KINETICS OF CHARGES ACCUMULATED IN Al-Al_2O_3-Ag TYPE SYSTEMS DUE TO ELECTRON BOMBARDMENT*

BY A. GIEROSZYŃSKI

Laboratory for Solid State Cryophysics, Institute of Experimental Physics, University of Wrocław**

(Received December 29, 1975; final version received September 29, 1976)

The time dependence of charges accumulated inside, and on the surface of thin films due to bombardment with electrons was investigated for Al–Al_2O_3–Ag type thin film systems. The dependence was determined from the analysis of energy distributions of exoelectrons emitted by the plastically deformed Al–Al_2O_3–Ag systems. It is shown that the surface charge originates from the transport of a part of the charge accumulated inside the oxide to the surface. The transport phenomenon is stimulated by a strong electric field existing inside the oxide layer. The values of surface resistance and volume resistance for the passage of charges to the base were estimated. They are confined to the interval $8\times10^9-6\times10^{10}$ Ω.

1. Introduction

The influence of strong electric fields on the exoelectron emission has been recorded since the fifties (Sujak 1956; Sujak 1961; Hanle, Kanzler, Scharmann 1961), and recently Dreckhan, Gross, Glaefeke (1970); Sujak, Gieroszyński, Gieroszyńska (1974). Much valuable information about their role in the emission mechanism has been obtained from the exploitation of the energy analysis of exoelectrons.

It was found (Gieroszyński 1977) that the emission of exoelectrons might be stimulated by strong electric fields appearing in the insulator after bombardment with electrons. In that work the tunneling model of exoelectron emission from electron traps was proposed. In the present investigation the model of Gieroszyński will be used to explain the time dependence of charges accumulated in the Al–Al_2O_3–Ag system.

2. Apparatus and measurements

The apparatus used in this experiment, and the performance of measurements were identical to those described in the papers, of Sujak, Gieroszyński, Gołek, Lesz (1975) and Sujak, Gieroszyński, Gieroszyńska (1977).

---

* Work supported by the Polish Academy of Sciences.

** Address: Zakład Kriofizyki Ciała Stałego, Instytut Fizyki Doświadczalnej, Uniwersytet Wrocławski, Cybulskiego 36, 50-205 Wrocław, Poland.

The thickness of the aluminium oxide in the $Al-Al_2O_3-Ag$ system was equal to 1960 Å, the thickness of the Ag layer was about 30 Å.

The samples were plastically deformed after bombardment with electrons. The time lapse t_{pb} from the end of bombardment to the beginning of deformation varied from 110 to 650 seconds. Measurements of electron emission were conducted from the moment of deformation until about 400 s afterwards. All measurements of emission were performed without light stimulation at room temperature.

3. The effect of the time lapse t_{pb} on emission kinetics and the energy distributions of exoelectrons

Emission kinetics and the energy distribution of exoelectrons essentially depend on the time interval t_{pb} separating the end of electron bombardment of a sample and the beginning of plastic deformation. In Fig. 1 emission kinetics recorded both during and

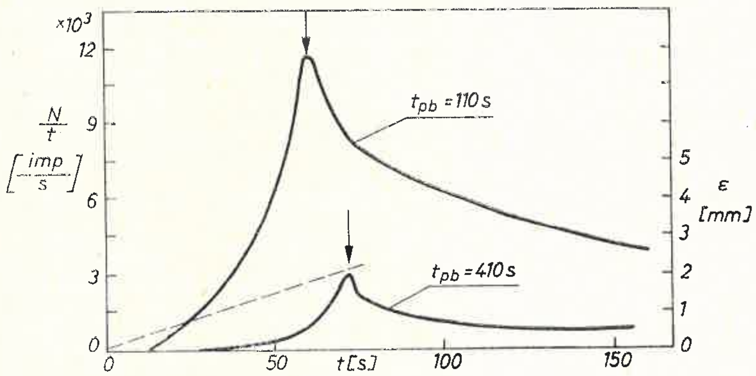

Fig. 1. Kinetics of exoelectron emission from the deformed $Al-Al_2O_3-Ag$ system for various times t_{pb} after the electron bombardment. Arrows mark the moment of sample break. The dotted line shows the dependence of sample elongation ε on time t

after the deformation of samples for various times t_{pb} are shown. It is observed that the emission intensity $(N/t)_{br}$ recorded at the moment of sample break strongly decreases with an increase in the time t_{pb} (Fig. 2).

Also the retardation curves (Fig. 3), and energy distributions (Fig. 4) depend strongly on the time t_{pb}. When t_{pb} increases the absolute value $|U_{c0}|$ of the potential at which the first retardation curve reaches the saturation point initially increases, develops a maximum for $t_{pb} \approx 250$ second and then decreases (Fig. 5). The value E_{m0} of the maximum energy corresponding to the first retardation curve decreases with a decrease of t_{pb} in the whole investigated interval (Fig. 6).

The absolute value $|U_c|$ of the saturation potential, and the maximum energy E_m monotically decrease with the time t after the deformation (Fig. 7).

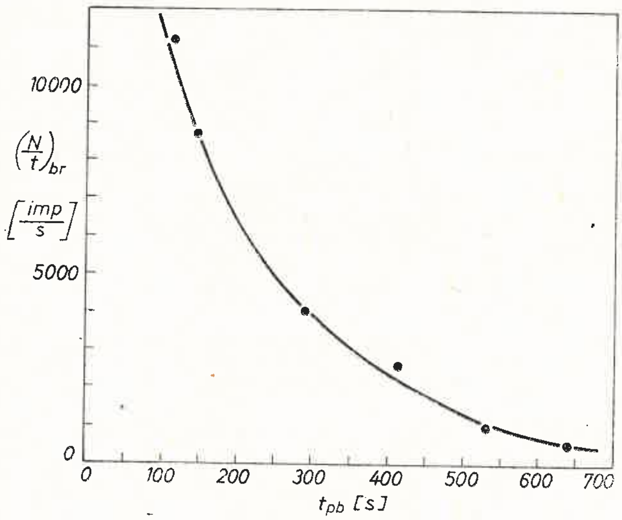

Fig. 2. Dependence of the emission intensity $\left(\frac{N}{t}\right)_{br}$ at the moment of sample break on time t_{pb} after the electron bombardment

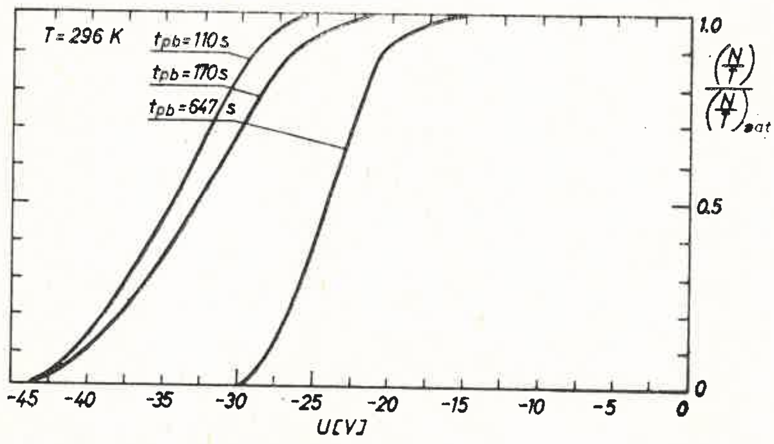

Fig. 3. Retardation curves for various times t_{pb}

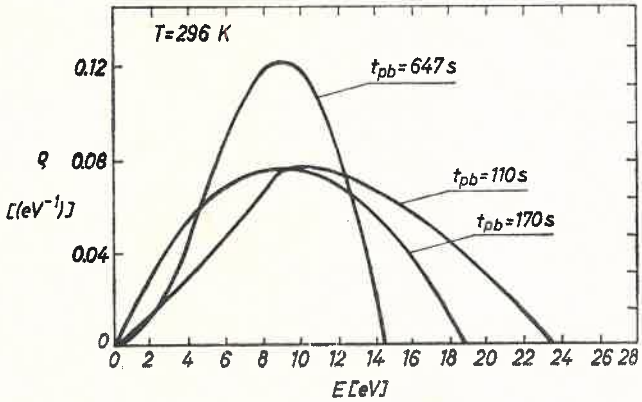

Fig. 4. Energy distributions of exoelectrons corresponding to the retardation curves of Fig. 3

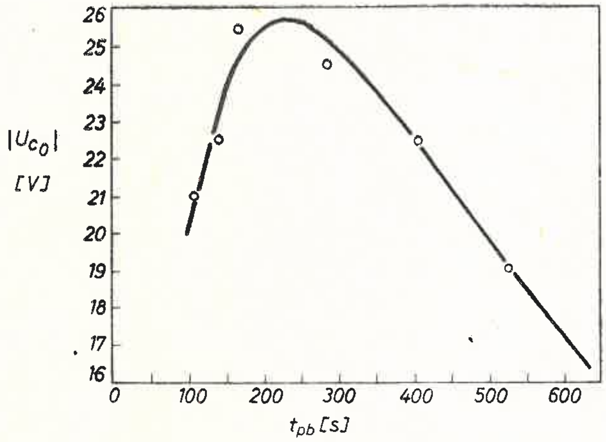

Fig. 5. Dependence of the absolute value $|U_c|$ of the saturation potential on time t_{pb} after the bombardment

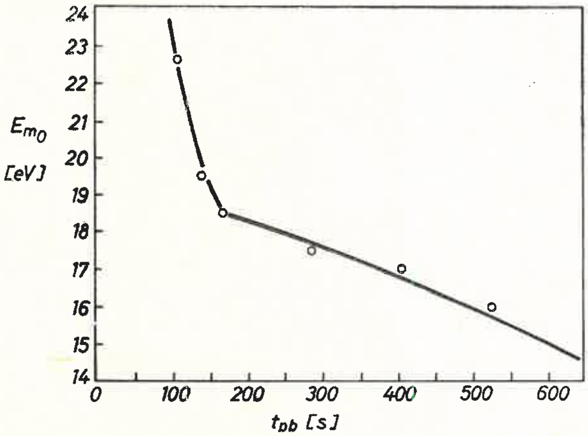

Fig. 6. Dependence of the maximum energy E_{m_0} on time t_{pb}

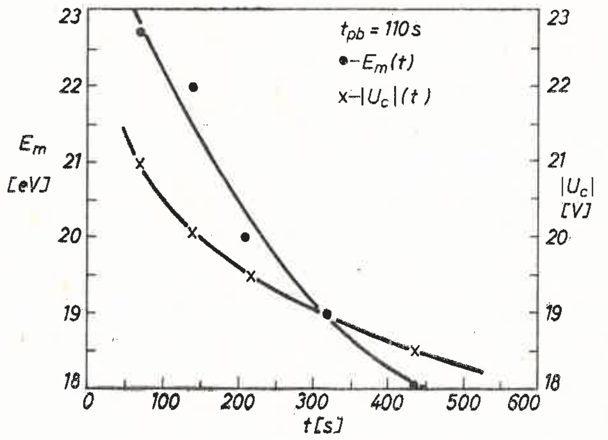

Fig. 7. Variation of the potential $|U_c|$ and energy E_m with time t after the deformation of a system

4. *Discussion*

Changes in emission intensity and energy distributions of exoelectrons described above can be explained by a consideration of the processes of transport and neutralization of charges accumulated inside and on the surface of the insulator layer.

In the paper of Gieroszyński (1977) it was shown that the intensity of emission of exoelectrons from electron traps via the tunneling effect depends on the magnitudes of volume charge Q_w and surface charge Q_p accumulated in the insulator. Also the maximum energy E_m of the emitted exoelectrons and the surface potential of the emitter are functions of Q_p and Q_w.

The charge Q_w accumulated inside the insulator is subject to neutralization due to the passage of charges from Al_2O_3 through a resistance R to the earthed Al base. Therefore, a change in the charge Q_w with time t can be described by the exponential function

$$Q_w = Q_{w_0} \exp\left[-\frac{t}{\tau_w}\right], \qquad (1)$$

where Q_{w_0} — initial charge ($t = 0$), τ_w — time constant specific for a system.

The constant τ_w is equal to $\tau_w = R \cdot C$, where R is the resistance which the system discharges through, and C is the electric capacity of a system.

If it is assumed that the time t is counted from the moment of the appearance of cracks in the oxide layer subject to deformation, then formula (1) describes the process of discharge of the deformed system; τ is the corresponding time constant of the deformed system.

The neutralization processes occur also within the time interval from the end of electron bombardment to the beginning of deformation, i.e. before the deformation starts.

The charge Q_{w_0} is, therefore, a function of time t_{pb} from the end of bombardment to the beginning of deformation.

$$Q_{w_0} = Q_{w_0}^* \exp\left[-\frac{t_{pb}}{\tau_w^*}\right], \tag{2}$$

where $Q_{w_0}^*$ is the magnitude of charge accumulated inside the oxide at the moment of the end of electron bombardment ($t_{pb} = 0$); τ_w^* is the time constant of the undeformed system.

The surface charge Q_p depends on the magnitude of charge transported from the volume to the surface, and on the neutralization rate of this charge via the surface resistance of the system.

Let us consider the process of accumulation of the charge Q_p for an undeformed system. The charge $Q_{p_0}^*$ supplied from the volume depends on the intensity I_p of the electron current reaching the base and on time t_{pb}

$$Q_{p_0} = I_p^* t_{pb}. \tag{3}$$

Now, taking into account the neutralization of this charge we get the following formula giving the charge Q_{p_0} accumulated on the surface.

$$Q_{p_0} = I_p t_{pb} \exp\left[-\frac{t_{pb}}{\tau_p^*}\right], \tag{4}$$

where τ_p^* is a time constant for the undeformed system.

Assuming further that after the deformation the neutralization process predominates over the charge supplying processes, we find that

$$Q_p = Q_{p_0} \exp\left[-\frac{t}{\tau_p}\right], \tag{5}$$

where τ_p is the time constant for the deformed system. The current I_p of electrons reaching the surface from the volume is also a function of time. The magnitude of I_p can be estimated by considering a formula for the current of electrons coming to the surface after their release from the traps in a strong electric field (Gieroszyński 1977).

I_p equals the difference between the intensity I_d of all electrons reaching the surface and the intensity I_e of the emitted electrons.

$$I_p = I_d - I_e. \tag{6}$$

The current intensity I_e can be described by the following approximate formula (Gieroszyński 1977):

$$I_e = Q_w H \exp\left[-\left(\frac{a}{Q_w - 2Q_p}\left(E_a^{3/2} + \frac{\Phi}{b}\right)\right)\right], \tag{7}$$

where H, a, b are constants, E_a is the trap activation energy ($E_a \approx 0.7$ eV), Φ — height of the potential barrier at the emitter surface ($\Phi \approx 0.6$ eV). The values of constants are

$a = 1.17 \times 10^{-7}\ C(eV)^{-3/2}$, $b = 6.62\ (eV)^{-1}$. We get the current intensity I_d by letting $\Phi = 0$ in formula (7). Substituting the formulas for I_d and I_e into (6) we get

$$I_p = Q_w H \exp\left[-\frac{aE_a^{3/2}}{Q_w - 2Q_p}\right]\left\{1 - \exp\left[-\frac{a\Phi}{(Q_w - 2Q_p)b}\right]\right\}. \tag{8}$$

In the paper of Gieroszyński (1977) it was shown that knowledge of the potential U_c of the surface relative to the base, and of the maximum energy E_m of exoelectrons suffices to estimate the charges Q_p and Q_w

$$Q_p = \frac{\varepsilon S}{8\pi\delta}|U_c|$$

$$Q_w = \frac{\varepsilon S}{4\pi\delta}\left[\frac{4E_m}{e} + |U_c| + \sqrt{\frac{4E_m^2}{e^2} + \frac{2E_m|U_c|}{e}}\right], \tag{9}$$

where ε — dielectric constant of aluminium oxide ($\varepsilon = 10$), S — surface area ($S = 0.88\ cm^2$), δ — half oxide thickness ($\delta \approx 10^{-5}$ cm).

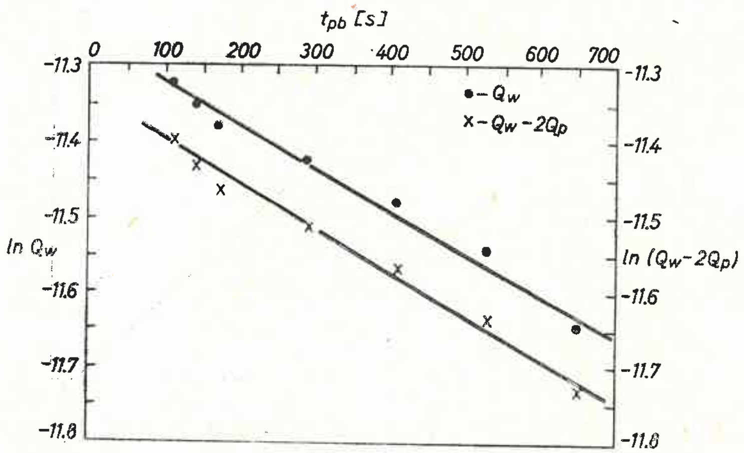

Fig. 8. Dependence of the charges Q_w and $Q_w - 2Q_p$ on time t_{pb}

The functional dependence of Q_w and $Q_w - 2Q_p$ on time t_{pb} is plotted in Fig. 8. It has been calculated with formula (9) from the experimentally determined dependence of energy E_m and potential $|U_c|$ on time t_{pb} (Fig. 5 and 6). Fig. 8 shows that the curves can be approximated by exponential functions.

$$Q_w - 2Q_p = Q_1 \exp\left[-\frac{t_{pb}}{\tau_1}\right]$$

$$Q_w = Q_2 \exp\left[-\frac{t_{pb}}{\tau_2}\right], \tag{10}$$

where the time constants $\tau_1 \approx \tau_2 = 1.8 \times 10^3$ sec, and the charge $Q_1 = 1.6 \times 10^{-5}$ C.

Since in the investigated time interval we have $\frac{t_{pb}}{\tau_1} \ll 1$, therefore we can write

$$Q_w - 2Q_p \approx \frac{Q_1}{1 + \frac{t_{pv}}{\tau_1}}. \tag{11}$$

Now, substituting (11) and (10) to (8) and dropping the terms slowly varying with t_{pb} we get

$$I_p = M \exp\left[-t_{pb} \cdot \beta_0\right], \tag{12}$$

where M is a constant, and $\beta_0 = \frac{\frac{aE_a^{3/2}}{Q_1} + 1}{\tau_1}$.

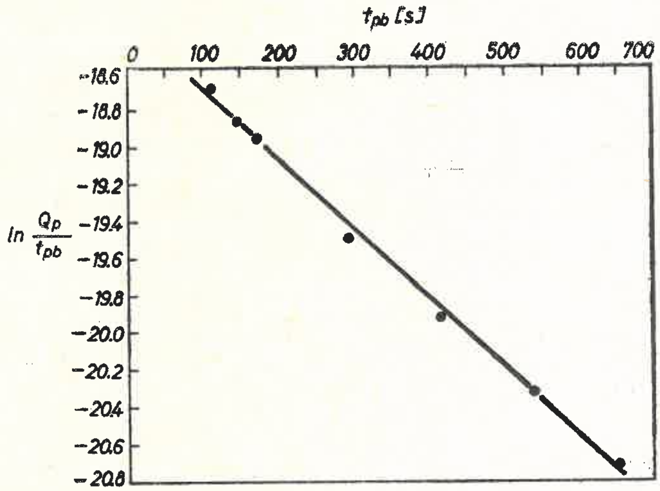

Fig. 9. Dependence of the surface charge Q_p on time t_{pb} in the system $\ln\left[\frac{Q_p}{t_{pb}}\right]$ versus t_{pb}

Substituting (12) into (4) and making use of relation (5) we have

$$Q_p = M t_{pb} \exp\left[-t_{pb}\left(\beta_0 + \frac{1}{\tau_p^*}\right)\right] \cdot \exp\left[-\frac{t}{\tau_p}\right]. \tag{13}$$

Formula (9) was used to determine the function $Q_p(t_{pb})$ from knowledge of the experimental curve $U_c(t_{pb})$ plotted in Fig. 5. When the curve $Q_p(t_{pb})$ is drawn in the system $\ln[Q_p/t_{pb}]$ versus t_{pb} then we get a straight line (Fig. 9). Therefore, the dependence. of Q_p on t_{pb} is described by

$$Q_p = A t_{pb} \exp\left[-\frac{t_{pb}}{\tau_3}\right], \tag{14}$$

where the time constant $\tau_3 = 2.65 \times 10^2$ sec.

In all measurements the time lapse from the beginning of deformation to the recording of the first retardation curve was approximately constant and equaled $t = t_n \approx 70$ sec.

Putting $t = t_n$ in formula (1) and substituting (2) to (1) we find $\tau_1 \approx \tau_2 = \tau_w^*$ after the comparison with the experimental dependence of Q_w on t_{pb} (formula 10).

The internal resistance R_w^* of an undeformed system can be determined from

$$R_w^* = \frac{\tau_w^*}{C}. \tag{15}$$

The capacity of the system was $C = 3.9 \times 10^{-8}$ F, therefore, we get $R_w^* \approx 4.6 \times 10^{10}\ \Omega$. Subsequently, putting $t = t_n$ in formula (13) and comparing it with formula (14) we find $\beta_0 + 1/\tau_p^* = 1/\tau_3$. Substitution of the known values of β_0 and τ_3 then yields $\tau_p^* = 3.1 \times 10^2$ sec. The surface resistance R_p^* is given by

$$R_p^* = \frac{\tau_p^*}{C}. \tag{16}$$

The value of R_p^* was $8 \times 10^9\ \Omega$.

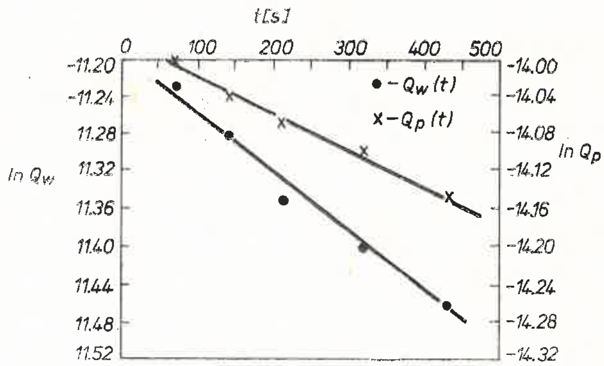

Fig. 10. Variation of the charges Q_p and Q_w with time t after the deformation of a system

For a fixed time lapse following the bombardment ($t_{pb} = 110$ sec) the changes in Q_w and Q_p with time t should be described by relations (1) and (5). Indeed, the experiments showed that the dependence of Q_w and Q_p on time t recorded from the moment of deformation can be fitted by exponential functions (Fig. 10)

$$Q_p = Q_{01} \exp\left[-\frac{t}{\tau_{01}}\right],$$

$$Q_w = Q_{02} \exp\left[-\frac{t}{\tau_{02}}\right], \tag{17}$$

where the time constants $\tau_{01} = 2.5 \times 10^3$ s, $\tau_{02} = 1.6 \times 10^3$ s. A comparison of formulas (1) and (5) with (17) yields $\tau_{01} = \tau_p$ and $\tau_{02} = \tau_w$.

Surface resistance R_p and volume resistance R_w were accordingly equal to

$$R_p = \frac{\tau_p}{C} = 6.4 \cdot 10^{10}\Omega,$$

$$R_w = \frac{\tau_w}{C} = 4.1 \cdot 10^{10}\Omega.$$

An increase of the surface resistance after deformation can be explained by the appearance of cracks in a deformed system.

The author would like to thank Professor B. Sujak, Director of the Laboratory of Solid State Cryophysics of the University of Wrocław, for suggesting the project and for valuable advice in the course of the investigation.

REFERENCES

Drenckhan, J., Cross, H., Glaefeke, H., *Phys. Status Solidi* (a) **3**, K 185 (1970).
Gieroszyński, A., *Acta Univ. Wratislaw.*, 1977, in press.
Hanle, W., Kanzler, G., Scharmann, A., *Z. Phys.*, **162**, 483 (1961).
Sujak, B., *Acta Univ. Wratislaw.*, **1B**, 151 (1956).
Sujak, B., *Acta Phys. Pol.* **20**, 969 (1961).
Sujak, B., Gieroszyński, A., Gieroszyńska, K., *Acta Phys. Pol.* **A46**, 3 (1974).
Sujak, B. Gieroszyński, A. Gieroszyńska, K., *Acta Univ. Wratisl.*, 1977, in press.
Sujak, B., Gieroszyński, A., Gołek, F., Lesz., J., *Acta Phys. Pol.* **A47**, 875 (1975).