SPECTRAL INVESTIGATIONS OF PHOTOSTIMULATED EMISSION OF (EXO)ELECTRONS IN VACUUM FROM PLASTICALLY DEFORMED ALUMINIUM COVERED WITH OXIDE LAYER PART III. SLOPE OF THE DECAY CURVE IN ITS INITIAL STAGE

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(Received July, 20, 1965)

It has been found that the slope of the decay curve in $\log \frac{N}{t}$ *vs* t coordinates is in the vicinity of $t = 0$ influenced by the wavelength of the stimulating light. The value of the slope tg α increases with the increase of the wavelength λ.

The dependence of tg α on the intensity I of white and monochromatic light, as well as on the oxide layer thickness D has been studied for various combinations of the parameters: intensity of the stimulating light I, its wavelength λ and thickness D of the oxide layer covering the deformed aluminium.

In the case of white light it was found that with increasing intensity the slope first decreases and then begins to increase in the high intensity range.

The dependences of the slope tg α on λ, I and D obtained in the present work, are analogous as those obtained in previous works under the assumption that the initial stage of the decay of the emission from deformed aluminium is controlled by the space charge of electrons which, do not get out of the fissures.

The results confirm the presumption that the emission of exoelectrons has its roots in the "bottom" of the fissures regarded as a semiconductor layer. It is namely a photoelectric effect on free electrons in aluminium. The electrons enter the oxide layer contacting the metal, and then are emitted from the transient oxide-metal layer into vacuum.

1. *Introduction*

The present work is the third part of extensive investigations on the excited emission of electrons (exoelectrons) in vacuum from plastically deformed aluminium covered with oxide layer. It is a result of continuation of studies on the elucidation of the irreproducibility of the exoelectron emission kinetics. The direct aim of this paper is the etasblishment

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of parameters which essentially influence the decay curve of the photostimulated emission of exoelectrons from deformed aluminium covered with an anodic oxide layer.

From studies carried out in gaseous atmosphere followed that the decay curves of the photostimulated emission from deformed aluminium are influenced by such factors as: composition of the gaseous atmosphere (Sujak, Bójko 1964), humidity in the case of studies in air and the thermal treatment of the samples (Mader 1962), and ions produced by the counter (Sujak, Gieroszyński, Mader 1965).

The studies in vacuum enabled to find the influence of further factors influencing the decay rate of the emission such as: oxide layer thickness D, strength of the external electric field E which accelerates the electrons and the intensity I of the stimulating light.

The parameter which has not been studied up to now and which influences the decay rate of the emission, is the wavelength of the stimulating light λ. The role of the latter in the decay process has been thoroughly discussed in the present paper.

As long as the dependences of the decay constants on all parameters influencing the decay rate are not known, the decay curves cannot be regarded as unambiguous description of physical processes, *e.g.* vacation of electron traps.

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

2. *Experimental arrangement*

The experimental arrangement and the method of preparing the samples have already been described in Part I of the present paper (Gieroszyński, Sujak 1965), where the interested reader may refer to.

3. *Procedure of decay curve studies*

Decay curves of the excited electron emission from deformed aluminium were studied after previous tensile failure of the sample, *i.e.* for $\varepsilon = \varepsilon_g$ (*cf.* Part II of this work). We were interested only in the initial part of the curves. The situation in which the measurements carried out, is illustrated in Fig. 1. The schematic diagram shown in the figure, is essentially

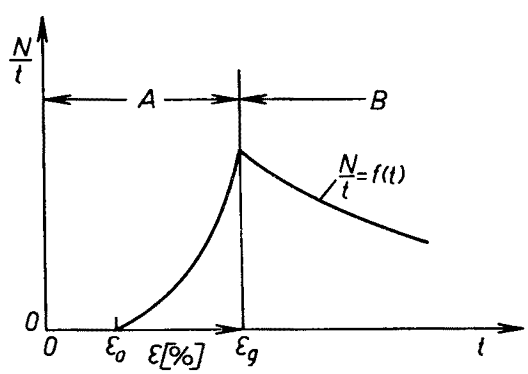

Fig. 1. Change of the emission intensity during the deformation of the aluminium sample (region A) and after failure (region B). ε_0 is the initial deformation, ε_g deformation at which there is a failure of the sample

composed of two regions A and B. Region A is a diagram of the dependence of the photo-stimulated emission intensity for uniform tensile strain on the deformation value $-\frac{N}{t}(\varepsilon)$. In this region we have plotted both the initial derofmation ε_0 and the limiting deformation ε_g for which the sample failure occurs. This part of the diagram was subjected to analysis in Parts I and II of the work. The present paper (Part III) deals with the region B of the diagram shown.

At the instant of the failure the stretching process was stopped and the detector recorded the decay curve $\frac{N}{t} = f(t)$ which has been schematically shown in the region B of Fig. 1. Similarly as in a previous work (Sujak, Gieroszyński, Gajda 1965), the decay of the emission was investigated only for a few minutes. Therefore we have accepted as a measure of the decay rate the value of the slope of the $\frac{N}{t}$ (t)-curve presented in $\log \frac{N}{t}$ *vs* t coordinates,

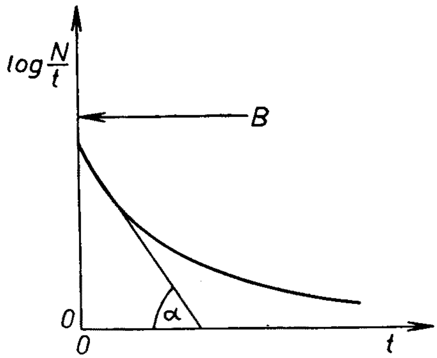

Fig. 2. Decay of the emission intensity after failure of the sample (region B from Fig. 1). α is the angle of the slope of the decay curve presented in $\log \frac{N}{t}$ *vs* t coordinates, in the vicinity of $t = 0$

for t-values close to zero. The curve $\log \frac{N}{t}$ *vs* t is shown schematically in Fig. 2, the measured angle α being marked in the figure.

As will be shown further on, the slope of the decay curve in the initial stage of the observation depends on the wavelength λ and on the intensity of the stimulating light I, as well as on the thickness of the oxide layer covering aluminium

$\mathrm{tg}\,\alpha = f(\lambda, I, D)$, $W_{\text{form}} =$ const, $\frac{d\varepsilon}{dt} =$ const, $E =$ const, $F = 0$, for constant accelerating field strength E $\left(E = \frac{\Delta U}{d}\right)$, fixed way of forming the sample W_{form}, constant deformation rate $\frac{d\varepsilon}{dt}$ and constant humidity of the atmosphere F (for vacuum $F = 0$).

In the consideration of the dependence of $\mathrm{tg}\,\alpha$ on the wavelength, the corrections $\Delta\,\mathrm{tg}\,\alpha$ resulting from the dependence of $\mathrm{tg}\,\alpha$ on the light intensity have been taken into

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

account. The latter dependence of tg α on I is due to the spectral distribution of the source and to different transmission of the interference filters. The relative change of tg α resulting from these corrections did not exceed 5 per cent.

4. *Influence of the wavelength λ of the stimulating light on the slope tg α*

The measurements which have been carried out show that the decay rate of the emission in its initial stage, the measure of with is the tg α-value (after correction by Δ tgα), is strongly influenced by the wavelength of the stimulating light λ. It turms out that tg α increases with the wavelength.

The dependence of tg α on λ found for various oxide layer thicknesses D and presented in $\frac{1}{\text{tg}\,\alpha}$ *vs* $\frac{1}{\lambda}$ coordinates, is a family of straight lines intersecting in a single point. This family can be approximately presented by the equation

$$\frac{1}{\text{tg}\,\alpha} = n_1 \left(\frac{1}{\lambda} - \frac{1}{\lambda_0}\right), \tag{1}$$

where $n_1 = f(D)$, $\lambda_0 = 5500$ Å (D being the parameter). The results of the measurements are shown in Fig. 3.

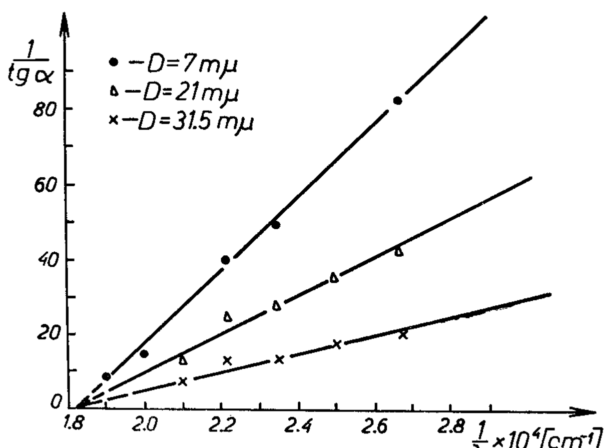

Fig. 3. Dependence of tg α on wavelength λ of the stimulating light for different oxide layer thicknesses. $I = I_0$, $E = 175$ V · cm^{-1}

The functional dependence between n_1 and D can be described by means of the equation (see Fig. 4).

$$n_1 = \frac{m_1}{D - D_1}, \tag{2}$$

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

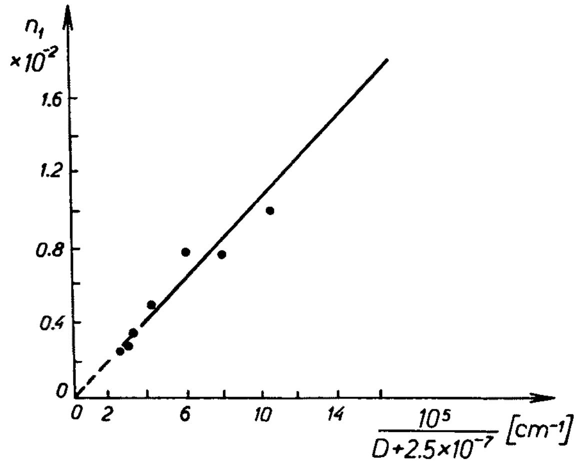

Fig. 4. Dependence of n_1 on the oxide layer thickness D

where $m_1 = 1.09\times10^{-8}$ cm^2, $D_1 = 2.5\times10^{-7}$ cm. Substituting (2) into (1) we obtain the dependence of tg α on λ and D

$$\mathrm{tg}\,\alpha = \frac{D-D_1}{m_1\left(\frac{1}{\lambda}-\frac{1}{\lambda_0}\right)}. \tag{3}$$

5. *Influence of the light intensity I on tg α*

In the first part of the work concerning the influence of the oxide and of measurement parameters on the decay of the photostimulated emission of exoelectrons in vacuum from plastically deformed aluminium (Sujak, Gieroszyński, Gajda 1965), it was found that tg $\alpha \sim I^{-\beta}$, *i.e.*, that the decay rate of the emission measured in the initial stage, decreases with the increase of the intensity of the light applied[1]. This was in contradiction with the results obtained with other Al-samples covered with aluminium oxide layer, and with other experimental arrangement (Lewowska, Sujak 1965; Lewowska, Ph. D. thesis 1965). According to the latter results, the influence of the intensity on tg α should be described by the relation[2] tg $\alpha \sim I^{\beta}$.

This contradiction gave rise to the present study of the influence of the stimulating light on tg α, carried out in a large interval of light intensity.

The light source used in this work was a 110 V/750 W tungsten lamp operated at a diminished voltage of 90 V.

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[1] The light source was a 6 V/50 W tungsten lamp. The incidence angle was 30° and the light converged in a large spot illuminating the sample. The dimensions of the spot were 6 mm×14 mm.

[2] In the investigations carried out by Lewowska the light source was also a tungsten lamp 6 V/50 W, the distance from the sample being however smaller. Besides, the hight pencil was perpendicular to the sample and the diameter of the illuminated spot was about 5 mm.

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

a) White light

It was found that for low intensities of white light the tg α-value really decreases with the increase of the intensity. However, in the higher intensity range there is an inversion of this dependence, *i.e.*, tg α increases with the intensity increase. The behaviuor of tg α with the increase of the intensity of white light is shown in Fig. 6. The parameter of the curves is the thickness of the oxide layer covering the deformed aluminium sample.

If the curves are plotted in the tg α *vs* $\left[\left(\frac{I}{I_0}\right)+a\right]\left(\frac{I}{I_0}\right)^{\beta}$ coordinates, where $\left(\frac{I}{I_0}\right)$ is the relative light intensity, $a = 0.25$, $\beta = 0.33$, then for various oxide layer thicknesses D we will obtain a family of straight lines intersecting in a simple point (Fig. 6). The equation by

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

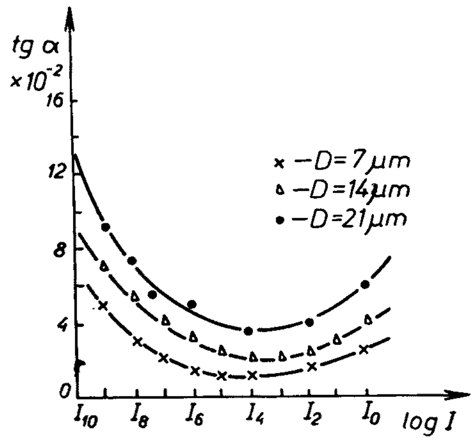

Fig. 5. Change of tg α with the intensity of white light for different oxide layer thicknesses. $E = 175\ \mathrm{V \cdot cm^{-1}}$

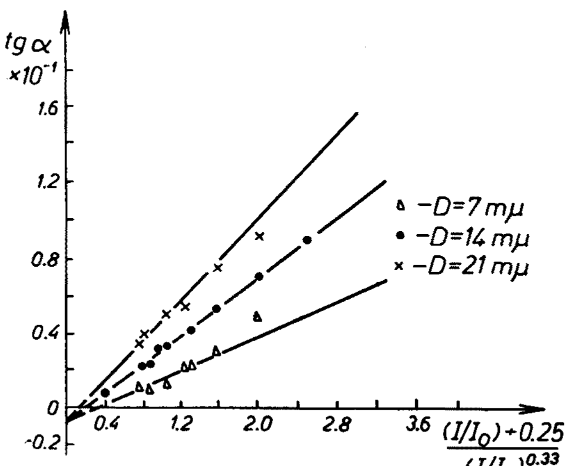

Fig. 6. Dependence of tg α on the light intensity I for different oxide layer thicknesses D. White light, $E = 175\ \mathrm{V \cdot cm^{-1}}$

means of which the experimental results shown in Fig. 6 can be approximately described is

$$\mathrm{tg}\,\alpha = n_2 \frac{\left(\frac{I}{I_0}\right) + a}{\left(\frac{I}{I_0}\right)^{\beta}} - b_1, \tag{4}$$

where $n_2 = f(D)$, $b_1 = 0.6\times 10^{-2}$.

The dependence of n_2 on the oxide layer thickness D is linear, as can be seen from the curves in Fig. 7; we have

$$n_2 = m_2(D - D_2), \tag{5}$$

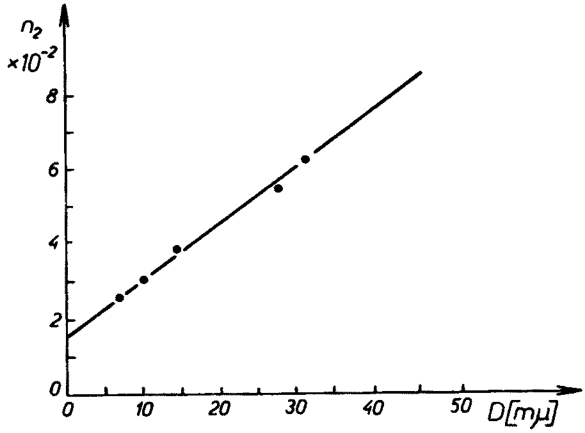

Fig. 7. Dependence of n_2 on oxide layer thickness D

where $m_2 = 1.5\times 104\ \mathrm{cm}^{-1}$, $D_2 = -10^{-6}$ cm.

After substituting (5) into (4) we obtain

$$\mathrm{tg}\,\alpha = m_2 \frac{\left(\frac{I}{I_0}\right) + a}{\left(\frac{I}{I_0}\right)^{\beta}} (D - D_2) - b_1. \tag{6}$$

b) Monochromatic light ($\lambda = 4000$ Å)

For monochromatic light tg α decreases monotonically with the increase of the intensity in the entire studied intensity range.

Fig. 8 shows curves of the dependence of tg α on the intensity of white and monochromatic light. It can be seen from the figure that the tg $\alpha = f(I)$-curve obtained in the case of monochromatic light ($\lambda = 4000$ Å) can be treated as a curve obtained at a smaller maximum light intensity, amounting to about 2^{-4} of the maximum intensity of white light (which corresponds to I_4). This can be understood, if we realise how small is the fraction of energy radiated by a black body into a spectrum interval transmitted by a filter of $\lambda = 4000$ Å.

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

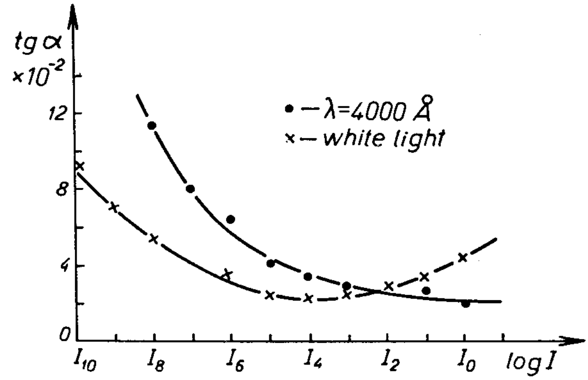

Fig. 8. Comparison of the curves of the relation tg α *vs* I in the case of white light (×—×—) and monochromatic light (●—●—).

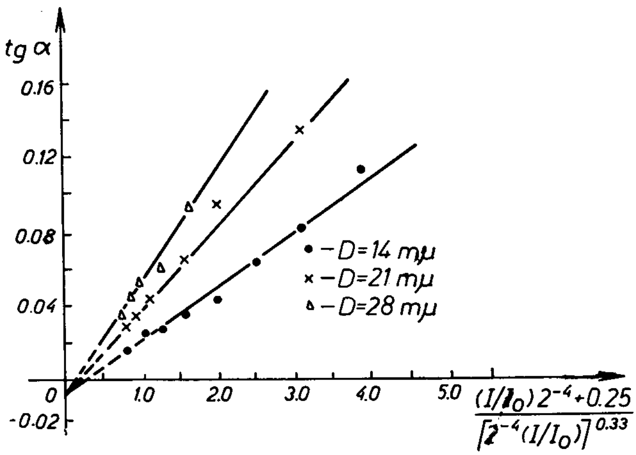

Fig. 9. Dependence of tg α on the light intensity I for different oxide layer thicknesses D. Monochromatic light $\lambda = 4000$ Å, $E = 175$ V · cm^{-1}

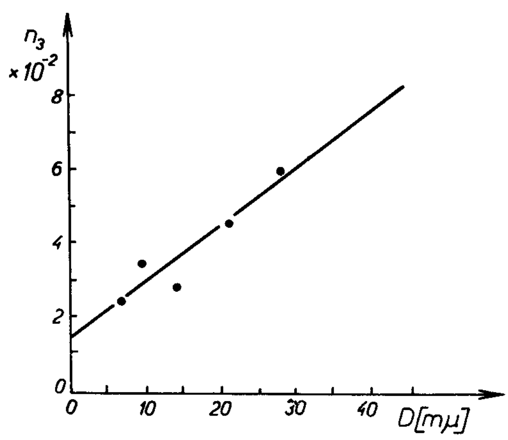

Fig. 10. Dependence of n_3 on oxide layer thickness D

In the case of monochromatic light the dependence of tg α on light intensity is described by an equation which is quite similae to Equation (6) (Fig. 9), namely

$$\operatorname{tg}\alpha = n_3 \frac{r\left(\frac{I}{I_0}\right)+a}{\left[r\left(\frac{I}{I_0}\right)\right]^{\beta}} - b_2, \tag{7}$$

where $n_3 = f(D)$, $b_2 = 0.6\times 10^{-2}$, $r = 2^{-4}$. The dependence of n_3 on the oxide layer thickness D for monochromatic light, is also linear (Fig. 10). There is

$$n_3 = m_3(D-D_3), \tag{8}$$

where $m_3 = 1.53\times 10^{-4}\ \mathrm{cm}^{-1}$, $D_3 = -10^{-6}$ cm. Substituting (8) into (7) we have:

$$\operatorname{tg}\alpha = m_3 \frac{r\left(\frac{I}{I_0}\right)+a}{\left[r\left(\frac{I}{I_0}\right)\right]^{\beta}} (D-D_3) - b_2. \tag{9}$$

6. Influence of the oxide layer thickness D on the tg α-value

a) Parameter λ (wavelength)

From studies of the dependence of tg α on the oxide layer thickness D for various wavelengths λ, we obtain in a tg α *vs* D plot a family of curves with the equation

$$\operatorname{tg}\alpha = n_4\,(D-D_4), \tag{10}$$

where $n_4 = f(\lambda)\mathrm{cm}^{-1}$, $D_4 = -2.5\times 10^{-7}$ cm

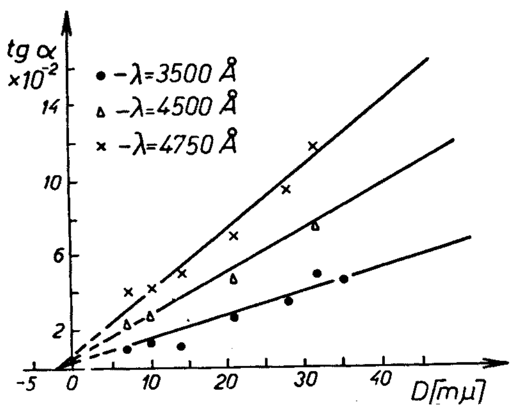

Fig. 11. Dependence of tg α on oxide layer thickness for various wavelengths λ of the stimulating light. $I = I_0$, $E = 175\ \mathrm{V\cdot cm^{-1}}$

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

The functional dependence of n_4 on λ is shown in Fig. 12. This function can be presented analytically by means of the equation

$$n_4 = m_4 \frac{\lambda_0 \cdot \lambda}{\lambda_0 - \lambda}, \tag{11}$$

where $m_4 = 0.91 \times 10^8 \text{ cm}^{-2}$, $\lambda = 5500$ Å. After substituting (10) into (11) we have

$$\text{tg}\,\alpha = \frac{m_4}{\frac{1}{\lambda} - \frac{1}{\lambda_0}} (D - D_4). \tag{12}$$

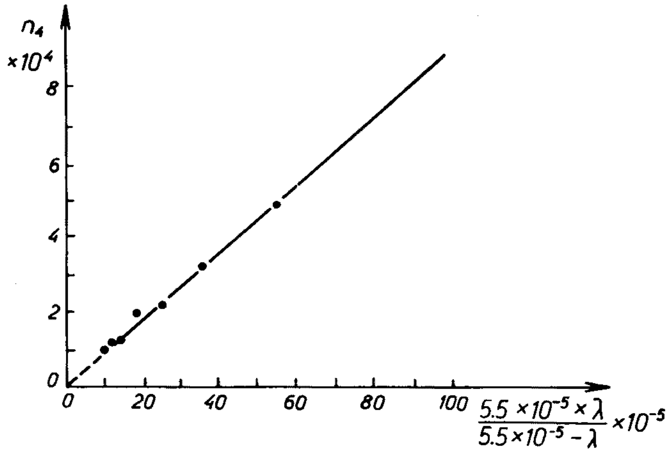

Fig. 12. Dependence of n_4 on the wavelength λ of the stimulating light

From comparison of the relation (12) with (3) it can be seen that $\frac{1}{m_1} \approx m_4$ and $D_1 \approx D_4$.

b) Parameter — I (intensity of white light)

If we analyse the dependence of tg α on the oxide layer thickness D for various intensities of white light I, we obtain in the tg α *vs* D plot also a family of straight lines intersecting in a single point (Fig. 13), with the equation

$$\text{tg}\,\alpha = n_5 (D - D_5), \tag{13}$$

where $n_5\,[\text{cm}^{-1}] = f(I)$ and $D_5 = -2.5 \times 10^{-7}$cm. The function $n_5 = f(I)$, plotted in Fig. 14 can be presented analytically as follows

$$n_5 = m_5 \frac{\left(\frac{I}{I_0}\right) + \alpha}{\left(\frac{I}{I_0}\right)^{\beta}} - b_3, \tag{14}$$

where $m_5 = 2.3 \times 10^4 \text{ cm}^{-1}$, $b_3 = 0.6 \times 10^4 \text{ cm}^{-1}$.

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

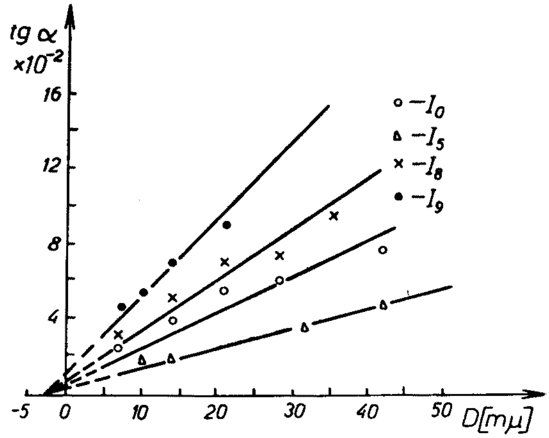

Fig. 13. Dependence of tg α on the oxide layer thickness for different light intensities. White light, $E = 175\text{V}\cdot\text{cm}^{-1}$

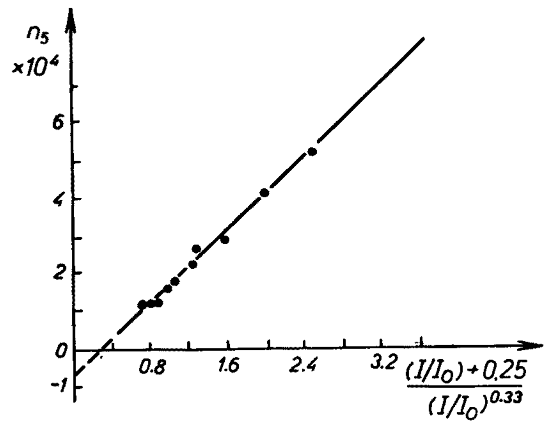

Fig. 14. Dependence of n_5 on the intensity of white light

Substituting (13) into (14) we obtain

$$\operatorname{tg}\alpha = \left[m_5 \frac{\left(\frac{I}{I_0}\right) + a}{\left(\frac{I}{I_0}\right)^{\beta}} - b_3 \right] (D - D_5). \tag{15}$$

c) Parameter I (intensity of monochromatic light $\lambda = 4000$ Å)

In the case of monochromatic light the dependence of tg α on the oxide layer thickness is also linear. The family of straight lines presented in Fig. 15 with I as parameter can be described by means of the equation

$$\operatorname{tg}\alpha = n_6 (D - D_6). \tag{16}$$

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

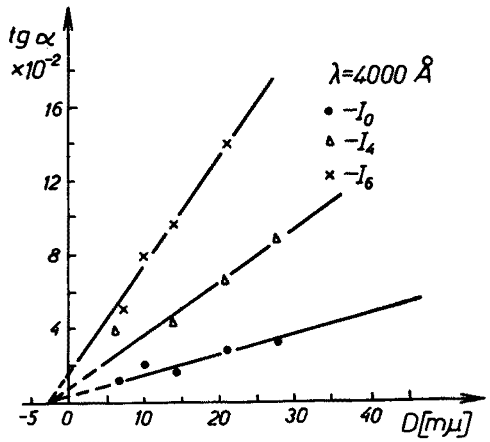

Fig. 15. Dependence of tg α on the oxide layer thickness for different light intensities. Monochromatic light $\lambda = 4000$ Å, $E = 175\ \mathrm{V \cdot cm^{-1}}$

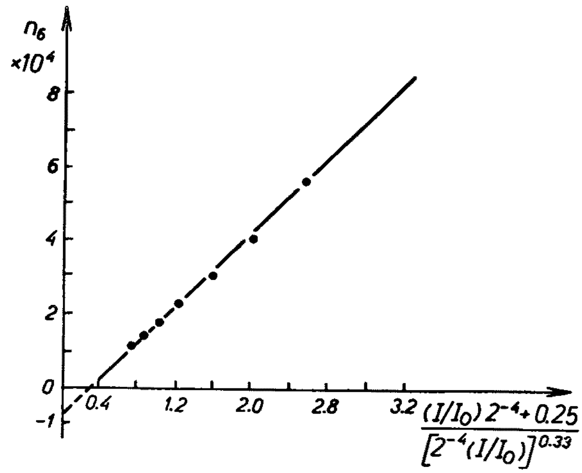

Fig. 16. Dependence of n_6 on the intensity of monochromatic light ($\lambda = 4000$ Å)

where n_6 [cm^{-1}] $= f(I)$ and $D_6 = -2.5 \times 10^{-7}$ cm. The dependence $n_6 = f(I)$ plotted in Fig. 16 can be described by means of the equation

$$n_6 = m_6 \frac{r\left(\frac{I}{I_0}\right) + a}{\left[r\left(\frac{I}{I_0}\right)\right]^{\beta}} - b_4, \tag{17}$$

where $m_6 = 2.25 \times 10^4\ \mathrm{cm^{-1}}$, $r = 2^{-4}$, $b_4 = 0.6 \times 10^4\ \mathrm{cm^{-1}}$.

Substituting (17) into (16) we obtain the following relation between tg α and the intensity of monochromatic light,

$$\operatorname{tg}\alpha = \left\{ m_6 \frac{r\left(\frac{I}{I_0}\right)+a}{\left[r\left(\frac{I}{I_0}\right)\right]^{\beta}} - b_4 \right\} (D-D_6). \tag{18}$$

As it is seen, the equations describing the dependence of tg α on the intensity of white light I, are qualitatively analogous, both for white (15) and monochromatic light (18).

The only condition required, is the presence of the wavelength $\lambda = 5500$ Å in the illuminating light beam.

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

7. Discussion

The theoretical considerations concerning the decay of emission from deformed aluminium, which were presented in a previous paper (Gieroszyński, Sujak 1965), contained already the idea of spacial charge of electrons which did not escape outside the fissure. The emission decay rate was determined by the kinetics of the increase of this charge. The formula for tg α derived from these considerations was

$$\operatorname{tg}\alpha = A \frac{DE_0}{T} + \lambda^*_{D=0}, \tag{19}$$

where A is a constant, D is the oxide layer thickness, E_0 the effective field strength in the fissure, T the kinetic energy of electrons emitted from the bottom of the fissure and $\lambda^*_{D=0}$ the decay constant of the first component for $D \to 0$. The relation (19) was qualitatively confirmed in a previous work. It has also been confirmed by the present results, as will be shown below.

From the relation between tg α and oxide layer thickness D (Formula 19) for tg $\alpha = 0$ we have $\lambda^*_{0=0} = -A \frac{D_0 E_0}{T}$, where D_0 is the value of the oxide layer thickness extrapolated to tg $\alpha = 0$. Formula (19) can then be written in the form

$$\operatorname{tg}\alpha = A \frac{E_0}{T} (D-D_0). \tag{20}$$

If the kinetic energy of electrons emitted from the "bottom" of the fissure T is somehow connected with the wavelength λ of the stimulating light by means of Einstein's equation for photoeffect on free electrons, then

$$\operatorname{tg}\alpha = A \frac{E_0}{hc\left(\frac{1}{\lambda} - \frac{1}{\lambda_k}\right)} (D-D_0), \tag{21}$$

where λ_k is the limiting wavelength for the photoelectric effect.

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

The empirical relations (3) and (12) which describe tg α as a function of λ and D, correspond to the above Equation (21). It is only necessary to put

$$\frac{AE_0}{hc} \approx \frac{1}{m_1} \approx m_4 \quad \text{and} \quad D_1 = D_0 .$$

The influence of the light intensity I on tg α can be understood, if we assume that there is a change of the effective emission work of electrons emitted from the "bottom" of the fissure, when the light intensity is changed. Thus the kinetic energy of electrons emitted from the "bottom" of the fissure will change as I^{β}, where $\beta = \frac{1}{3}$ for nonlinear and $\beta = \frac{2}{3}$ for linear recombination of electrons from the conductivity band.

After taking into account the surface density of emitting centres $\sigma = f(I)$ which is inherent in the constant A, and which has not been considered in previous papers (Gieroszyński, Sujak 1965), Formula (19) becomes

$$\text{tg}\,\alpha \sim \frac{A_0 \sigma E_0}{I^{\beta}} (D - D_0), \tag{22}$$

where $A_0 = \frac{A}{\sigma}$, $\beta = \frac{1}{3}$ or $\frac{2}{3}$.

The experimental relations of tg α (6), (9), (15) and (18) with light intensity I and oxide layer thickness D correspond as far as the shape is concerned, to the relation (22), for both white and monochromatic light.

From relation (6) one can determine the value of σ, by comparing the former relation with Formula (22).

$$\sigma = \left(\frac{I}{I_0}\right) + a - c \left(\frac{I}{I_0}\right)^{\beta}, \tag{23}$$

where $c = f(D)$ in the considered thickness interval amounts from 0.25—0.08. In Fig. 17 σ is presented as a function of I. In the case of white light we obtain an increase of σ with

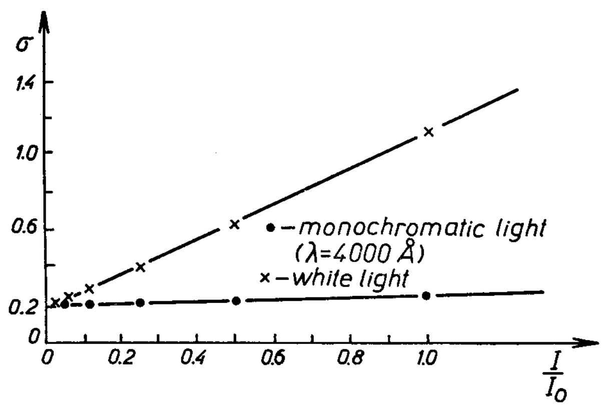

Fig. 17. The shape of the function σ *vs* intensity of white light (× — × —), and monochromatic light (λ = 4000 Å) (•—•—)

increasing light intensity and this relation can be approximated by a straight line. For monochromatic light ($\lambda = 4000$ Å) σ is almost independent of the intensity.

For this reason, while stimulating the emission with the use of monochromatic light, no increase of tg α with increasing intensity was observed. Similarly, in a previous paper (Sujak, Gieroszyński, Gajda 1965) it was found that tg α decreases with the increase of the intensity of the stimulating light. It should be reminded that the light source used in the latter work yielded a much smaller intensity than that used in the present one.

The assumption that in Formula (20) the kinetic energy of the emitted electrons will display its influence according to the relation which results from Einstein's equation for photoelectric effect on free electrons, has been confirmed experimentally (*cf.* relation (3)). The limiting wavelength of the photoelectric effect $\lambda_k = 5500$ Å determined experimentally, in agreement with that obtained from considerations concerning the limiting oxide layer thickness D_g (Part II of the present paper). Furthermore, the linear incresse of the surface density of the emitting centres σ with increasing light intensity in the high intensity range is also a check of the photoeffect occuring on free electrons.

The shift of the photoelectric wavelength limit from 2950 Å for pure aluminium surface to the value of 5500 Å obtained in our experiments, should therefore be regarded as due to the diminishmet of the emission work of electrons from metal to oxide through contact of metal with semiconductor.

Thus, it seems that direct, photostimulated emission of exoelectrons from deformed aluminium takes place from the transient metal-oxide layer which is the "bottom" of the fissure, or from rapidly oxidized pure aluminium surface and is photoelectric effect on free electrons supplied from the metal to the oxide.

The experimental value of the exponent $\beta = \frac{1}{3} = 0.33$ would suggest again a nonlinear recombination of electrons from the conductivity band of this transient layer.

ACTA PHYSICA POLONICA Vol. XXIX (1966) Fasc. 4

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