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On a current mechanism in Ta_2O_5 thin films

Research Article

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Abstract: Electrical conduction in the temperature range of 120-370 K has been studied in sandwiched structures of Al/Ta_2O_5/Si. The tantalum oxide films were prepared by evaporation of tantalum on a p-Si crystal substrate, followed by oxidation at a temperature of 600˚C. The temperature-dependent current-voltage ($I-V$) characteristics are explained on the basis of a phonon-assisted tunnelling model. The same explanation is given for $I-V$ data measured on Ta_2O_5 films by other investigators. From the comparison of experimental data with theory the density of states in the interface layer is derived and the electron-phonon interaction constant is assessed.

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Keywords: Ta_2O_5 • Frenkel emission • phonon-assisted tunnelling

1. Introduction

Tantalum pentoxide (Ta_2O_5) has been extensively investigated as a promising material in a number applications such as dielectric materials for storage capacitors, thin film transistors, and the coating material for solar cells. Since Ta_2O_5 has a dielectric constant k of 25-35 compared to ≈4 for SiO_2, Ta_2O_5 is one of the most suitable materials to replace SiO_2 in metal-oxide-semiconductor (MOS) devices, and particularly for development of future high-density dynamic random access-memories (DRAMs) [1] .

There are many studies on the leakage current mechanism in thin films grown by various methods, with different electrode materials deposited on different substrates [2–13]. Leakage current-voltage characteristics have been explained in most cases by the Frenkel-Poole (FP) mechanism (bulk limited currents) [3–5, 7–13] or by the Schottky emission of electrons from electrodes [2, 7]. However, these models often faced difficulties when a quantitative comparison with experimental data was performed. For example, in Ref. [2] the fit of $I-V$ data measured in Al/Ta_2O_5/Al structures with the FP model required a very large dielectric constant (≈299), whereas the Schottky model application required the value of $k = 3.09$ for the best fit. In Ref. [7], the conduction of amorphous and crystalline films has been explained by the Schottky mechanism at intermediate fields, whereas at higher fields (>350 kV/cm) priority has been given to the FP model. The authors of [7] decline the possible tunnelling mechanism of conduction because, in their mind, Fowler-Nordheim tunnelling is inherently independent of the temperature, while the current exhibited an obvious dependence on temperature. However, in [6] it was shown that temperature-dependent $I-V$ characteristics can be successfully explained on the

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basis of phonon-assisted tunnelling, which is an inherently temperature dependent tunnelling process.

Nevertheless, in a series of recently published works [10–13], the authors also point to Frenkel emission as the dominant mechanism of current in metal/Ta_2O_5/Si structures. Therefore, we want to show that in order to explain the temperature dependent $I-V$ data measured for Ta_2O_5 thin films, as well as for the ones of the other oxides [14], phonon-assisted tunnelling must be taken into account. This assertion will be proved by the analyses of our own data measured on metal/Ta_2O_5/Si structures and also data obtained by other investigators.

2. Experiment

The metal/Ta_2O_5/Si structures were prepared by evaporation of tantalum on a p-type single crystal substrate heated to 150°C. The tantalum film oxidation was performed in an oxygen atmosphere at a temperature 600°C. The thickness of the Ta_2O_5 films was found to be 0.1 to 0.5 μm. The aluminium electrodes with a diameter of 1 mm were sputtered through a shadow mask. The dc leakage currents were measured both for positive and negative potentials of the Al electrode, and the measurements were carried out with a 5 s charging time to reach the steady state dc regime.

The current was measured in a vacuum cryostat using either an electrometer (for the current range from 10^{-12} to 10^{-8} A) or a double-coordinate potentiometer (10^{-9} to 10^{-3} A).

The diodes under investigation have been found to rectify the current. Negative voltage applied to the Al-electrode resulted in current values two orders of magnitude higher than those at positive voltages. Current-voltage ($I-V$) dependences at various temperatures were measured for twenty samples of different thickness.

The typical curves of current dependence on voltage measured at different temperatures for the Al/Ta_2O_5/Si sample of 0.5 μm thickness for negative and positive Al electrode potentials are shown in Figs. 1 and 2, respectively. The distinct features of these $I-V$ data is superlinearity in the region of higher bias voltages and evident dependence of the $I-V$ characteristics on temperature. The dependences of the current on the temperature were found to be stronger in the low-voltage region, where the activation energy was estimated to be about 0.9 eV for Al^- and 1.05 eV for Al^+. The influence of thickness on $I-V$ data for our samples was negligible. The dependences of the current on temperature at different applied voltages have been also measured, due to their importance for the adequate choice of the charge carrier transport model.

The current dependences on the reciprocal temperature measured at different values of applied voltages are shown in Fig. 3. The $\ln I$ against $1/T$ dependences are seen to be linear only at low voltages. With the rise in voltage, the slope of these curves is found to increase (the thermal activation energy decreases) and bowing of the curves is also observed. Such behaviour of the $\ln I$ against $1/T$ dependences obtained in Ta_2O_5 films can be explained by neither the FP nor Schottky mechanism because these mechanisms predict only a linear dependence of $\ln I$ on $1/T$. Therefore, in order to explain the observed peculiarities of current dependence on temperature and applied voltage, the phonon-assisted tunnelling (PhAT) model, which was formerly used for describing similar data measured in some diodes [6, 14, 15], will be used.

3. Comparison of experimental results with theory

In accordance with the PhAT model, current transport through the barrier is governed by

the process of phonon-assisted electron tunnelling from states nearby the metal-oxide interface to the conduction band of the oxide. The electron population in the states is assumed to be independent of bias voltage due to the continuous filling of interface states from the electrode. If the electrons released from traps dominate the current in the oxide, then the current density will be equal to $j = eN_SW$, where e is the electronic charge unit, N_S is the carriers density in the interface states, and W is the electron tunnelling rate from these states, which is a function of field strength E and temperature T. Thus, $j \approx W(E,T)$ and that is why we can fit the current dependences obtained by measurement with the theoretical tunnel transition through the barrier rate dependences on temperature.

For the computation of the transition rate $W(E,T)$ a relatively simple equation derived for electron tunnelling from the deep centre to the conduction band presented in [16] is used:

$$W = \frac{eE}{(8m^*\varepsilon_T)^{1/2}}\left[\left(1+\gamma^2\right)^{1/2}-\gamma\right]^{1/2}\left[1+\gamma^2\right]^{-1/4}\exp\left\{-\frac{4}{3}\frac{(2m^*)^{1/2}}{eE\hbar}\varepsilon_T^{3/2}\left[\left(1+\gamma^2\right)^{1/2}-\gamma\right]^2\left[\left(1+\gamma^2\right)^{1/2}+\frac{1}{2}\gamma\right]\right\}, \quad (1)$$

where

$$\gamma = \frac{(2m^*)^{1/2}\Gamma^2}{8e\hbar E \varepsilon_T^{1/2}}. \quad (2)$$

Here $\Gamma^2 = 8a(\hbar\omega)^2(2n+1)$ is the width of the centre absorption band, $n = [\exp(\hbar\omega/(k_B T)) - 1]^{-1}$, where $\hbar\omega$ is the phonon energy, ε_T is the energetic depth of the centre and a is the electron-phonon interaction constant ($a = \Gamma_0^2/8(\hbar\omega)^2$).

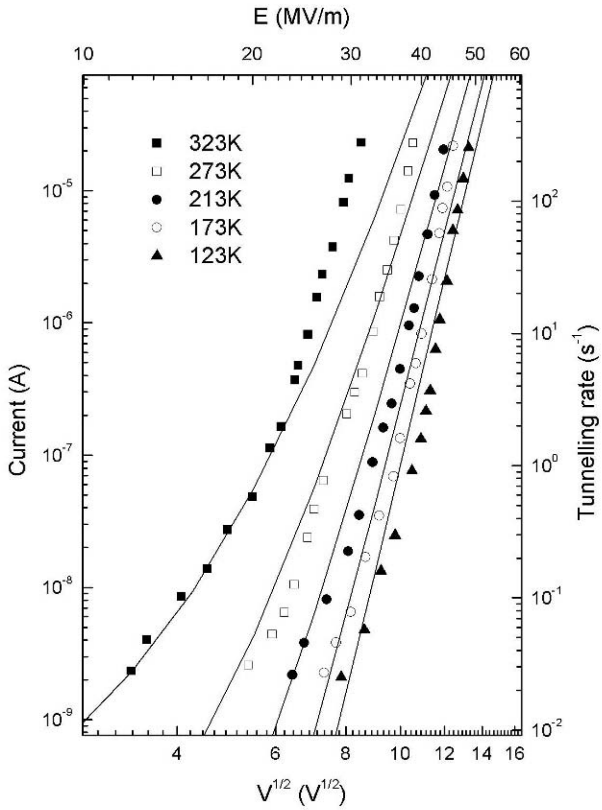

Figure 1. Current-voltage characteristics at different temperatures for p-Si/Ta_2O_5/Al (for negative Al electrode) (symbols) and theoretical dependences $W(E,T)$ (solid lines) calculated using Eq. (1). The calculation have been carried out for the parameters: ε_T=0.9 eV, phonon energy $\hbar\omega$ = 43 meV, electron effective mass m^*= 0.1 m_e, electron-phonon interaction constant a= 4.2. The estimated $N_s \approx 2{\cdot}10^{13}$ cm^{-2}.

The comparison of the $I-V$ data measured at different temperatures for both polarities of the Al electrode with theoretical $W(E,T)$ dependences are shown in Figs. 1 and 2. The calculation was carried out using an electron effective mass of 0.1 m_e [17]. For the phonon energy, a value of 43 meV was used, for ε_T the activation energy of 0.9 eV was taken in the negative Al electrode case and 1.05 eV for the positive Al electrode. The electron-phonon

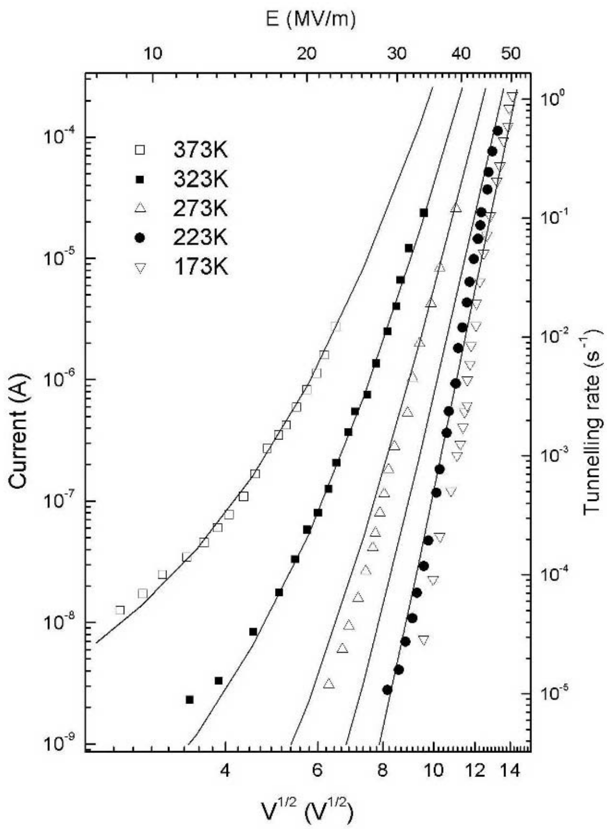

Figure 2. The same as in Fig. 1 for positive Al potential (symbols) fitted to theoretical dependences $W(E,T)$ (solid lines) calculated using Eq. (1). The fitting parameters: ε_T= 1.05 eV, phonon energy $\hbar\omega$ = 43 meV, electron effective mass m^*= 0.1m_e, electron-phonon interaction constant a= 4.5.

coupling constant a was chosen so as to get the best fit of the experimental data with calculated dependences, on the assumption that field strength for tunnelling is proportional to the square root of the applied voltage. In Figs. 1 and 2 one can see that theoretical curves in the entire measured temperature range describe the experimental data quite well. However, discrepancy between theory and experiment is observed, especially at higher temperatures. Namely, at a temperature of 323 K and higher voltage the slope of $I-V$ data is larger than the theoretical ln$W$ vs ln$E$ dependence. The transition from a slope of 2.4 in a log$I$ vs $E^{1/2}$ plot to 4.8 at higher field strength has been observed for Ta_2O_5 films [3] and such behaviour was attributed to the influence of adsorbed water. The density of states in the interface N_S estimated from the comparison was found to be equal about 10^{13} cm^{-2}.

The comparison of the current temperature dependences with the theory, presented in Fig. 3, also shows a good

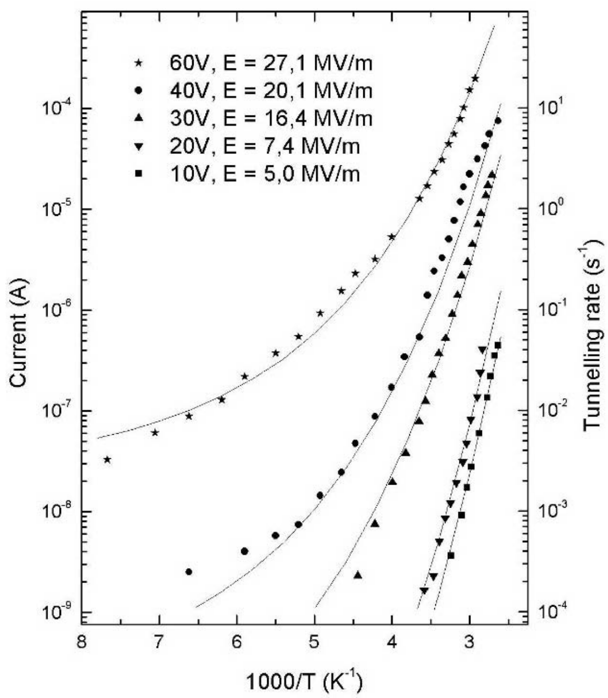

Figure 3. Current-inverse temperature characteristics at different voltages for the same structure as in figure 1 fitted to theoretical $W(E,T)$ dependences vs inverse temperature $1/T$ (solid lines). The fitting parameters are the same as in Fig. 1 ones.

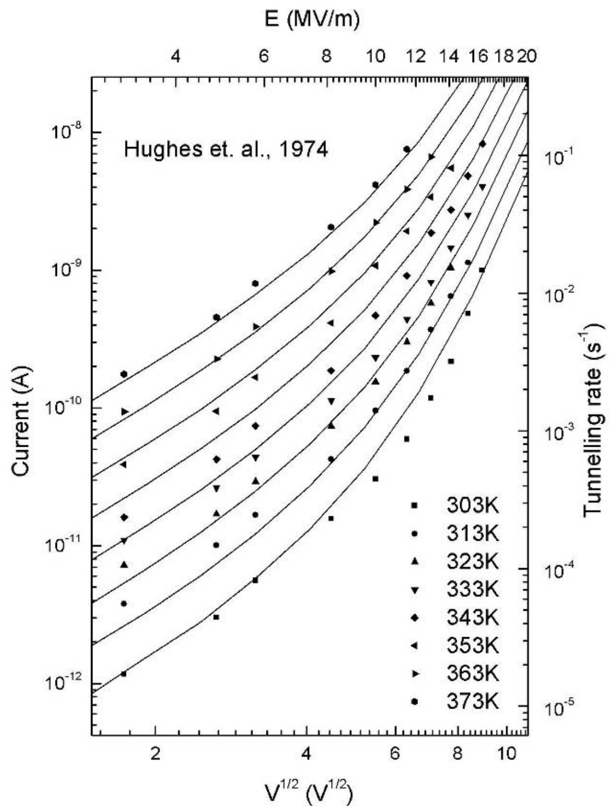

Figure 4. I-V dependences of Si/Ta_2O_5/Al at different temperatures extracted from [2], Fig. 7(a) (symbols) fitted to theoretical $W(E,T)$ vs E dependences (solid lines) computed using the parameters: ε_T= 0.9 eV, $\hbar\omega$ = 43 meV, m^*= 0.1 m_e, and a= 4. $N_s \approx 10^{13}$ cm^{-2}.

agreement of the experimental data with computed curves. A similarly good agreement of the experimental data with the theory is obtained fora comparison of the temperature-dependent $I-V$ data measured by Hughes and Jones [2] in reactively sputtered tantalum oxide thin films. The authors of [2] envisaged a strong resemblance of the conduction to either a FP or the Schottky mechanism, with the latter prevailing. The comparison of the experimental results with theory in Fig. 4 is also done on the assumption that the Schottky barrier exists at the electrode-oxide contact.

Finally, in Fig. 5, the $I-V$ data for Ta_2O_5 thin films obtained by Lee *et al.* [9] using an anodizing method, fitted with computed dependences, are displayed. The authors of [9] identified two types of conduction in the Ta_2O_5 films, i.e. Schottky emission at low electric fields below 2.2 MV/cm and the Poole-Frenkel emission at high fields above 2.2 MV/cm. As is seen in Fig. 5 the computed $W(E)$ describes the experimental data well for the entire range of fields.

4. Conclusion

In conclusion, the phonon-assisted tunnelling model, as we have shown, describes the temperature-variation of the $I-V$ data and peculiarities of current dependence on temperature in Ta_2O_5 films - with a unique set of parameters characterizing the material, such as effective mass of the electron, the phonon energy, as well as the electron-phonon coupling constant. The problem of the curved behaviour of the $\ln I$ vs $1/T$ plots obtained at higher fields falls away as well. The fit of the experimental data with computed tunnelling rate allows an estimate of the field strength at which the free charge carriers are generated, and the density of charged states near the interface between metal and dielectric. Thus, the phonon-assisted tunnelling mechanism must be taken into account in explaining the leakage current characteristics for thin films of Ta_2O_5. The PF/Schottky mechanisms, due to their classical physics nature, are strongly limited in a certain range of temperature and field strength.

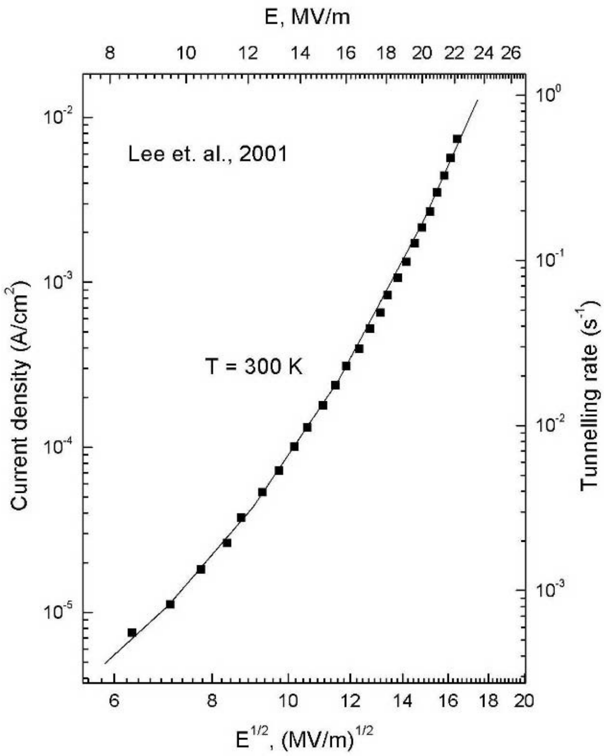

Figure 5. Current density J vs E_{ex} for the Ta_2O_5 films extracted from [9], Fig. 7 (symbols) fitted to theoretical $W(E,T)$ vs E dependences (solid lines) computed using the parameters: ε_T= 0.9 eV, $\hbar\omega$ = 43 meV, m= 0.1 m_e, a= 4. $N_s \approx 10^{13}$ cm^{-2}.

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