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# Surface-roughness influence on the optical properties of aluminium films**

The relation between surface-roughness and optical properties of Al-fim was studied in the range from 210 to 600 nm. Al-film were evaporated on heated quartz substrates in a perpendicular electric field, and without electric field. It has been found that in the short-wavelength range of the spectral reflection decreases with increasing roughness. Experimental and theoretical curves were compared, the latter being calculated in terms of Davies' theory.

## 1. Introduction

Films deposited in the evaporation process have a grained structure and, usually, rough surfaces. Optical properties, and especially the coefficient of reflection at normal incidence, are considerably sensitive to surface roughness [1, 2]. Studies of the relation between surface irregularities and optical properties were performed both theoretically [3, 4] and experimentally [5, 6].

When irregularities appear at the surface of the film, light scattering takes place; thus, the coefficient of reflection at normal incidence for a rough surface is always lower than the same coefficient for a smooth surface. In the theory explaining this phenomenon and was developed by Davies [3], the following assumption have been taken: 1) the root mean square roughness $\sigma$ is small compared to the wavelength $\lambda$; 2) the surface is perfectly conducting, and the specular reflection coefficient of the smooth surface is close to unity; 3) the distribution of heights of the surface irregularities is Gaussian about the mean.

Under these assumptions Davies has derived the following formula for the coefficient of specular reflection at normal incidence to the rough surface

$$R_1 = R_0 \exp\left[-\left(\frac{4\pi\sigma}{\lambda}\right)^2\right], \tag{1}$$

where $R_0$ is the coefficient of reflection for a perfectly smooth metallic film. As can be seen from eq. (1), the decrease of reflection due to light scattering is especially pronounced in the short wavelength; for sufficiently long wavelengths this decrease is negligible. Eq. (1) can also be written as follows:

$$R_1 = R_0 - \Delta R_1, \tag{2}$$

where

$$\Delta R_1 = R_0\left\{1 - \exp\left[-\left(\frac{4\pi\sigma}{\lambda}\right)^2\right]\right\}$$

is a correction resulting from light scattering. Surface roughness also leads to the excitation of surface plasmons. This problem has been theoretically treated in [4, 7, 8], where the influence of surface-plasmon excitation on the specular coefficient of reflection has been determined. The decrease of the coefficient of reflection $R_2$ at normal incidence associated with this effect becomes

$$R_2 = R_1 - \Delta R_2, \tag{3}$$

where $\Delta R_2$ depends on the wave frequency ($\omega$), ($\omega_p$ is plasmon frequency root mean square roughness $\sigma$), the autocorrelation length assuming a Gaussian hill distribution (a) [6].

To compare the theoretical relations with the experimental results, films of given roughnesses have been evaporated by the following methods:

1. A rough dielectric film is evaporated onto a substrate, and then a conducting film is superimposed to the latter reproduce the irregularities of the dielectric film [6, 9].
2. In a slow evaporation process the metal film is deposited on a substrate heated up to centigrades [10, 11, 12].
3. A metal film is evaporated on a rough substrate with given irregularities [5].

The optical properties of the films prepared (e.g. of Al-films) were studied in the vacuum UV and in the IR [5, 6]. It has been found that in vacuum UV the theoretically predicted reflection minimum appears at $\lambda \sim 1300$ Å, and is associated with the excitation

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of surface plasmons [6]. A similar minimum appears for silver films at $\lambda \sim 3200$ Å [13]. It is verified by experiments that the reflection for rough surfaces obtained by using methods 1), 2), and 3) is smaller, especially at the short-wavelength, than for smooth films in [9, 14].

In the present paper the optical properties of Al-films evaporated by the method 2) are related to the surface roughness. The assumed surface roughnesses were: $\sigma \sim 10$ nm and $\sigma \sim 20$ nm. Al-films of the roughness $\sigma \sim 10$ nm were deposited on heated quartz substrates at high evaporation rates, while those with $\sigma \sim 20$ nm were obtained on the same substrates but in a perpendicular electric field and at low evaporation rates.

## 2. Experimental

Al-films were prepared by the methods reported earlier [15, 16]. Some films were deposited in the evaporation process involving a perpendicular electric field [17]. During one evaporation process three Al-films were obtained, of which two were prepared on a NaCl substrate, and one on an amorphous quartz substrate. The films deposited onto a quartz substrate were examined by electron microscopy with the application of carbon replica and by X-ray methods. Fig. 1 (a, b, c) represents the electron micrographs of Al-films evaporated under different conditions. As can be seen in fig. 1, these are films with rough surfaces. It is to be noted that the greatest irregularities appear when the evaporation process involves a perpendicular electric field of 80–100 V/cm and is performed at low rates.

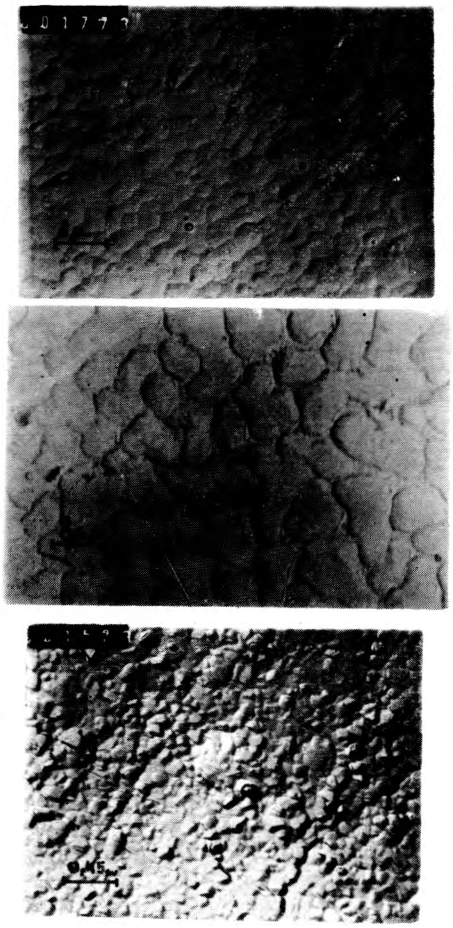

Fig. 1. An electron micrograph of an Al-film deposited on a quartz substrate:
a)*) $t = 115$ nm, $T_{\text{substrate}} = 340°\text{C}$, $v = 40$ nm/s, b) $t = 195$ nm, $T_{\text{substrate}} = 350°\text{C}$, $v = 13$ nm/s, $\varepsilon = 80$ V/cm, c)*) $t = 360$ nm, $T_{\text{substrate}} = 400°\text{C}$, $v = 4.5$ nm/s, $\varepsilon = 105$ V/cm

Structural investigations have been revealed that the films prepared on quartz substrates show only partial ordering of the crystalline structure. The presence of the (111) orientation was established for the maximum sensitivity of the X-ray detector (fig. 2) [15, 16].

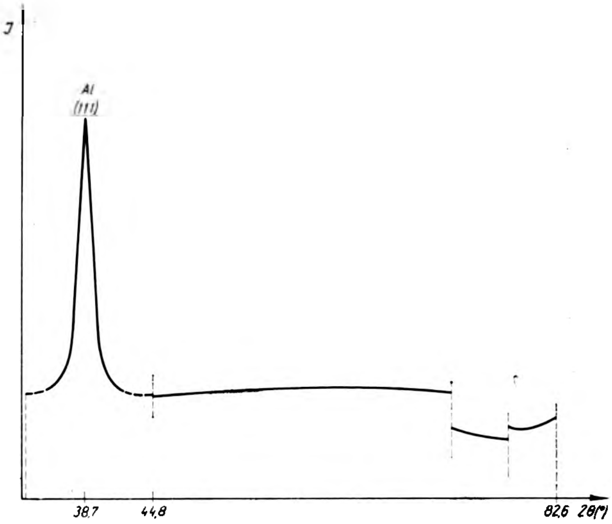

Fig. 2. X-ray intensity distribution versus diffraction angle for Al-film deposited on a quartz substrate

The coefficients of specular reflection ($R$) at normal incidence were measured in the range from 210 to 600 nm, using Zeiss Specord UV VIS equipped with a reflection unit. The film thickness ($d$) determined by multiple-beam interference method, varies from 100 to 360 nm.

## 3. Discussion of results

The coefficients of reflection are plotted in figs 3 and 4. It is easily seen that for films prepared at a high evaporation rate without the electric field the dependence of $R$ on $\lambda$ (fig. 3) differs from that for the films deposited at low evaporation rates with the application of a perpendicular electric field (fig. 4). In

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* The photograph has been made at the Laboratory of Electron Microscopy, Wrocław Technical University.

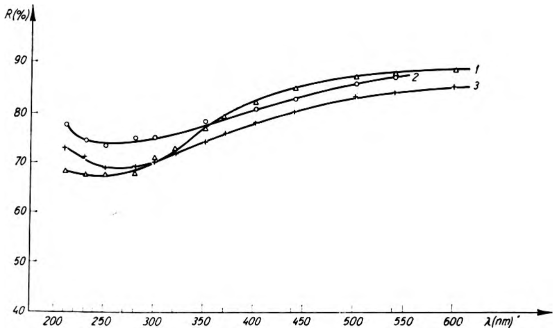

Fig. 3. The experimental curves of reflection as a function of wavelength for Al-films deposited without electric field

Curve *1*: $t = 150$ nm, $T_{\text{substrate}} = 350°C$, $v \approx 75$ nm/s; curve *2*: $t = 121$ nm, $T_{\text{substrate}} = 340$ °C, $v = 60.5$ nm/s; curve *3*: $t = 115$ nm, $T_{\text{substrate}} = 340°C$, $v = 40$ nm/s

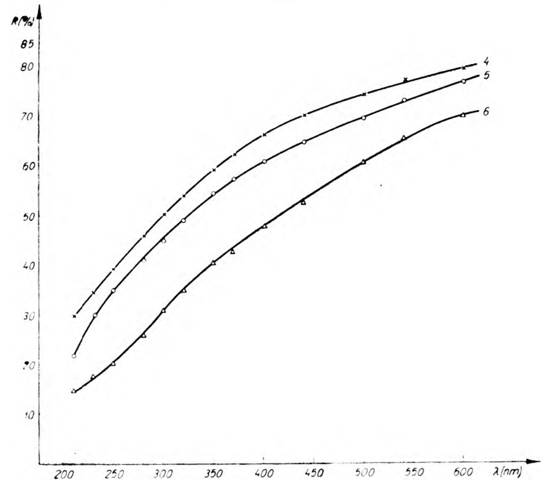

Fig. 4. The experimental curves of reflection as a function of wavelengths for Al-films deposited in an electric field

Curve *4*: $t = 360$ nm, $T_{\text{substrate}} = 400°C$, $v = 10.3$ nm/s, $\varepsilon = 105$ V/cm; curve *5*: $t = 195$ nm, $T_{\text{substrate}} = 350°C$, $v = 13$ nm/s, $\varepsilon = 80$ V/cm; curve *6*: $t = 225$ nm, $T_{\text{substrate}} = 320°C$, $v = 4.2$ nm/s $\varepsilon = 80$ V/cm

the first case the coefficient of reflection decreases with decreasing wavelength, reaches an insignificant minimum at $\lambda = 270$ nm, and slightly increases toward the UV region.

The shapes of the $R(\lambda)$ curves represented in fig. 3 seem to be caused by two effects; at long wavelengths they are due to light scattering at the rough surface, and below $\lambda = 300$ nm ($\varepsilon \sim 4.2$ eV) to the absorption related to the transition of electrons from the surface states of the (111) plane to the Fermi level.

The dependence of $R$ on $\lambda$ determined theoretically from eq. (1) for $\sigma \sim 10$ nm, are shown in fig. 5 together with the experimental curves. From a comparison it follows that the experimental results are in good agreement with the theoretical predictions for a wide 300 to 600 nm range of wavelengths. This may be attributed to the effect of light scattering described by the correction of $\Delta R_1$ (eq. 2).

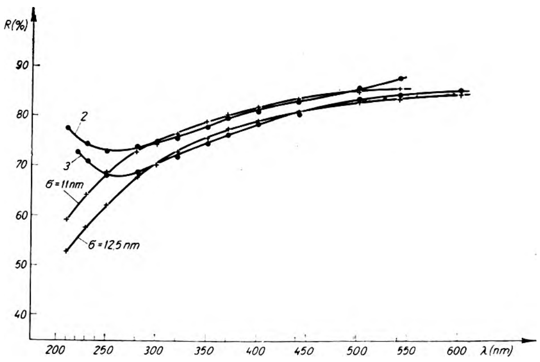

Fig. 5. The theoretical (+) and experimental (.) curves of reflection as a function of wavelength for Al-films deposited without an electric field

For wavelength below 300 nm the theoretical and experimental plots become inconsistent. An absorption band found for these wavelengths in the case of epitaxial films may correspond to the transitions from the surface states [18, 19]. X-ray examinations have shown that the films of interest display only a partial ordering of the crystalline structure and a (111) orientation (fig. 2).

Fig. 4 represents the dependence of $R$ on $\lambda$ for Al-films prepared at low evaporation rates with the use of a perpendicular electric field. The coefficients of reflection are evidently decreasing towards short wavelengths. From the eq. (1) it follows that the surface irregularities are large. Assuming that $\sigma$ ranges from 18 to 25 nm the theoretical curves of the $R$ on $\lambda$ dependence have been calculated from eq. (1) and shown in fig. 6. In the spectral range under consideration the agreement between the theoretical and experimental results is good. It is supposed that for

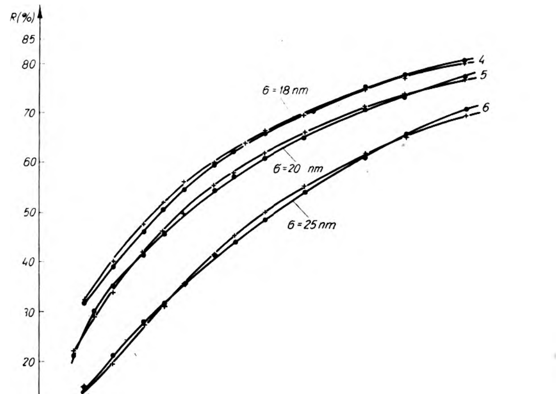

Fig. 6. The theoretical (+) and experimental (.) curves of reflection as a function of wavelength for Al-films deposited in an electric field

the roughness exceeding $\sigma \sim 20$ nm, the light scattering effect at the irregular surface may be of considerable importance.

## 4. Conclusions

Al-film evaporated onto a heated amorphous substrate display a partial ordering of the crystalline structure and a rough surface. When the evaporation process has involved a perpendicular electric field and a low evaporation rate, then surface roughness increases. For such films the coefficient of reflection decreases toward UV. The experimental dependence of $R$ on $\lambda$ is in good agreement with the theoretical curves and justifies the dominating role of the effect of light scattering. When $\sigma$ takes smaller values ($\sigma \sim 10$ nm), it is probable that in addition to light scattering we should take account of the absorption phenomenon associated with the transition of electrons from the surface states of the (111) plane and corresponding to the wave energy $\varepsilon \sim 4.2$ eV ($\lambda \sim 300$ nm).

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### Влияние шероховатости поверхности слоев Al на их оптические свойства

Исследовано влияние шероховатости слоев Al на их оптические свойства в пределах 210–600 нм. Слои осаждались из газовой фазы на нагреваемое кварцевое основание в перпендикулярном электрическом поле и без поля. Обнаружено, что в коротковолновой части спектра отражение уменьшается с возрастанием неровностей. Опытные кривые сопоставлены с теоретическими, вычисленными на основе теории Девиса.

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