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Optical properties of aluminium island films on NaCl substrates near the percolation threshold*

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Optical properties of aluminium island films on NaCl substrates were investigated. Reflectivity was measured in the spectral range of 220 to 700 nm. The imaginary part of dielectic permittivity was calculated for films of a volume fraction $q < q_c$. A distinct change in the character of optical properties was found to occur at the volume fraction of $q_c \simeq 0.45$. Films having a volume fraction of $q < q_c$ exhibit an optical anomaly which takes the form of a maximum in the wavelength dependences of reflectivity and imaginary part of dielectic permittivity. This anomaly disappears for $q > q_c$ and the character of reflectivity is the same as that for thick films with diffuse scattering.

1. Introduction

Experiments on the optical properties of cermets and metal island films prepared on dielectric substrates have been reported a number of times in the past decades [1–5]. The optical anomalies which are found to occur take the form of maxima appearing in the imaginary part of dielectric permittivity ε_2' or in the reflectance R of the film as a function of wavelength.

Many classical models are available to describe the optical properties of dispersion systems. In these models, metal islands deposited on dielectric substrates influenced by an external electromagnetic field are assumed to form a system of interacting dipoles. The theoretical approach by Maxwell–Garnett [6] was first to describe the problem in question. This theory has been modified a number of times, e.g., by David [7], Doremus [8], Jarret and Ward [9], Yamaguchi [10], Norrman et al. [11].

The optical properties of metallic island films may also be interpreted in terms of the Bruggeman theory of the effective medium [12].

Having at hand a wide spectrum of theoretical approaches, we can determine the influence of wavelength λ on the imaginary part of dielectric permittivity ε_2' on reflectivity R or on the transmittance T of island films characterized by a volume fraction q [13]. When the films are sufficiently thin ($d \ll \lambda$), the imaginary part of

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dielectric permittivity is established by experiments using Wolter's approximation [14]

$$\varepsilon_2' = \frac{\lambda}{2\pi d} n_s \frac{1-R-T}{T} \quad (1)$$

where: n_s – refraction coefficient of substrate,
d – films thickness.

The agreement between experimental and theoretical wavelength-dependence of ε_2', R, and T for metallic island films was found to be good [11], [15], [16]. The reflectivity of smooth continuous films displays a monotonous plot in the wavelength range with no interband transition or plasma excitation [17]. When a continuous film exhibits a rough surface, reflectivity may be defined as [18], [19]:

$$R = R_0 \exp\left[-\left(\frac{4\pi\sigma}{\lambda}\right)^2\right] \quad (2)$$

where R_0 is reflectivity of a perfectly smooth conducting surface and σ denotes mean square deviation of the surface from the mean surface level. Equation (2) holds when $\sigma \ll \lambda$. When the film shows only partial coverage, reflectivity may be calculated as follows:

$$R = R_0 q_s \exp\left[-\left(\frac{4\pi\sigma}{\lambda}\right)^2\right] + (1-q_s)R_s \quad (3)$$

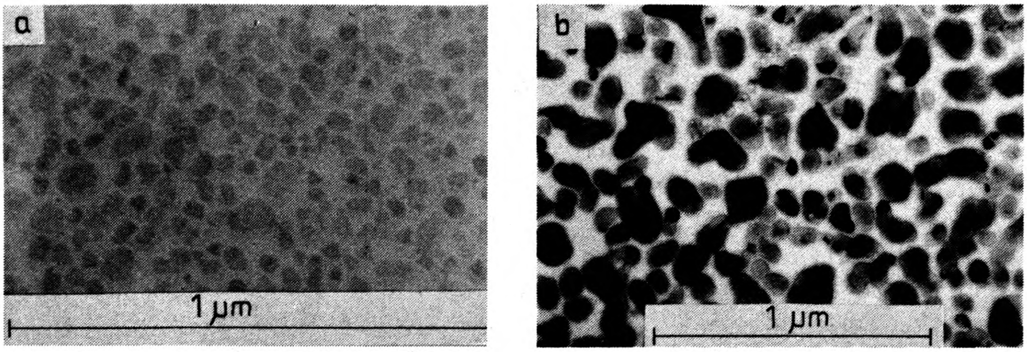
where R_s denotes reflectivity of the substrate, and q_s is coefficient of coverage; the relation between coverage coefficient and volume fraction takes the form [13]

$$q_s = \frac{3}{2}q. \quad (4)$$

The volume fraction which characterizes the percolation threshold (q_c – critical volume fraction) occurs between two q-value ranges — that with optical anomalies and that with monotonous R, T, ε_2' plots. In this study, the optical properties of aluminium island films were investigated in the range of optical anomalies ($q < q_c$), in the range of monotonous plots ($q > q_c$), and on the percolation threshold ($q \approx q_c$).

2. Experimental

Aluminium island films of volume fractions varying from 0.3 to 0.66 were deposited by thermal evaporation. The method of depositing aluminium island films onto NaCl substrates, as well as that of examining their structure, thickness and optical properties were described in papers [15], [20]. The microstructures of films with different volume fractions are shown in Fig. 1**a**, **b**, **c**. Thus, when the volume fraction increases, some changes occur in the shape of the islands and in the type of the microstructure. A spectrophotometer UV VIS (Carl Zeiss Jena) was used for



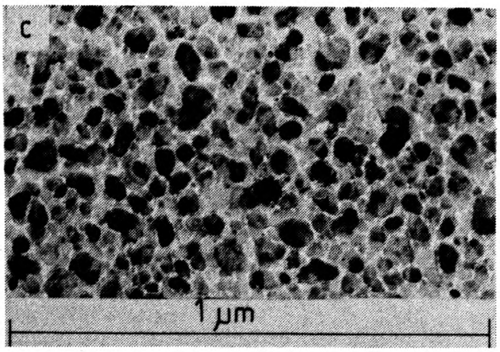


Fig. 1. Microstructures of Al films with different volume fractions: **a** — $q = 0.31$, **b** — $q = 0.43$, **c** — $q = 0.56$

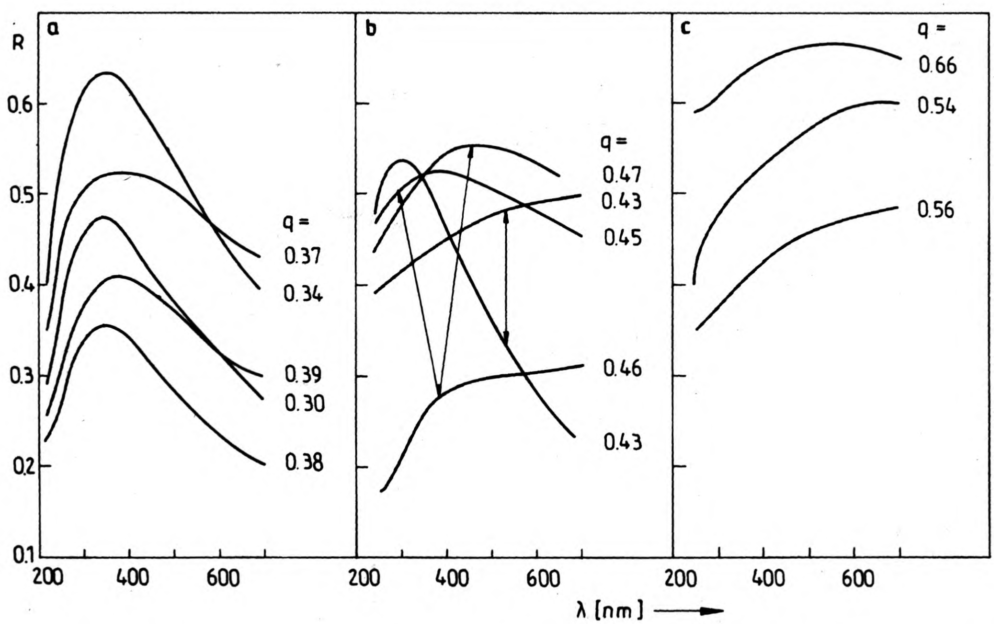


Fig. 2. Reflection ceofficient as a function of wavelength λ for Al films with different volume fractions: **a** — $q < q_c$, **b** — $q \simeq q_c$, **c** — $q > q_c$

measuring the energetic coefficients of reflection in the wavelength of 250 to 700 nm. The results are given in Fig. 2**a**, **b**, **c**. As shown by these data, volume fraction has an evident influence on the optical properties of the film. When q ranges from 0.30 to 0.42, the reflectivity curves are of a resonant nature, a distinct maximum being observed in the range $\lambda = 350$ to 400 nm (Fig. 2**a**). This relation is not so clear when the volume fraction range is $0.43 \leqslant q \leqslant 0.47$. Films of identical volume fraction, e.g., 0.43, may either display optical anomalies or not. Films of volume fractions 0.45 and 0.46 or 0.47 bahave in a similar manner (Fig. 2**b**). A further increase in the volume fraction value ($q > 0.54$) brings about a change in the nature of optical properties. No maxima are found to occur, and reflectivity decreases towards shorter wavelengths. This is a typical plot of rough films creating favourable conditions for the scattering of radiation (Fig. 2**c**).

3. Discussion of results

Making use of Eq. (1), the wavelength-dependence of ε_2' was plotted for ($q < q_c$) films. The results are given in Fig. 3. The curves display distinct maxima which have a

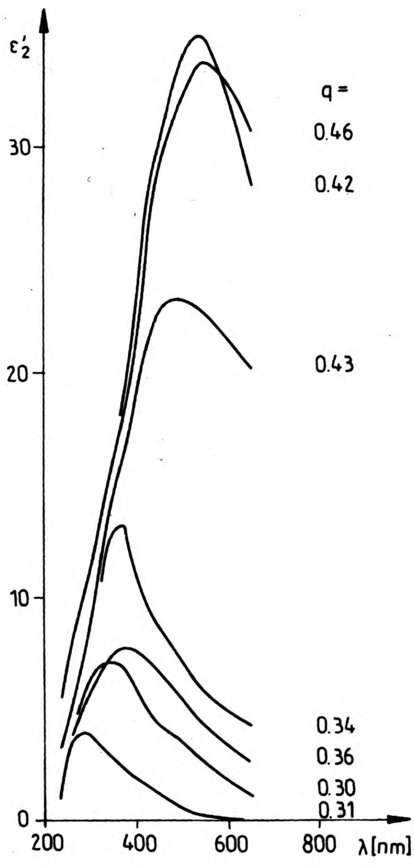


Fig. 3. Imaginary part of the permittivity ε_2' as a function of wavelength λ for Al films with different volume fractions

tendency to shift towards longer wavelengths with the increasing volume fraction [15]. On the basis of the available theories, the curves may be regarded as typical of isolated islands which, under the influence of an electromagnetic wave, behave in a manner similar to that of dipole systems [6]–[11].

A further increase of the volume fraction yields films of a specific feature – in spite of an identical q-value, optical anomalies are either present or absent. This behaviour may be explained as follows: there is a certain probability that, if q approaches q_c, the films will either consist of isolated islands or will display islands connected to one another with conducting pathways. Optical anomalies appear in the isolated island system and disappear when the islands are connected. Taking into account the substantial change in the nature of optical properties, it seemed advisable to determine the range of the critical volume fraction values q_c which characterize the percolation threshold. For aluminium island films on NaCl substrates the threshold volume fraction interval becomes $0.43 \leqslant q_c \leqslant 0.47$. Similar threshold values were obtained by electric measurements conducted simultaneously with optical examinations [21].

Using Equation (3), the wavelength-dependence of reflectivity was calculated for the experimental volume fractions 0.43, 0.54, and 0.66, adopting σ-values of 0, 5, 10 and 15 nm, respectively. The results are plotted in Fig. 4**a**, **b**, **c**. From these curves it can be seen that as the volume fraction increases, so does reflectivity (for determined

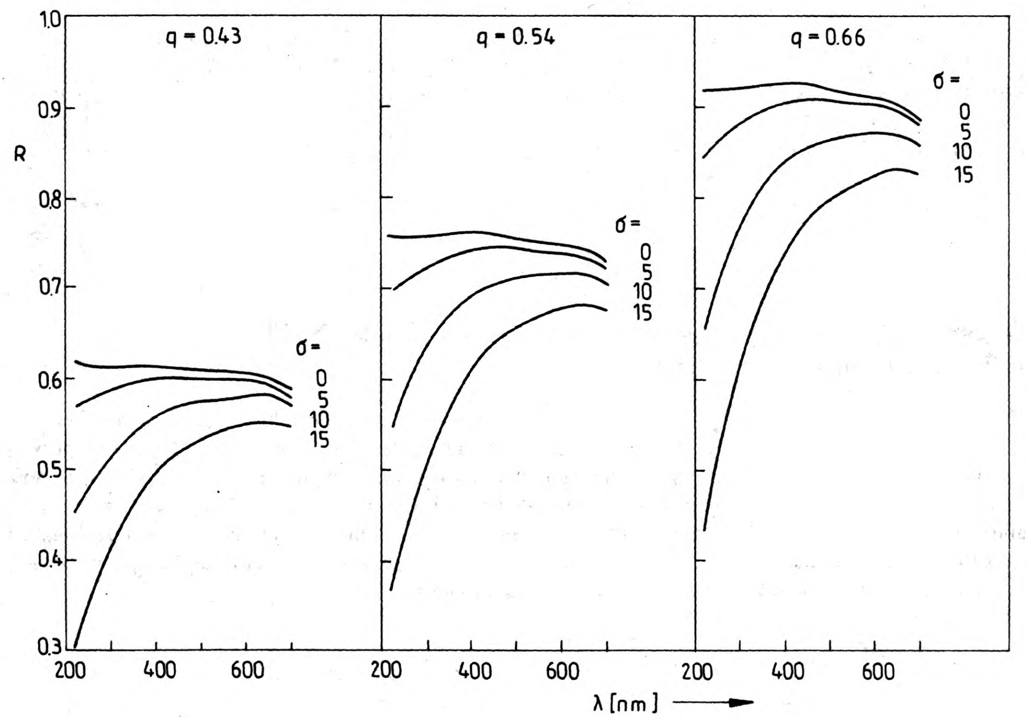


Fig. 4. Calculated value of R as a function of wavelength λ for Al films with different volume fractions: **a** – $q = 0.43$, **b** – $q = 0.54$, **c** – $q = 0.66$

σ- and λ-values). When the σ-value increases, reflectivity decreases evidently, especially in the shortwave range of the spectrum. Films of the volume fraction ($q > q_c$), Fig. 2c, follow a similar behavioral pattern. It may be agreed that the islands of such films are interconnected (anomalies are absent) and that scattering on the surface roughness is a dominant effect.

References

[1] SHKLYAREVSKII I. H., BLYASHENKO I. H., Opt. Spektrosk. **44** (1978), 545–549 (in Russian).
[2] AL-ABDELLA P. B., SHKLYAREVSKII I. H., Zh. Prikl. Spektrosk. **33** (1980), 712–717 (in Russian).
[3] VO-VAN TRUONG, GIROUARD E., BASI G., Appl. Opt. **21** (1982), 2508.
[4] BERTHIER S., LAFAIT J., J. Phys. **47** (1986), 249.
[5] NOH T. W., SONG Y., LEE S., GAINES J. R., PARK H. D., Phys. Rev. B **33** (1983), 3793.
[6] MAXWELL-GARNETT J. C., Philos. Trans. R. Soc. **203** (1904), 385. Ibidem **205** (1906), 237.
[7] DAVID E., Z. Phys. **114** (1939), 389.
[8] DOREMUS R. H., J. Appl. Phys. **37** (1966), 2775.
[9] JARRETT D. N., WARD L., J. Phys. D **9** (1976), 1515.
[10] YAMAGUCHI S., J. Phys. Soc. Jpn. **17** (1962), 1172.
[11] NORRMAN S., ANDERSSON T., GRANQVIST C. G., HUNDERY O., Phys. Rev. B **18** (1978), 674.
[12] BRUGGEMAN D. A. G., Ann. Phys. (Leipzig) **24** (1935), 636.
[13] MCKENNA W. T., WARD L., Phys. Status Solidi A **68** (1981), K11.
[14] WOLTER H., Z. Phys. **105** (1937), 269. Ibidem **113** (1939), 547.
[15] DOBIERZEWSKA-MOZRZYMAS E., RADOSZ A., BIEGAŃSKI P., Appl. Opt. **24** (1985), 727.
[16] GRANQVIST C. G., HUNDERY O., Phys. Rev. B **16** (1977), 3513.
[17] WOOTEN F., *Optical Properties of Solids*, Academic Press, New York, London 1972.
[18] BENNETT H. E., PORTEUS J. O., J. Opt. Soc. Am. **51** (1961), 123.
[19] DAUDE A., SAVARY A., ROBIN S., Thin Solid Films **13** (1972), 255.
[20] DOBIERZEWSKA-MOZRZYMAS E., BIEGAŃSKI P., RADOSZ A., Thin Solid Films **102** (1983), 77.
[21] DOBIERZEWSKA-MOZRZYMAS E., BIEGAŃSKI P., to be published.

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Оптические свойства островных слоев Al на основе NaCl в близости порога перколяции

Исследованы оптические свойстве островных слоев Al на основании NaCl. В спектральном пределе от 220 до 700 нм определены зависимости коэффициента отражения и мнимой части электрической проницаемости от длины волн для слоев с коэффициентом заполнения $\Delta q_c = 0{,}43$–$0{,}47$, в котором происходит изменение характера оптических свойств. Для $q < \Delta q_c$ слои проявляют оптическую аномалию заключающуюся в присутствии максимума коэффициента отражения и мнимой части электрической проницаемости. Когда $q > \Delta q_c$ оптическая аномалия исчезает, характер отражения становится типичным как для слоев рассеивающих свет.