

Role of Carrier Injection and Localization in Degradation of Oxide Films

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Most current electronic and electrochemical devices are stacks of thin films and interfaces operating under electrical stress. Nanometre-thick oxide films play crucial role in performance of these devices. Injection of excess electrons and holes into oxide films is responsible for the mechanisms that govern the formation of conductive filaments in resistance switching memory devices, the dielectric breakdown in microelectronic devices, and the performance of photo-electrochemical and oxide fuel cells. Our theoretical modelling combined with experimental observations demonstrates that structural disorder in amorphous SiO₂, Al₂O₃, TiO₂, ZnO and HfO₂ films creates precursor sites which can spontaneously trap up to two electrons or holes in deep states in the band gap. The results demonstrate that single- and bi-polaron electron and hole states can form in a- SiO₂ [1] and a- HfO₂ [2] where the effect of local disorder is amplified by polaronic relaxation of amorphous network. Only hole trapping is found in a-Al₂O₃ [3] The electron localization weakens Me-O bonds, which can be broken upon thermal activation, creating an O²⁻ interstitial ion and a neutral O vacancy [4]. O²⁻ interstitial ions can easily diffuse through the oxide and in devices are guided to the positive electrode by the electric field [5]. Multi-scale modelling [6] including electron injection rates, defect creation and electron hopping rates through created defects is used to describe dielectric breakdown in oxide films. The results demonstrate that the creation and field-driven movement of oxygen ions causes changes in oxide structure on a much larger scale than previously thought.

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[6] A. Padovani et al., *J. Appl. Phys.* **121**, 155101 (2017).