satisfies $1 \leq \epsilon_m \leq \epsilon_s$; ϵ_s is the dielectric constant of the substrate (glass in this case). The depolarization factors in the direction of the long, $k=1$, and the short, $k=2$, axis parallel to the surface of the substrate are given by⁴⁾

$$L_{(1)}(e) = A(e) - B(e) \quad \text{and} \quad L_{(2)}(e) = \frac{1}{2} - \frac{1}{2} A(e) - B(e) \\ A(e) = \frac{1}{2} e^{-3} (1-e^2) \left[\ln \left(\frac{1+e}{1-e} \right) - 2e \right] \quad \text{and} \quad B(e) = \frac{1}{24} \left(\frac{\epsilon_s - 1}{\epsilon_s + 1} \right) (1-e^2)^{-\frac{1}{2}} \tag{4}$$

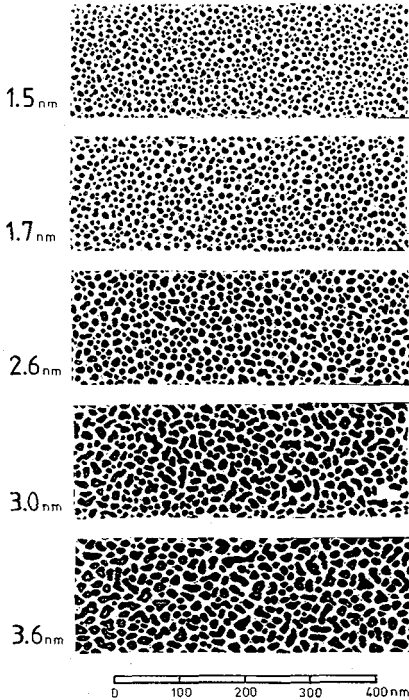


Fig.1- High-resolution electronmicrographs of regions of the films

B is due to the interaction with the image dipole and we note that the spheroid touches the substrate surface. For different islands e_i and V_i will be different. The average polarizability parallel to the surface α_{\parallel}^s is given by

$$\alpha_{\parallel}^s = \frac{1}{2} t_w \frac{\sum_i \left[\alpha^{(1)}(e_i, V_i) + \alpha^{(2)}(e_i, V_i) \right]}{\sum_i V_i} \quad (5)$$

The weight thickness t_w is in principle equal to $\sum_i V_i$ per unit of surface area. In practice one finds a somewhat lower value²⁾ due to so-called "lost weight". We used the experimental values $t = 1.5, 1.7, 2.6, 3.0$ and 3.6 nm in our calculation. Using a computer analysis⁵⁾ of the electron micrographs, fig.1, we obtained the average polarizabilities. In fig.2 these are given using $\epsilon_m = \frac{1}{2}(1 + \epsilon_s)$. The description using the dielectric susceptibility γ for the film^m gives the same transmittance as a plane-parallel plate if one uses

$$\frac{\epsilon_{eff} - \epsilon_m}{\epsilon_{eff} + 2\epsilon_m} = \frac{f}{3\epsilon_m} \left(\frac{\alpha_{\parallel}^s}{t_w} \right) \quad \text{with } f = \frac{t_w}{t_{opt}} \quad \text{and } t_{opt} = (\epsilon_m \kappa_{\parallel})^{-1} \quad (6)$$

for the dielectric constant, filling fraction and thickness of the plate. If one would use spherical islands and neglects image dipoles eqs.(2)-(6) give the original Maxwell Garnett formula¹⁾. This shows the fact that the optical thickness in the usual description may be calculated in terms of the local field factor κ_{\parallel} . This factor, and thus the optical thickness, is given in our theory in terms of the correlation function $g_w(r)$ for the distribution of island mass along the surface of the substrate by

$$\kappa_{\parallel} = \frac{1}{\epsilon_m t_{opt}} = \frac{3}{4} \int_0^{\infty} \frac{dr}{r^2} g_w(r) \left[1 + \left(\frac{1 - \epsilon_s}{1 + \epsilon_s} \right) \frac{r^3(r^2 - 8d^2)}{(r^2 + 4d^2)^{5/2}} \right] \quad (7)$$

Here d is the average distance of the centre of the islands to the substrate. The

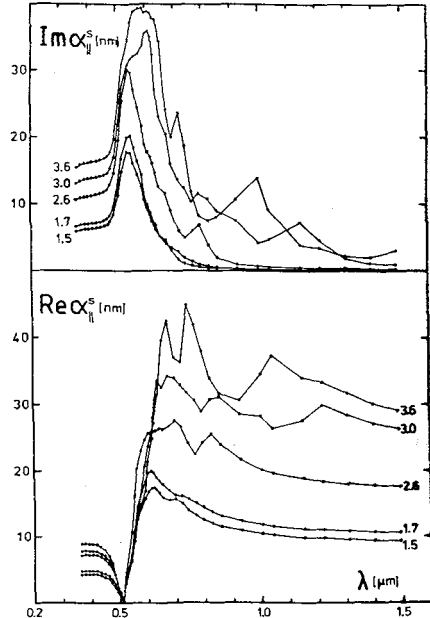


Fig.2- The polarizability density of the films using $\epsilon_m = \frac{1}{2}(1 + \epsilon_s)$

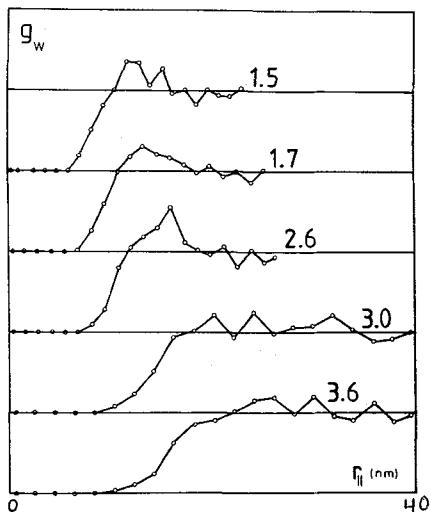
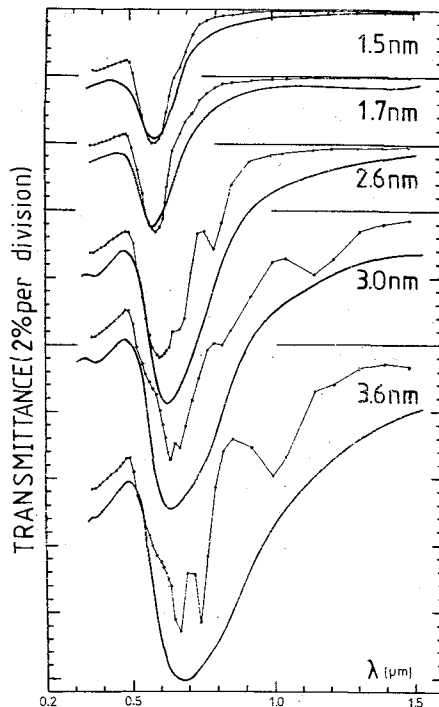


Fig.3- The mass distribution correlation functions of the films

Fig.4- The transmittance as a function of the wavelength for the films. The continuous curves give the experimental values. The dots give the computed values.



second term between the square brackets is due to the image dipoles. In fig.3 the correlation functions, obtained from the electronmicrographs, are given for the various films. The resulting transmittances are compared with the experimental values in fig.4, where we used $\epsilon_m = \frac{1}{2}(1+\epsilon_s)$. In order to gain some insight into how the various parameters like the eccentricity, ϵ_m , the optical thickness and the image charge affect the computed transmittance T , we have varied all these quantities. Fig.5 shows the values of T for 3 different choices of ϵ_m and it is apparent that both the location of the minimum and the amount of absorption depend strongly on ϵ_m . For all films we find that $\epsilon_m = \frac{1}{2}(1+\epsilon_s)$ is the best choice. A theoretical motivation for this choice may also be given^{1b)} based on the observation that ϵ_{eff} should approach ϵ in the $f=1$ limit. Fig.6 shows that accounting for the image dipoles increases the width of the minimum in T . This improves the agreement with the experimental shape. In fig.7 the value of T is compared with a value obtained by stretching all the islands. This shows that the location of the minimum is a sensitive function of the shape of the islands. Clearly the values of the eccentricities found from the electronmicrographs and used in the calculation lead to a satisfactory value of this location. In fig.8 the value of T is compared to a value obtained by decreasing t_{opt} . It is interesting to note that the decrease of t_{opt} has an effect comparable to an increase of ϵ_m . This explains why Norrman et al.²⁾ were able to fit their data using $\epsilon_m=1.2$,^m rather than the value $\epsilon_m = \frac{1}{2}(1+\epsilon_s) = 1.67$ we used, using t_{opt} as an adjustable parameter. It should be emphasized however, that the statistical analysis leads to a unique value of t_{opt} so that we obtain our values of T without fitting any parameter.

REFERENCES

- /1/ Vlieger, J. and Bedeaux, D., Thin Solid Films 69 (1980) 107
Bedeaux, D. and Vlieger, J., Thin Solid Films, 102 (1983) 265
- /2/ Norrman, S., Andersson, T., Granqvist, C.G. and Hunderi, O., Phys.Rev.B18 (1978)
- /3/ Bedeaux, D. and Vlieger, J., Physica 67 (1973) 55 /674
- /4/ Yamaguchi, T., Yoshida, S and Kinbara, A, Thin Solid Films 21 (1974)173
- /5/ Kwee, R.S.H., Kandidaatsverslag (T.H.Delft, the Netherlands, unpublished)

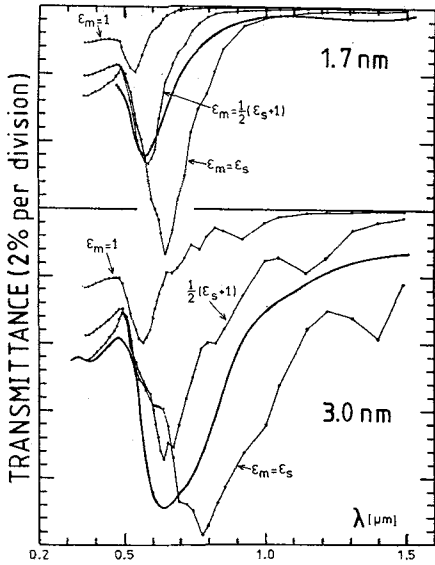


Fig.5 - The computed transmittance for $\epsilon_m = 1$, $\frac{1}{2}(\epsilon_s + 1)$ and ϵ_s for two films

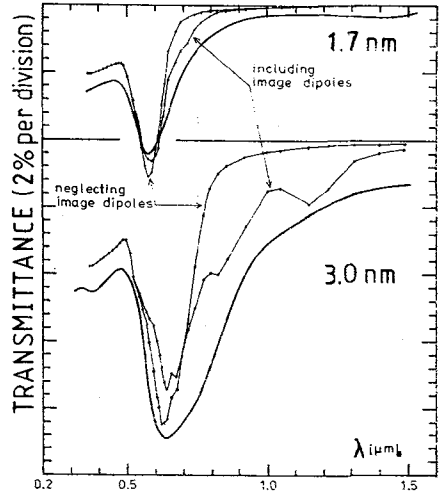


Fig.6 - The computed transmittance including and neglecting image dipoles for two films

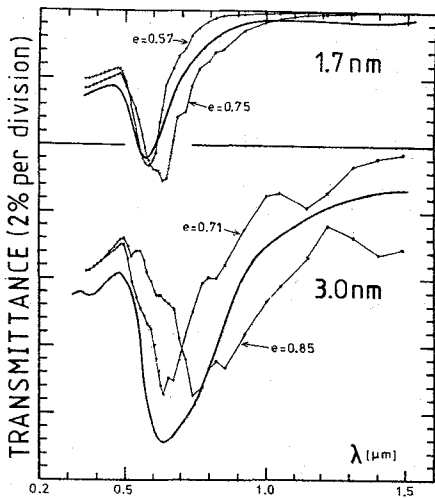


Fig.7 - The computed transmittance for two values of the average eccentricity for two films

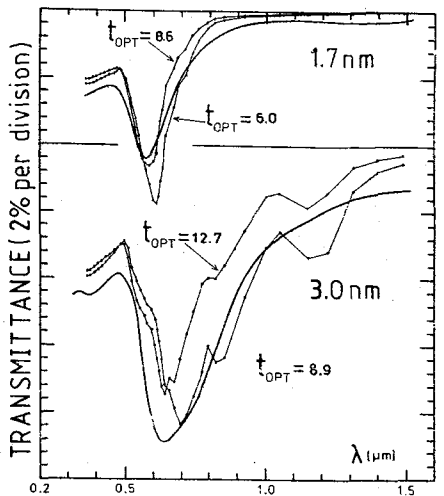


Fig.8 - The computed transmittance for two values of the optical thickness for two films