

## Optical properties of epitaxial aluminium films\*\*

Epitaxial Al films (111) orientation were evaporated onto a NaCl substrate and their optical properties were studied. The coefficients of reflection ( $R$ ) and transmission ( $T$ ) at normal incidence were measured both in the UV and visible regions. The optical constants  $n$  and  $k$ , determined by numerical method, were found to be lower than those of Al films which have been evaporated on a glass substrate held at room temperature. The imaginary part of the dielectric constant ( $\epsilon_2$ ) and optical conductivity ( $\sigma$ ) were considered as a function of wave energy. For thick films ( $d \sim 200$  nm) the  $\epsilon_2$  and  $\sigma$  plots are monotonic, and in the case of thinner films ( $d \lesssim 100$  nm) there appears a pronounced peak which may be attributed to the theoretically expected transition corresponding to the surface states.

### 1. Introduction

The optical properties of polycrystalline aluminium films were studied over a wide spectral range [1–3]. The optical properties, as well as the real and imaginary part of the dielectric constant, have been determined in the range from UV to far IR [4, 5]. On the other hand, the band structure of Al is known and the interband transitions appearing in IR near 1.5 eV and 0.5 eV are theoretically determined [6, 7]. Hence, in the case of Al films we are able to compare theoretical and experimental results. A review of the so far obtained theoretical and experimental results is reported in [8] and [9]. The theoretically determined interband transition near 1.5 eV corresponding to the Fourier coefficient  $V_{200}$  of the pseudopotential has been justified by experiments. In the near UV and visible range the optical properties of thick polycrystalline Al films are studied in terms of Drude's free electron theory [1]. Studies are also reported for the influence of the substrate roughness on the optical properties of Al films. For this purpose a dielectric film of a coarse-grained and porous structure was deposited onto a glass plate and then covered with aluminium. The imaginary part of the dielectric constant in these films, determined for 1–2 eV, was found to be significantly smaller than that of the Al films directly evaporated on a glass substrate; nevertheless, the position of the peak corresponding to 1.5 eV remained unchanged [10]. It was also found that owing to light scattering the coefficient of reflection for coarse-grained films was low [11].

In the literature discussed, the optical properties of Al films are considered from the viewpoint of the action of electromagnetic wave with free electrons and from the viewpoint of interband transitions.

The authors of [12] suggest that the optical properties of Al films may be influenced by surface states. Based on theoretical studies, two optical transitions are expected. The first transition is produced by the surface states of the (100) plane and corresponds to the energy 1.5 eV [13], while the second transition is due to the surface states of the (111) plane and falls to the wave energy range of 3.3–6.6 eV [14]. The peak at 1.5 eV which also corresponds to the interband transition was proved experimentally. However, optical studies in UV for Al films did not justify the existence of the second transition.

In the present paper the aim was to study the optical properties of monocrystalline Al films of (111) orientation. It has been found that there exist differences in the optical properties of monocrystalline and polycrystalline films especially for lower thickness ranges.

### 2. Experimental

Monocrystalline Al films were prepared using the method reported in [15] and [16]. Three film samples were made during one preparation process. One sample, evaporated on a NaCl substrate, was used for electronographic investigations. Another sample, also prepared on a NaCl substrate, was employed for electrical and optical measurements. The third one was evaporated onto a quartz substrate and used in X-ray examinations, as well as electrical, optical and thickness measurements. Both the samples prepared on a NaCl substrate were examined by transmission electron microscopy, while the sample evaporated on a quartz substrate was studied by means of carbon

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replica and X-ray methods. Figs 1\* and 2 represent the electron micrographs of Al films evaporated on NaCl and quartz substrates, respectively.

Structural studies have revealed that Al films which were evaporated on NaCl at a suitable temperature of the substrate and an appropriate evaporation rate are monocrystalline. The presence of the (111) orientation for films on quartz substrates was established for the maximum sensitivity of the detector. It seems reasonable to suppose that Al films deposited onto a quartz substrate are only partially ordered [16].

Optical properties of Al films prepared on both the substrates were measured in the near UV and visible range. The coefficients of reflection ( $R$ ) and transmission ( $T$ ) were determined in the range from 210 to 600 nm, using Zeiss Specord UV VIS. Measurements of reflection related to the specimen for normal incidence were performed on a reflection unit which has been designed for the spectrophotometer used in our studies. The coefficients of transmission for films characterized by a low transmittance were measured in a spectrophotometer VSU 2-P with various measuring ranges. Film thickness ( $d$ ), determined using

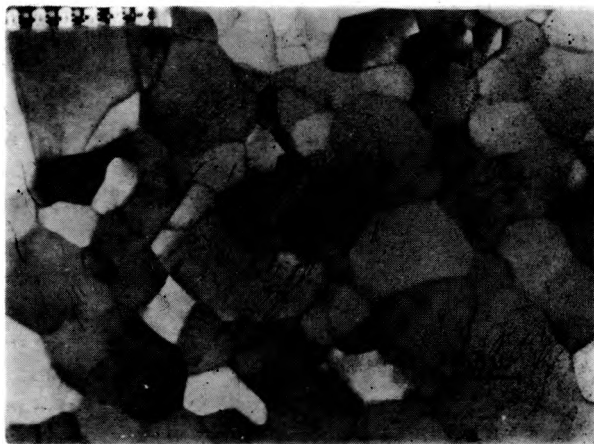


Fig. 1. Electron micrographs of Al film evaporated onto NaCl substrate

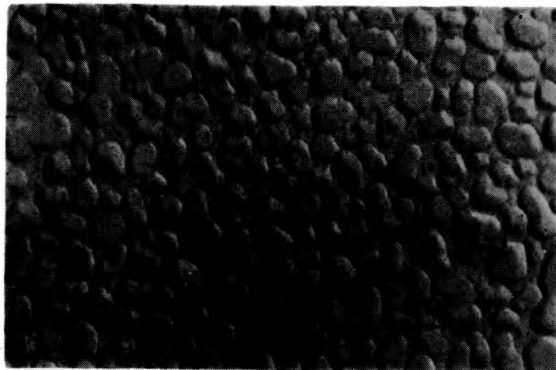


Fig. 2. Electron micrograph of Al film evaporated onto quartz substrate

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

the method of multiple beam interference, varies from 50 to 240 nm. Based on the measured coefficients of reflection and transmission, as well as on the determined film thickness the optical constants, refraction index ( $n$ ) and absorption coefficient ( $k$ ) have been calculated in the above range of wave energy, by using the method described in [18]. The imaginary part of the dielectric constant ( $\epsilon_2 = 2nk$ ) and the optical conductivity

$$\sigma = \frac{c}{2\lambda} \epsilon_2$$

as a functions of wave energy have also been determined.

### 3. Discussion of results

In fig. 3, the coefficient of reflection is plotted against wave energy for monocrystalline Al films. The same figure represents the reflection of thick polycrystalline films [2]. The dependence of  $R$  on  $E$  for monocrystalline films is different from that for polycrystalline films. In the case of monocrystalline films the coefficient of reflection behaves as follows: it remains almost constant in the visible region, shows the minimum in UV, evidently increases toward higher energies, and within 5.4–5.9 eV it reaches higher values than in the case of polycrystalline films. Hence it seems reasonable to employ this phenomenon in practice as monocrystalline Al films can be used in UV as mirrors of a high coefficient of reflection.

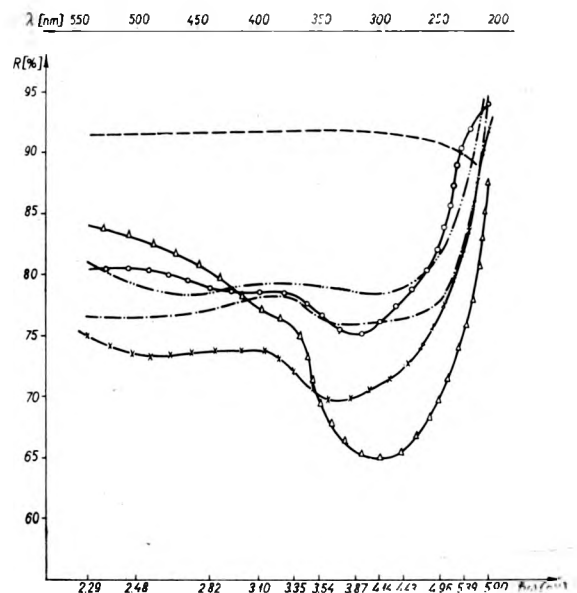


Fig. 3. Coefficient of reflection vs. wave energy for monocrystalline Al films

-----	Al sample
-----	239 nm
-x-x-x-x-x	198 nm
.....	195 nm
-o-o-o-o-o	115 nm
-Δ-Δ-Δ-Δ	56 nm

It was already mentioned in section 2 that the investigated Al film samples were deposited on to NaCl and quartz substrates under the same evaporation conditions. The experiments have shown that these films displayed various ordering of the crystalline structure [17] and different surface states (figs 1 and 2). Fig. 4 represents the coefficient of

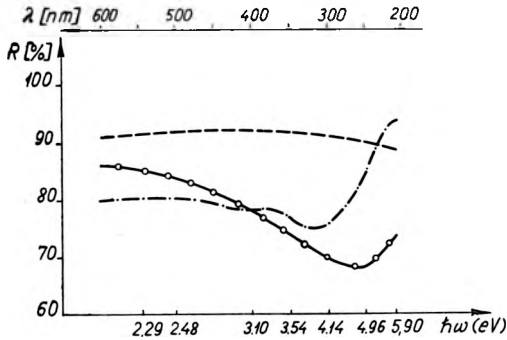


Fig. 4. Coefficient of reflection vs. wave energy both for monocrystalline and slightly textured Al films

- Al/quartz
- - - - - Al/NaCl
- · - · - · - · Al sample

reflection as a function of wave energy both for monocrystalline Al films and Al films prepared on a quartz substrate. In the UV region the optical properties of both the films show considerable differences. The coefficient of reflection of monocrystalline films evidently increases toward the UV region, while that of the Al film on a quartz substrate decreases and reaches an insignificant minimum at 4.9 eV. This character of the optical properties was observed in any experiment performed during our studies. The optical properties of monocrystalline films are such that the coefficient of reflection always increases in the direction of UV. The decrease of this coefficient in Al films on quartz substrate may be due to light scattering [19].

As can be seen in fig. 2, the surface of this film is

grained and rough. The dependence of the coefficient of reflection on the surface roughness is the subject of another study (in preparation).

Based on the measurements of reflection coefficients, transmission and thickness, the optical constants (*n*) and (*k*) were calculated. The coefficients of reflection and optical constants for monocrystalline Al films of various thicknesses are listed in table 1.

The refractive index and coefficient of absorption for thicker films increase toward IR. The increase is attributable to the interband transition at 1.5 eV, for which the optical constants reach their maximum values. Increasing film thickness contributes to the decrease of the optical constants.

The imaginary part of the dielectric constant and the optical conductivity were also determined. Both the magnitudes are plotted against wave energy in figs. 5 and 6, respectively. The shape of the  $\epsilon_2$  and  $\sigma$

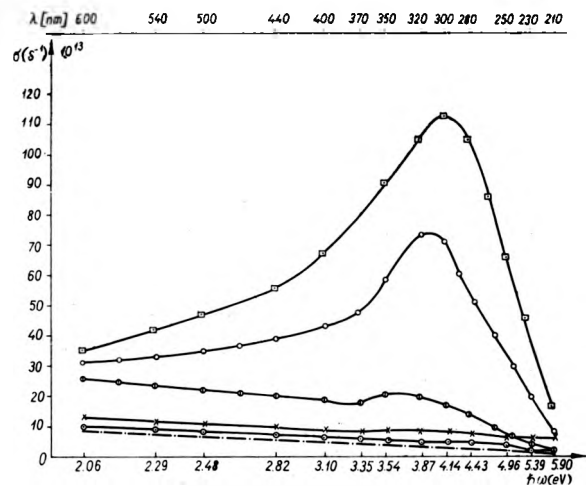


Fig. 5. Imaginary part of dielectric constant vs. wave energy for Al films of various thickness

- 56 nm
- 75 nm
- ◇-◇-◇-◇-◇ 115 nm
- ×-×-×-×-× 180 nm
- △-△-△-△-△ 195 nm
- +-+-+-+ 239 nm

Table 1

λ nm	d = 56 nm			d = 115 nm			d = 195 nm			d = 239 nm		
	R	n	k	R	n	k	R	n	k	R	n	k
210	0.878	0.076	1.405	0.940	0.021	0.788	0.945	0.015	0.421	0.936	0.017	0.312
230	0.767	0.207	1.534	0.914	0.034	0.861	0.864	0.043	0.448	0.850	0.046	0.326
250	0.697	0.330	1.654	0.843	0.075	0.922	0.810	0.065	0.489	0.800	0.065	0.357
280	0.650	0.512	1.906	0.780	0.125	1.033	0.790	0.077	0.560	0.766	0.080	0.411
300	0.651	0.559	2.015	0.760	0.152	1.123	0.784	0.085	0.640	0.761	0.085	0.483
320	0.651	0.594	2.093	0.750	0.173	1.206	0.793	0.084	0.682	0.761	0.086	0.500
350	0.694	0.517	2.184	0.768	0.179	1.333	0.802	0.087	0.777	0.765	0.091	0.595
370	0.761	0.388	2.286	0.784	0.164	1.348	0.790	0.099	0.844	0.780	0.089	0.669
400	0.768	0.381	2.360	0.786	0.177	1.445	0.793	0.106	0.932	0.780	0.095	0.736
440	0.803	0.325	2.511	0.790	0.193	1.571	0.780	0.124	1.020	0.773	0.105	0.812
500	0.830	0.290	2.733	0.806	0.207	1.768	0.788	0.137	1.167	0.764	0.125	0.937
540	0.841	0.271	2.854	0.804	0.230	1.880	0.810	0.133	1.266	0.767	0.133	1.018
600	0.854	0.233	3.000	0.800	0.263	2.025	0.810	0.151	1.402	0.752	0.160	1.132

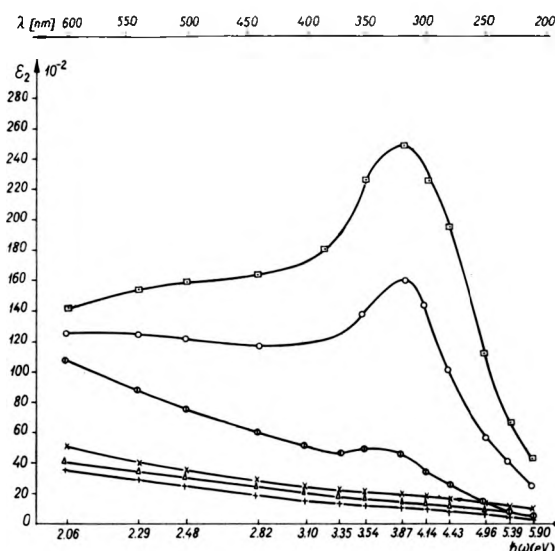


Fig. 6. Optical conductivity vs. wave energy for Al films of various thickness

—□—□—□—□—	56 nm
—○—○—○—○—	75 nm
—△—△—△—△—	115 nm
—×—×—×—×—	180 nm
—◇—◇—◇—◇—	195 nm
— · — · — · — · —	259 nm

curves for thick films ( $d \sim 200$  nm) is monotonic — similarly as that for polycrystalline films [1] — and markedly differs from the shape of the  $\epsilon_2$  and  $\sigma$  curves for thin films ( $d \lesssim 100$  nm), which is characterized by pronounced peaks falling at 3.5–3.9 eV and increasing with decreasing thickness.

#### 4. Conclusions

The appearance of an ordered crystalline structure (of (111) orientation) influences the optical properties of the films. The coefficient of reflection of monocrystalline films is markedly increased in UV and reaches higher values than in the case of polycrystalline films. The imaginary part of the dielectric constant and the optical conductivity of thin monocrystalline films have pronounced peaks at the energies of 3.5–3.9 eV, while the dependence of  $\epsilon_2$  and  $\sigma$  on  $E$  for thicker films has a monotonic plot.

Following ref. [12] and [14] it seems reasonable to consider our results together with the absorption from the surface states of the (111) plane of aluminium. It has been theoretically predicted that the transition of electrons from the surface states of the (111) plane to the Fermi level falls in the range 3.3–6.6 eV. The experimental peaks of  $\epsilon_2$  and  $\sigma$  correspond to the energies of 3.5–3.9 eV. It is of interest to note that these peaks are typical of thinner films alone, for which the surface effects may be of considerable importance.

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#### Оптические свойства эпитаксиальных слоев из алюминия

Напыливались испарением эпитаксиальные слои из алюминия ориентацией (111) на подложке из NaCl и исследовались их оптические свойства. Измерены коэффициенты отражения ( $R$ ) и пропускания ( $T$ ) при нормальном падении как в ультрафиолетовой, так и видимой областях света. Оптические константы  $n$  и  $k$ , определенные с помощью численных расчетов, оказались меньше констант слоев Al, которые напыливались на стеклянной подложке при комнатной температуре. Мнимые части диэлектрической константы ( $\epsilon_2$ ) и оптической проводимости ( $\sigma$ ) обсуждались с учетом зависимости их от энергии волны. Для толстых слоев ( $d \sim 200$  nm) диаграммы  $\epsilon_2$  и  $\sigma$  были монотонными, а в случае более тонких слоев ( $d < 100$  nm) появился заметный пик, который можно связывать с теоретически ожидаемым переходом, соответствующим поверхностным состояниям.

#### References

- [1] MENDLOWITZ H., Proc. Phys. Soc. **75**, 667, 1960.
- [2] HASS G., WAYLONIS J. E., J. Opt. Soc. Am. **51**, 719, 1961.
- [3] BENNET H. E., SILVER M., ASHLEY E. J., J. Opt. Soc. Am. **1089**, 1963.
- [4] COOPER B. R., EHRENREICH H., PHILIPP H. R., Phys. Rev. **132**, 18, 1963.
- [5] HUNTER W. R., J. Opt. Soc. Am. **55**, 1197, 1965.
- [6] SHKLYAREVSKY I. N., YAROVAYA R. J., Optics and Spectroscopy **14**, 130, 1963.
- [7] BURST D., Phys. Rev. **B2**, 818, 1970.
- [8] POWELL C. J., J. Opt. Soc. Am. **60**, 1, 1970.
- [9] ASHCROFT N. W., STURM K., Phys. Rev. **3**, 6, 1971.
- [10] OTTO A., SOHLER W., Solid State Comm. **6**, 17, 1319–1323, 1975.
- [11] PALATNIK L. S. et al., J. Prikl. Spekt. **XXI**, 5, 905–909, 1974.
- [12] TOMÁŠEK M., TALAT G. H., Opt. Appl. **V**, 1, 25, 1975.
- [13] HOFFSTEIN V., Solid State Comm. **10**, 605, 1972.
- [14] BAUDREAU D. S., Surface Sci. **28**, 344, 1971.
- [15] DOBIERZEWSKA-MOZRZYMAS E., Acta Phys. Pol. **A47**, 93, 1975.
- [16] DOBIERZEWSKA-MOZRZYMAS E., OHLY T., WARKUSZ F., J. Cryst. Grow **32**, 129, 1976.
- [17] DOBIERZEWSKA-MOZRZYMAS E., WARKUSZ F., Thin Solid Films **43**, 267, 1977.
- [18] DOBIERZEWSKA-MOZRZYMAS E., LEWANOWICZ S., MOZRZYMAS J., Acta Phys. Pol. **A41**, 3, 251, 1972.
- [19] DAUDE A., SAVARY A., ROBIN S., Thin Solid Films **13**, 255–259, 1972.

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