Opto-electronic characterization of electron traps upon forming polymer

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Opto-electronic characterization of electron traps upon forming polymer oxide memory diodes

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Metal-insulator-polymer diodes where the insulator is a thin oxide (Al2O3) layer are electroformed by applying a high bias. The initial stage is reversible and involves trapping of electrons near the oxide/polymer interface. The rate of charge trapping is limited by electron transport through the polymer. Detrapping of charge stored can be accomplished by illuminating with light under short-circuit conditions. The amount of stored charge is determined from the optically induced discharging current transient as a function of applied voltage and oxide thickness. When the charge density exceeds $8 \times 10^{17}/m^2$, an irreversible soft breakdown transition occurs to a non-volatile memory diode.

Non-volatile memories are being developed for data storage applications. A promising candidate is a resistive random access memory. The memory consists of a simple metal-insulator-metal stack based upon organic or inorganic semiconductors and oxide layers or nanoparticles. Diodes comprising as insulator an oxide have to be formed by applying high electric field corresponding to soft breakdown. After forming the diode behaves as a capacitor. The charging of the capacitor can be followed by applying a step voltage. The charging current $(I)$ decays with time $(t)$, following a power law $I(t) = 1/t^\alpha$. Where $\alpha$ is a constant parameter. The inset of Fig. 2 shows the linearity of $\log(I)$ vs $\log(t)$ over four decades with a slope $\alpha \approx 1.06$. An explanation of this remarkable behavior is given below.

Figure 1 illustrates the electroforming of the diode. The bias voltage was swept at a ramp rate of 0.1 V/s up to 10 V and the current density recorded with time. The $J$-$V$ characteristic shows three distinct regions. At applied voltages below 6 V (region I), the current density has the characteristics of a displacement current, approaching an almost constant value depending on the ramp rate. The diode behaves as a capacitor.

Upon reversing the direction of the voltage sweep, the current is negligible (not shown). This implies that the charges are trapped. With impedance spectroscopy or
mental data is shown together with the $J$-$V$ traces are measured in the diode. Once emptied, identical $J$-$V$ traces are observed, hence the trap levels must be deep. This is confirmed by the fact that under short-circuit conditions, it takes more than four orders of magnitude with $\alpha = 1.06$. The experimental data is shown together with the $1/t^n$ and $1/t$ behavior.

In region (I), at bias voltages between 6 V and 10 V, the current rises exponentially with bias and is due to tunneling of electrons through the oxide. In this regime, the $J$-$V$ characteristics are reversible; no hysteresis is observed.

In region III, near 10 V, the current rises steeply and becomes noisy which are both typical characteristics of soft breakdown of the oxide. This process is irreversible; the electrical characteristics have changed permanently. The formed device behaves as a non-volatile memory, as reported previously. The resistance of the diode can now be switched between a low resistive on-state and a high resistance off-state.

The trapped charge in region I can be neutralized by photogenerated charge carriers. Under illumination, transient discharge currents are observed on the time scale of hundreds of seconds. Optical detrapping is only observed for light with photon energies higher than the polymer band gap (3.1 eV) showing that the charge carriers inducing the neutralization are generated in the polymer. Furthermore, detrapping transients are faster for higher optical power, in agreement with previous findings using quasi-static capacitance-voltage techniques. The oxide thickness was varied. The resistance of the diode can only be switched between a low resistive on-state and a high resistance off-state.

The inset in Fig. 4(b) shows the total amount of charge released upon illumination as a function of the maximum applied charging voltage in the preceding charging $J$-$V$ sweep. The amount of charge released depends linearly on the applied maximum charging voltage. Extrapolation to the voltage necessary to fully electroform the device (10 V, see Fig. 1) predicts that a total amount of $8 \times 10^{17}$ m$^{-2}$ charges must be stored in the device before soft-breakdown occurs and memory characteristics are induced. This density of trapped charge ($\rho_t$) corresponds to a critical electric field strength for electroforming ($\rho_t/\varepsilon_{\text{poly}}$) of $1.5 \times 10^{9}$ V/m, which is close to the field strength required for electrical breakdown of Al$_2$O$_3$ ($10^9$ V/m). Furthermore, $\rho_t$ is in agreement with previous findings using quasi-static capacitance-voltage techniques. The oxide thickness was varied. The amount of stored charge when applying the same bias voltage for charging varies with oxide thickness. Yet, the amount of charge needed to reach the critical electrical field strength for electroforming remained the same.

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\[ I(t) = \frac{1}{r} \left( \frac{1}{n \rho C} t + \left( \frac{1}{V_0} \right)^n \right)^{1+n} \]

for \( t \gg \left( rC/nV_0^n \right) \),

where, \( V_0 \) is the initial voltage drop over the polymer. From the experiment, we obtain a power-law dependence of the current \( I \) on time with an exponent of 1.06. We fabricated electron-only diodes of the present poly(spirofluorene) and obtained superlinear \( J-V \) characteristics following Eq. (2) with exponent \( n \) around 5. This is in good agreement with the exponent \( x = 1.06 \) from time domain measurements as described by Eq. (3).

If the electric field across the oxide is increased, hole injection from the aluminum anode into the oxide will be favored. Hole trapping in the oxide encourages further accumulation of electrons at the interface, creating a layer of increasing polarization which eventually causes soft-breakdown across the oxide. We suggest that this process is responsible for the oxide electroforming.

In summary, we have determined density and spatial location of electron traps in polymer-oxide memories employing an electro-optic method. The trapped charges establish an electric field across the oxide. When this field reaches a critical value, soft breakdown occurs, yielding an active memory diode.

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