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THE THERMALLY STIMULATED CURRENTS IN THIN-FILM STRUCTURES Al—SiO_x—Al WITH DIELECTRIC OF HIGH DEFECT CONCENTRATION

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The maxima of thermally stimulated currents for thin SiO_x films deposited in "sandwich" structure Al—SiO_x—Al were investigated. The I—T characteristics were measured as function of heating rate B, biasing temperature T_b, bias voltage V_b, biasing time t_b and collecting voltage V_c. The optimal parameters for revealing of thermostimulated peaks were determined. From the increasing slope of peaks the activation energy was calculated. The results were compared with those obtained by Servini, Jonscher (*Thin Solid Films* **3**, 341 (1969)) and Berkovich, Gorokhovatskii (*Izv. Akad. Nauk Latv. SSR* **2**, 45 (1971)).

1. *Introduction*

The investigation of current-voltage characteristics at the constant temperature $(I-U)_T$ is one of the ways which may be used for conductor characterization. The supplementary characteristic, which is peculiarly useful for dielectrical or weak conducting systems, is the current-temperature characteristic at constant voltage $(I-T)_U$. This characteristic defines a thermal deliverance of accumulated charge or polarization in unconducting materials. This method is known as thermostimulated current method or method of thermoluminescence. Preliminary excitation is the fundamental condition for the thermostimulated peaks to be obtained.

This method [1, 3] is based on the bias voltage V_b application at defined biasing temperature T_b. Next, the temperature is reduced to some low temperature T_0 and all the time the bias voltage is being applied. At the temperature T_0 the current becomes negligibly small. The collecting voltage V_c, which is different from V_b, is applied at this temperature T_0 and then the temperature is uniformly raised with heating rate $B = dT/dt$ and the current as a function of temperature is registered [1]. One or more peaks appear, which correspond to the deliverance of trapped charge or polarization. Suitable activation energies are connected with the temperatures of peaks maxima and their half-widths [2].

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The $I-T$ characteristics are strongly dependent on the bias voltage V_b, biasing temperature T_b, biasing time t_b, collecting voltage V_c, and heating rates B. Usually, $I-T$ characteristics are presented as a function of one of these parameters, while the other parameters are constant.

In present work this method has been applied to the analysis of Al—SiO$_x$—Al system properties. The current-voltage characteristics for these structures were presented in our earlier works [5, 10].

2. *Experimental*

Thin film structures Al—SiO$_x$—Al in "sandwich" configuration have been deposited on the cleaved mica and BK-7 glass substrates, in vacuum of the order 10^{-5} torr from tungsten crucible. The temperatures of substrates have been 20°C or 150°C. The thicknesses of SiO$_x$ films measured by interference method were about 0.5 μm. The rate of deposition was about 20 Å/s. The areas of active surfaces of investigated structures were smaller than 1.0×10^{-2} cm^2. The silicon monoxide of Balzers (SiO 99.8%) was the starting material for preparing SiO$_x$ films. The exact parameters of the preparation of films have been presented in Ref. [10].

The thermostimulated currents have been measured in vacuum of 10^{-5} torr. For measurements the sample was placed on the copper basis in a metallic dewar which was mounted directly on diffusion pump. The sample was cooled with liquid nitrogen to the temperature 80 K. The resistance heater was regulated with autotransformer. The electric motor with suitable mechanical gear changed the voltage of autotransformer. The velocity of autotransformer slider shift was such that the rise of temperature was linear in the defined range.

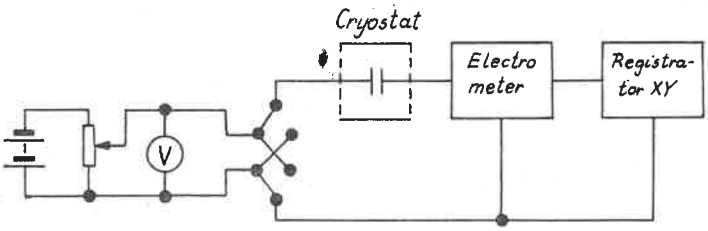

Fig. 1. The idea-scheme of the system utilized in measurements of $I-T$ characteristics

The current-temperature characteristics were measured in the system which idea-scheme is presented in Fig. 1. The correct grounding of individual branches of the system was very significant during the measurements.

3. *Results*

For investigated structures Al—SiO$_x$—Al, the peaks of stimulated-dielectric-relaxation currents (SDRC) have been determined. The influences of heating rate B, biasing temperature T_b, biasing voltage V_b, biasing time t_b and collecting voltage V_c on the value and position of these peaks have been studied.

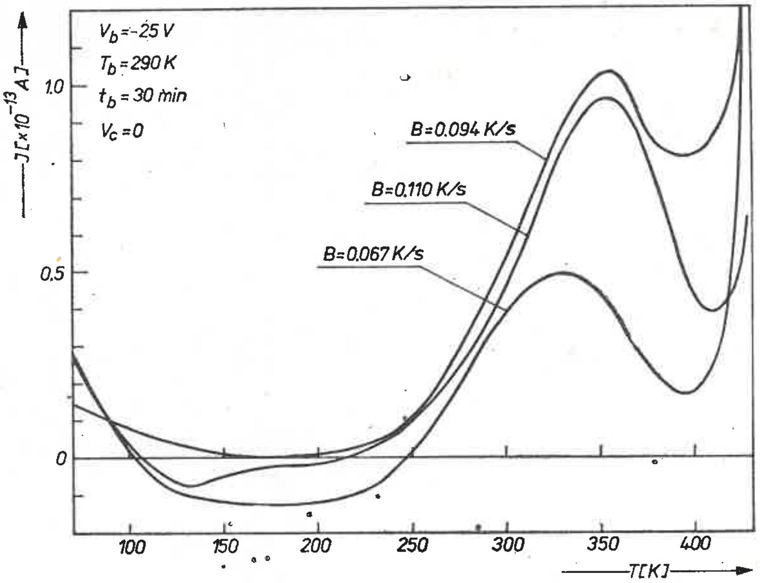

Fig. 2. $I-T$ characteristics as a function of heating rate, B

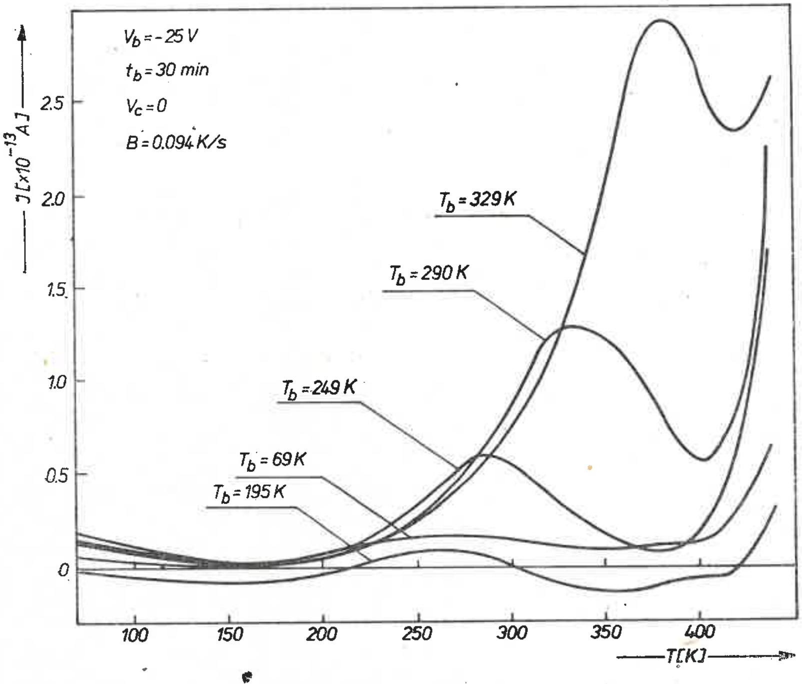

Fig. 3. $I-T$ characteristics for different biasing temperatures, T_b

The $I-T$ characteristics have been investigated for three different heating rates: 0.067 K/s, 0.094 K/s and 0.11 K/s. Results are presented in Fig. 2. It is seen that the highest peak is derived for the heating rate 0.094 K/s.

The biasing temperature T_b has also influenced the increase of peaks. This dependence is presented in Fig. 3. The increase of the current peak with increasing temperature T_b has been observed over the whole investigated range of temperatures.

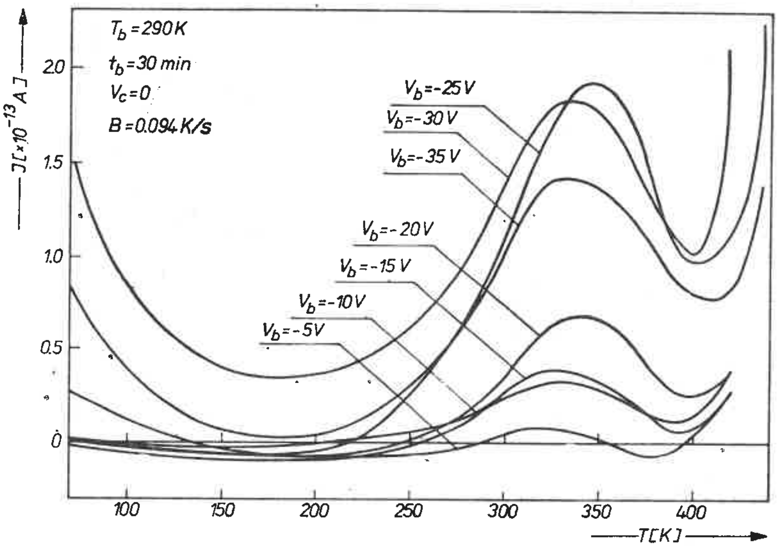

Fig. 4. The dependence of $I-T$ characteristics on bias voltage, V_b

If it is going about the influence of biasing voltage, the optimal voltage, $V_b = -25$ V, is observed. For this value the peak becomes the highest one. This dependence is presented in Fig. 4.

Likewise for the case of biasing temperature, the increase of current peak with increasing biasing time has been also observed for the all investigated ranges of time (Fig. 5).

The current peaks as a function of collecting voltage are presented in Figs. 6 and 7. When the negative or positive collecting voltage is applied (relative to the negative biasing voltage) then the monotonic increase of current is observed above the temperature of peak maximum T_m which disguise the peak of SDRC. The direction of current flow was changed together with the change of biasing voltage peak. This is visible from the diagram, presented in Fig. 6.

The SDRC peaks for structures on the mica and glass substrates have been presented in Fig. 7.

The optimal parameters for revealing the current peaks have been determined as: $V_b = -25$ V, $B = 0.094$ K/s, $T_b = 290$ K, $t_b = 30$ min, $V_c = 0$. The SDRC peak which was obtained for optimal parameters was compared with those obtained by Servini and Jonscher [3] and Berkovich and Gorokhovatskii [4]. This comparison is presented

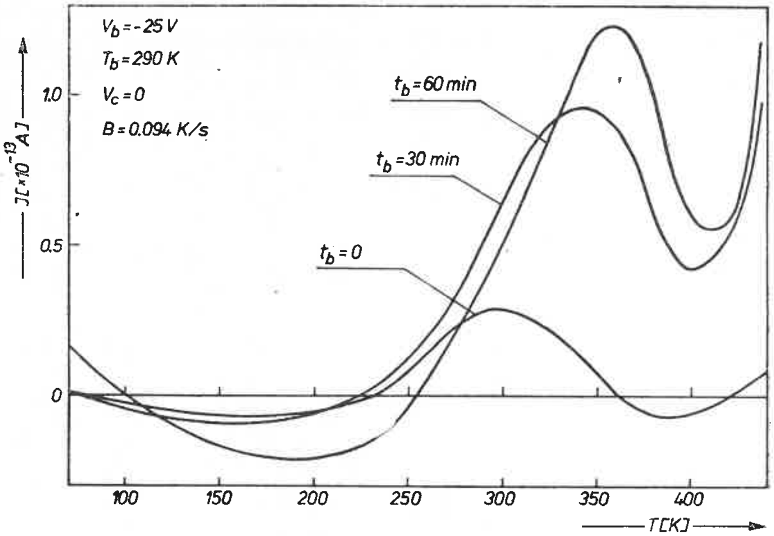

Fig. 5. The influence of biasing time, t_b, on $I-T$ characteristics

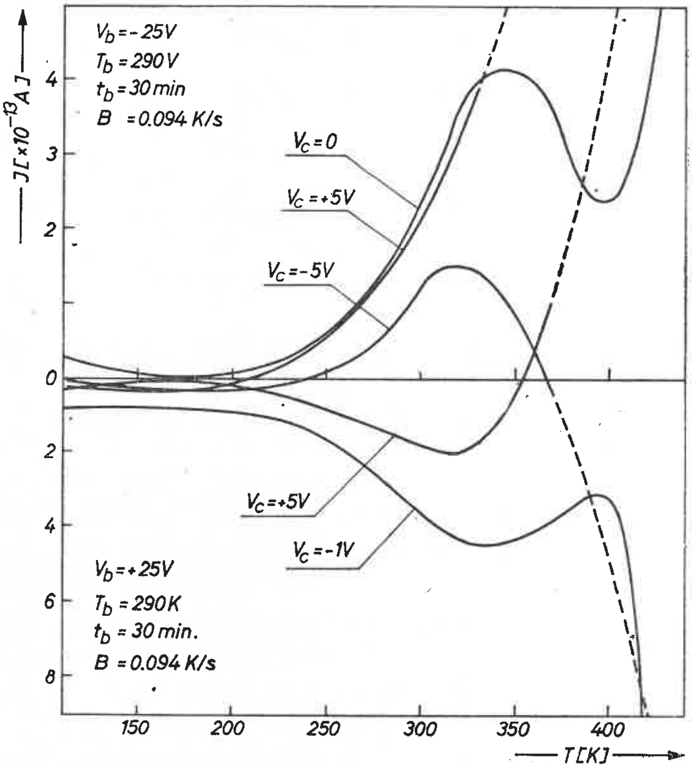

Fig. 6. $I-T$ characteristics as a function of collecting voltage, V_c, for negative and positive bias voltage V_b

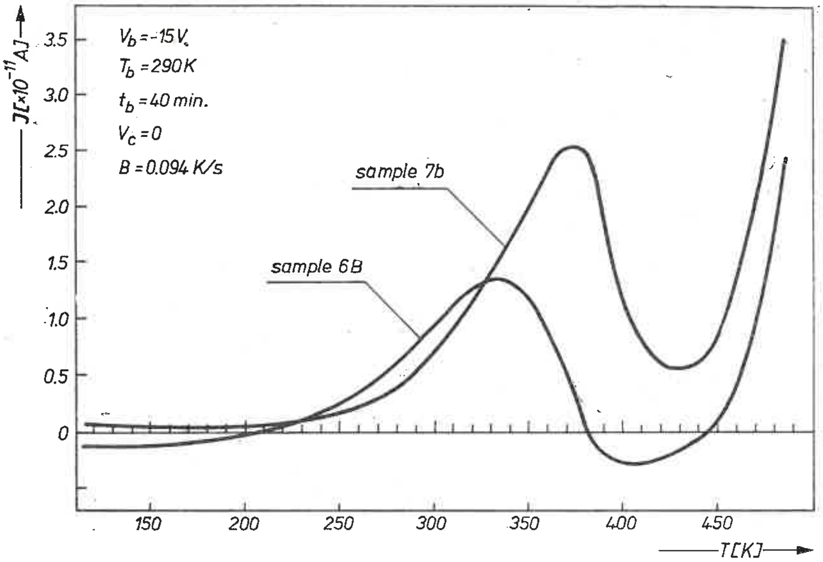

Fig. 7. $I-T$ characteristics for structures deposited on mica (6b) and glass (7b) substrates

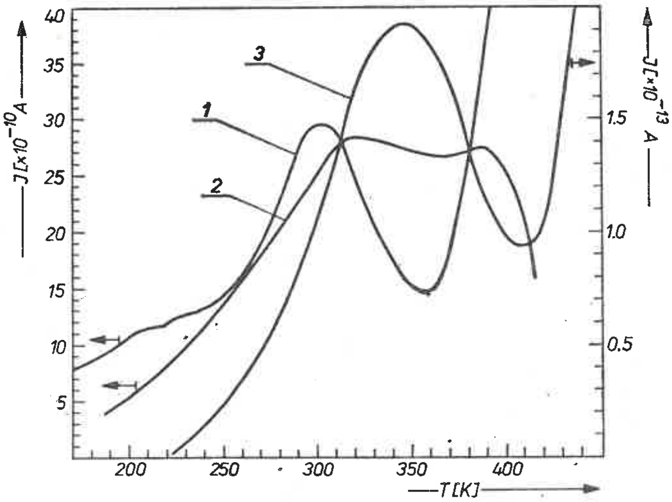

Fig. 8. Comparison of current peaks: the left-hand scale *1* — Servini, Jonscher [3] and *2* — Berkovich, Gorokhovatskii [4]; the right-hand scale *3* — on the base of result obtained in present work

in Fig. 8. Satisfactory reproducibility of $I-T$ characteristics can be obtained only for the defined technology of deposition at given laboratory conditions.

The ln I as a function of $1000/T$ has been derived from the increasing branch of SDRC peaks (Fig. 9). The activation energies 0.25 eV, 0.22 eV and 0.20 eV have been calculated on this base. The reproducibility of the $(I-T)_U$ characteristics was good, it changed for the samples obtained in different evaporation cycles.

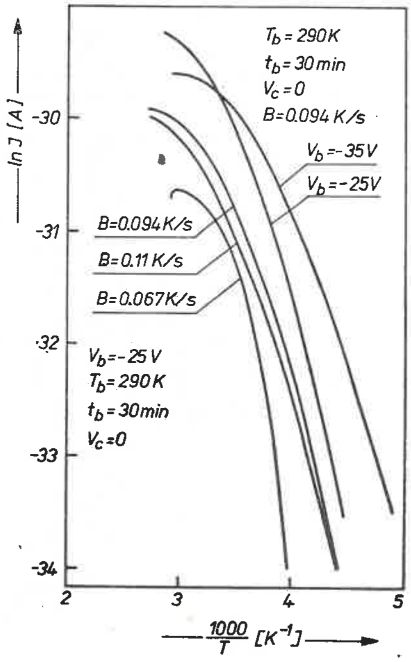

Fig. 9. Dependence of ln I on the reciprocal temperature for the increasing slope of peaks

4. *Discussion*

The results presented refer to amorphous films. In investigated "sandwich" structures Al—SiO$_x$ interface was strongly defected. The principal cause of this was the oxidation of aluminium electrodes during the masks exchange. The strongly defected interface has influenced the thermostimulated currents more than the current-voltage characteristics ([5,10] and also [3,4]). Long-term establishment of current which was observed, suggests that the dielectric is initially in the unstable state [5,6, 10]. Assuming that electrons are the current carriers and that the time of electron staying in a trap depends on its depth and on temperature, this phenomenon is due to the trapping of electrons [7]. The formulas which define the dielectric relaxation time refer usually to dielectric with one precisely defined trapping level. For investigated SiO$_x$ films such a level is absent. In amorphous materials, to which SiO$_x$ belongs, deeply lying localized states are responsible for trapping [8].

For equilibrium state at given temperature there is a trapping level with the greatest number of captured electrons. Such a state is established as a result of overlapping of two effects: decrease of localized states density when the distance from the conduction band bottom increases, and increase of the life-time of an electron in a given state when the depth of this state increases.

In Fig. 10 the proposed energetic model of metal-dielectric contact is presented. The height of potential barrier and the depth of trapping levels, which are denoted on the diagram, have been derived from the analysis of current-voltage characteristics [5, 10] and from the increasing branch of SDRC peak (Fig. 9).

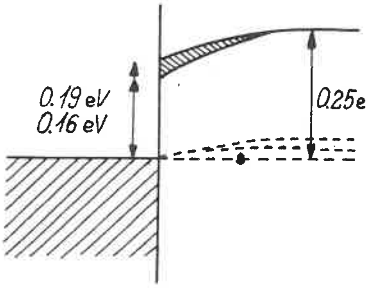

Fig. 10. Energy diagram of metal-insulator contacts

The theoretical $I-T$ characteristic is determined over the whole range of temperatures by [9]

$$I(T) = A(J_{\mathrm{SDRC}}+J_{\mathrm{RS}}), \tag{1}$$

where J_{SDRC} is the initial SDR current in empty cathodic domain, J_{RS} — Richardson-Schottky current, A — the surface of investigated structures.

SDR current can be expressed exactly only for the case of precisely defined non-interacting trapping levels and is given by [9]

$$I(T) = q\mu N_c(V_i/L)A \exp\{-[E_l-\beta(V_i/L)^{1/2}]/kT\}, \tag{2}$$

where q is the electron charge, μ — mobility of electrons, N_c — effective density of states in the conduction band, V_i/L — electric field inside the insulator, E_l — depth of trapping level below the conduction band, $\beta = (q^3/\pi\varepsilon)^{1/2}$ — Richardson-Schottky coefficient, T — temperature. With respect to the investigated structures this expression is valid for orientation only.

During the heating process, the $I-T$ characteristic is described by J_{SDRC} current for low temperatures. For higher temperatures the dominating role is played by J_{RS} current. As the Richardson-Schottky barrier is low (0.14—0.19 eV [5, 10]) due to high concentration of defects at interface, J_{RS} current starts to dominate at room temperature. For the deliverance of current carriers when trapped, 0.25 eV energy is needed which corresponds to higher temperature than that which is necessary for $R-S$ barrier overcoming. This is the reason for which all the $I-T$ characteristics with non-zero collecting voltage give monotonically increasing current without appearance of SDRC peaks (Fig. 6).

When collecting voltage is zero, the observed peaks are due mainly to the dielectric relaxation (transition of the system from one stable state to the other through the unstable state). Forming of stable state after applying of the biasing voltage, V_b, accompanied by the positive spatial charge which is due to the emptying of trapping levels. Depending on temperature and time the maximum of density of empty traps is nearer or further from the bottom of conduction band. Such a dependence can take place only for the case of energetic model of amorphous dielectric with tails of localized states [8]. In this model the distribution of trapping levels is continuous (Fig. 11) and the number of electrons captured at the given trapping level is a function of time and temperature. When the biasing temperature, T_b, increases then the increase and shift of a peak toward higher temperatures can be observed (Fig. 3). So, for lower biasing temperature, T_b, the traps of lower energy are emptied and for higher T_b — those of higher energy. Similar shift of peaks is observed for different biasing times, t_b, (Fig. 5). This can be explained as being due to the increase of time of electrons staying in traps proportionally to their depth. So, discussed $I-T$ characteristics as a function of T_b and t_b correspond phenomenologically to the proposed model.

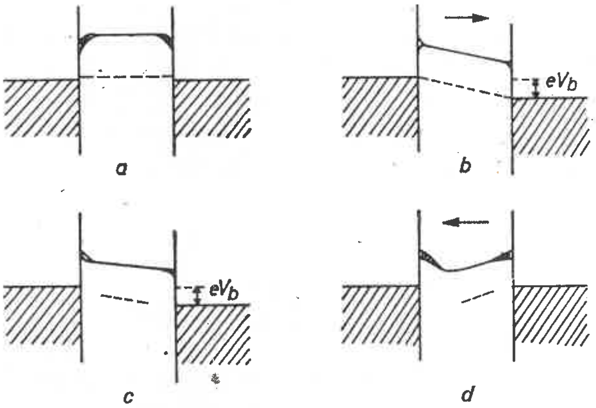

Fig. 11. Energy diagram of Al—SiO$_x$—Al structures for different states of dielectric relaxation: a — before bias voltage V_b applicated — steady-state; b — immediately after bias voltage V_b applicated — unsteady--state; c — new steady-state which was established; d — at low temperature immediately before the starting of heating — broken-down electrodes. The empty traps has been marked on the diagram. Arrows indicate the conventional direction of current flow

Applied biasing voltage (V_b) does not shift the peak but influence on its rise (Fig. 4), for together with the increase of V_b the width of empty cathodic domain increases too. The biasing voltage $V_b \simeq -25$ V for which the maximal SDRC peak is observed can be assumed to exist. This is equivalent to the saturation of positive charge collected in investigated structure. Such a high voltage of saturation cannot be explained in terms of surface effects only. It seems to be reasonable that the regions which are far from electrodes take part in the processes of capturing and deliverance of electrons. The lack of peak shift as a function of biasing voltage can suggest that localized levels are similar, at least to some distance from electrodes. This distance can be determined by the propagation of positive charge in cathodic domain, but the determination of this on the basis of the obtained results is impossible. Technological conditions indicate that the structure should be non-symmetrical in spite of the fact that the electrodes were made of the same metal (Al). This asymmetry should be introduced by the layer of oxidized aluminium due to infection of the bowl during the exchange of mask. However, for the $I-T$ characteristics derived for positive and negative biasing voltage no difference in the shape of SDRC peaks was observed (Figs. 6 and 7). The symmetry of investigated structures and low contact barriers justify the lack of second peak connected with transition from unstable to stable state.

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