

Ascorbate Detection Using Single-Trap Phenomena in Two-Layer Si NW FETs

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Abstract—Biosensing and detection of various biological and chemical species are extremely important for the diagnosis of different diseases. Nowadays, liquid-gated silicon nanowire field-effect transistors (Si NW FETs) are attracting considerable attention as powerful sensing transducers. We have recently designed and fabricated novel silicon nanowire structures that consist of two silicon layers with different doping concentrations and applied them for the detection of ascorbate biomolecules in electrolyte solutions. The response of the sensor to different concentrations of target molecules was revealed by monitoring the changes in both the drain current of the transistor and the capture time as a characteristic of a single trap responsible for a random telegraph signal (RTS) noise of the device. The results demonstrated that changes in the surface potential introduced by ascorbate molecules are registered with enhanced sensitivity in the case of a single-trap-based biosensing approach compared to the standard drain current approach. Here we report that single-trap phenomena in fabricated two-layer structures are favorable for biosensing applications and can be effectively used as a highly sensitive tool for monitoring such antioxidant molecules as ascorbate in order to measure oxidative stress in chemical, biochemical, and biological systems.

Keywords—Nanowire biosensors, single trap, low-frequency noise, ascorbate molecules, oxidative stress.

I. INTRODUCTION

Research into and development of new devices, as well as new methods enabling a direct, sensitive, and rapid analysis of biological and chemical substances, are essential for both theoretical studies and practical applications. Nowadays, among a large variety of biosensing platforms, silicon nanowire field-effect transistors (NW FETs) are considered as excellent candidates for biosensing from the viewpoint of the development of label-free, high-speed and ultrasensitive FET-based biosensors [1-3]. Although the utilization of NW FETs as biochemical transducers offers considerable advantages such as mass fabrication and biocompatibility, there is still considerable debate concerning both the best transducer configuration and approach for the detection of different target bio-objects, e.g. ions, proteins, DNA molecules, viruses. To address these problems, the noise level of the devices has to be considered, which usually affects the sensitivity of the sensors and defines their detection limit.

Typically, the physical sources of noise in large-area devices originate from fluctuations of microscopic entities, such as mobility or number of charge carriers, due to their interactions with different traps randomly distributed in a gate insulator layer [4, 5]. However, with a scaling down of small devices the noise might be determined by a single carrier

phenomenon [5, 6]. In particular, a small device can be fabricated with characteristic sizes below 100 nm, where a point defect (a single trap in a nanotransistor gate dielectric layer) can exchange charge carriers with the conductive channel of a nanotransistor. In this case, a two-level discretized fluctuation signal known as random telegraph signal (RTS) noise can be observed [5-7]. Being a fully stochastic process, RTS noise can be characterized by its main parameters, i.e., capture (τ_c) and emission (τ_e) time constants as well as RTS amplitude, e.g. a change of the signal when a transition event occurs. At the same time, the capture time constant indicates the average time during which the trap is empty, while the emission time constant reflects the time during which the trap is occupied by a charge carrier.

Usually, an RTS phenomenon in a nanoscale device is treated as noise affecting its performance. However, due to the discrete nature of the phenomenon and its specific properties, the phenomenon provides a significant opportunity for practical applications including biosensing [6, 7]. Recently, in order to achieve a better single-trap switching kinetic (more readily controllable capture/emission time characteristics of a single trap) we fabricated a new type of silicon nanowire consisting of two silicon layers with different concentrations of dopants: the first layer is low-doped silicon with an impurity concentration of 10^{15} cm^{-3} , while the second layer is highly-doped silicon with an impurity concentration of 10^{17} cm^{-3} .

In this study, we show that the fabricated devices indeed display pronounced RTS noise with advanced characteristics favorable for biosensing. We apply such two-layer (TL) nanowire sensors for the detection of ascorbate molecules in buffer solutions and demonstrate that nanotransistor sensors exploiting single-trap phenomena allow different concentrations of target molecules, i.e. ascorbate molecules, to be detected with enhanced sensitivity. It should be noted that no attempts to measure changes in ascorbate concentration using nanowire-based sensors have yet been published.

II. EXPERIMENTAL METHODS

A. Fabrication procedure

The structures under study are silicon two-layer nanowire FETs with p-type conductivity configured as accumulation mode transistors. Nanowire structures were fabricated using a CMOS-compatible top-down approach. The fabrication process was based on silicon-on-insulator (SOI) wafers with a 50 nm thick p-type silicon layer and a 145 nm thick buried oxide layer (BOX). A second p-type silicon layer with a boron concentration of 10^{17} cm^{-3} was grown epitaxially using the

chemical vapor deposition method on top of the active silicon layer of SOI wafers with a boron concentration of 10^{15} cm^{-3} . Single nanowire structures were defined using e-beam lithography and patterned using tetramethylammonium hydroxide (TMAH) wet chemical etching in order to provide atomically flat surfaces as well as reduced defect density of the nanowires. Source and drain contacts were highly implanted with boron dopants resulting in p^+-p-p^+ transistor structures. An 8 nm thick SiO_2 layer was grown thermally in order to protect the silicon nanowires and to serve as a gate dielectric layer. Low-resistive 200 nm thick contact feedlines were formed by thermal evaporation followed by a lift-off process and high-temperature annealing in order to achieve an ohmic contact. Finally, samples were passivated with a polyimide layer in order to prevent current flow between metal leads and liquid during measurements. Small windows allowing a gating solution to exclusively access the nanowire area were opened by means of photolithography. All fabrication processes were performed at the Helmholtz Nano Facility of Forschungszentrum Jülich.

B. Experimental setup

A schematic illustration of the fabricated structure including the measurement configuration is shown in Fig. 1a. The PDMS microchannel constructed in our laboratory was used for liquid delivery. An Ag/AgCl reference electrode was inserted into the outlet tube close to the microchannel in order to avoid any pollution of the gating solutions flowing through the microfluidic channel. Both current-voltage characterization and noise measurements were performed in order to characterize the nanowire FETs.

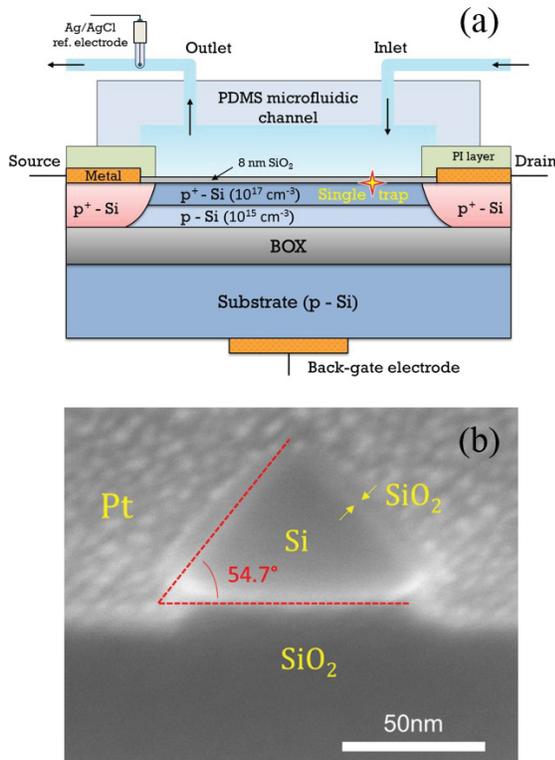


Fig. 1. (a) A schematic view of a Si TL NW FET with a microfluidic channel (cross section along the nanowire). (b) SEM micrograph of the fabricated single silicon nanowire cross section obtained using focused ion beam milling.

Current-voltage characteristics of the nanostructures were measured using Keithley 2400 and 2430 current/voltage measurement units providing an accurate measurement of both the drain-source and the gate-source currents with a resolution of less than 10 pA. Low-frequency noise characteristics were measured using a fully automated ultralow noise measurement setup. A detailed description of our measurement setup is presented elsewhere [7]. The noise data collection was performed using a U2542A data acquisition module. A fast Fourier transform method was then applied to the measured data in order to translate time-dependent source-drain voltage fluctuations into a voltage power spectral density (PSD) S_V . In order to achieve reliable noise characteristics of the samples under study, all electrical measurements were performed in a custom-built Faraday cage to protect against any external electromagnetic fields.

III. RESULTS AND DISCUSSION

A. I - V and noise characterization

In the present study, we discuss the sensing results obtained at room temperature for a 70 nm wide and 400 nm long two-layer silicon nanowire with a triangular cross-section as shown in Fig. 1b. Transfer and output curves measured for the same liquid-gated nanowire transistor are shown in Fig. 2a and Fig. 2b, respectively. A phosphate-buffered saline (PBS) solution with $\text{pH} = 7.4$ was used as the gating solution in this case. It should be noted that the nanowire demonstrated typical p-type FET behavior with a subthreshold swing of 100 mV/decade (see Fig. 2a). At the same time, the leakage current through the 8nm thick SiO_2 gate dielectric layer was negligibly small during the current-voltage and noise measurements.

As can be seen from Fig. 2b, at liquid-gate voltages higher than $V_{LG} = -1.4 \text{ V}$, noticeable fluctuations in the drain current were observed. Such stepwise behavior of the drain current implies the presence of a single trap in the front-gate dielectric layer exchanging the carriers with the conductive channel of the nanowire transistor. In order to investigate the observed fluctuation phenomena, noise spectroscopy was used as a powerful tool for the analysis of device performance.

B. Noise spectroscopy analysis

Fig. 3a illustrates the noise measurement results obtained for the same 70 nm wide and 400 nm long two-layer silicon nanowire structure at different liquid-gate voltages. The drain-source bias was kept constant at -100 mV, which corresponds to a linear working regime of the transistor, and in this case the back-gate electrode was grounded. As can be seen from Fig. 3a, a drain-source voltage noise PSD demonstrates Lorentzian-shaped noise behavior with the characteristic frequency shifting to higher frequencies with increasing liquid-gate voltage. Such noise behavior is typical for nanowire transistors yielding RTS noise [7-9]. At the same time, as can be seen in the inset of Fig. 3a, the drain current of the transistor is indeed affected by RTS noise thus demonstrating switching kinetics between two distinct states (levels): the low level, which corresponds to the case when the trap is occupied, and the high level of the drain current, which corresponds to the state when the trap is empty (a carrier is in the channel).

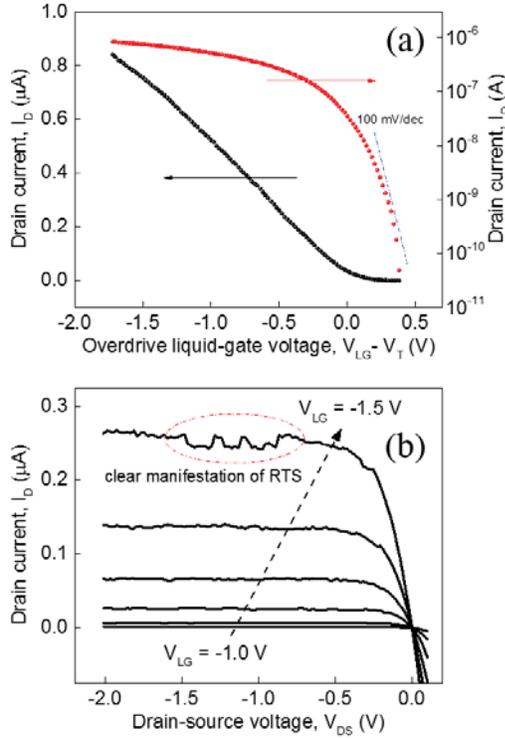


Fig. 2. (a) Transfer curves of a 400 nm long and 70 nm wide Si TL NW FET measured with a liquid gate at a constant V_{DS} of -100 mV (left axis, linear scale; right axis, semi-logarithmic scale). (b) Output characteristic of the same Si TL NW FET measured at different liquid-gate voltages that were varied in the range from -1.0 V to -1.5 V with a step size of -0.1 V.

As was mentioned above, RTS noise is a fully stochastic process that can only be characterized by the average capture and emission time constants. Therefore, in order to estimate RTS characteristic times, a statistical analysis using the amplitude histogram method was performed [7, 10]. The histograms of RTS time traces measured at different liquid-gate voltages are shown in Fig. 3b. Two Gaussian peaks corresponding to the capture and emission states can be clearly resolved. It should be noted that the amplitude of the peaks reflecting a probability of the trap being in a certain state (occupied by a carrier or empty) strongly depends on the V_{LG} applied to the sample. The calculated average capture τ_c and emission time τ_e constants are presented in Fig. 4a as a function of the drain current controlled by the liquid gate. It can be seen that both time constants depend on the drain current, i.e. density of carriers, and demonstrate behavior which deviates from that predicted by the classical Shockley-Read-Hall (SRH) theory. It should be emphasized that similar dependences of RTS noise time constants were observed for different devices fabricated in the same technological run. Note that here we report the results obtained for accumulation mode Si TL NW FETs. The similar characteristics of RTS noise observed for the inversion mode TL NW structures are published in [7]. In the case of the inversion regime, such strong dependences of time constants on the density of carriers can be explained in the framework of the Coulomb blockade energy model [7, 11]. At the same time, an additional energy barrier should be considered in order to explain the dynamics of capture and emission processes in the accumulation working regime [11].

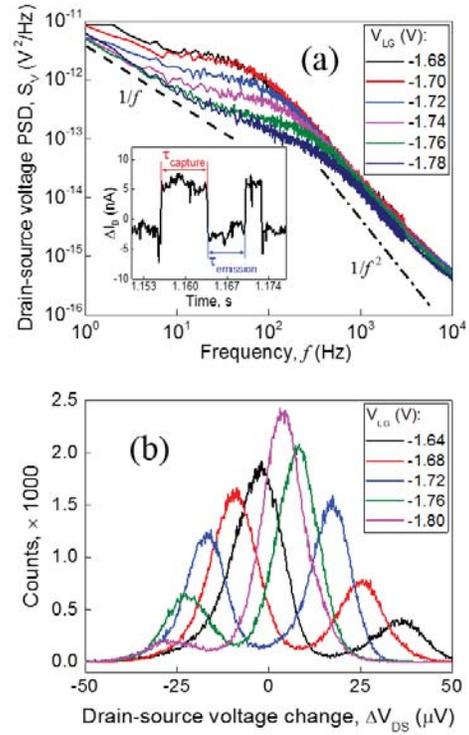


Fig. 3. (a) Drain-source voltage noise PSD measured at different liquid-gate voltages and at a constant drain-source bias of -100 mV (inset: two-level RTS fluctuations measured at $V_{LG} = -1.70$ V). (b) Histograms of measured time traces confirming two-level fluctuations.

Fig. 4b illustrates the probability of the trap being occupied by a single carrier (a hole for the p-type FET) $g = \tau_e / (\tau_e + \tau_c)$ at different liquid-gate voltages. It can be seen that trap occupancy probability is indeed effectively controlled by the liquid-gate implying that the single trap characteristics are also very sensitive to the changes of surface potential. As was mentioned above, RTS noise is usually considered an undesirable effect that affects transistor performance. However, in this work we applied the nanowire transistor which demonstrated single-trap phenomena with advanced characteristics for the detection of different concentrations of ascorbate molecules diluted in the 180 mM PBS solution with initial pH = 7.4. In this respect, the current-voltage as well as the low-frequency noise measurements at different concentrations of ascorbate molecules were performed at a defined working point ($V_{DS} = -100$ mV, $V_{LG} = -1.8$ V).

The results of the sensing experiment are presented in Fig. 5 in term of changes in both the drain current (the standard I - V approach) and the capture time (the single-trap approach) as a characteristic parameter of the single trap. As can be seen from Fig. 5, changes in both parameters were registered with increasing concentration of ascorbate molecules. However, the average capture time constant changes much more strongly in response to the change of the concentration of target molecules compared to the drain current. This result demonstrates that the single-trap approach is indeed favorable for biosensing with enhanced sensitivity using the fabricated TL NW-based sensors.

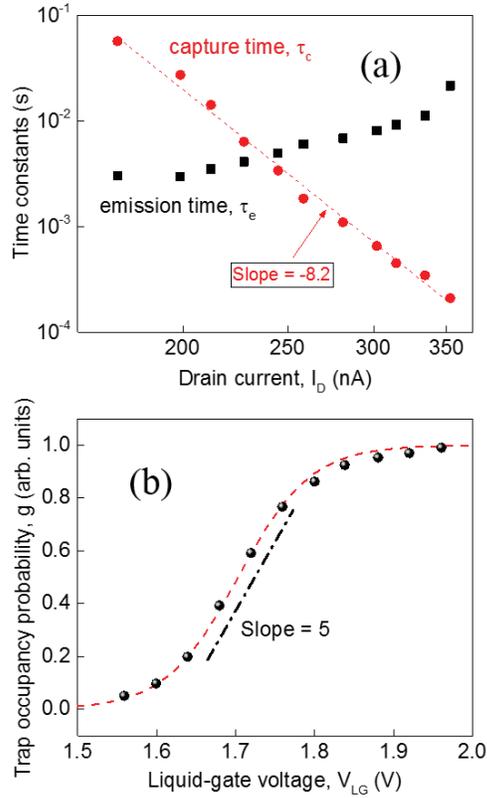


Fig. 4. (a) Capture and emission characteristic time constants as a function of the drain current controlled by the liquid-gate. The dashed red line with slope -8.2 reflects a strong dependence of capture time on the drain current. (b) Trap occupancy probability as a function of the liquid-gate voltage applied. The dashed red line here represents a guide for the eye.

IV. CONCLUSIONS

To conclude, we fabricated liquid-gated silicon two-layer nanowire structures and applied them for the detection of different concentrations of ascorbate molecules. Electrical and transport properties as well as noise phenomena in Si TL NW FETs were investigated utilizing both the current-voltage characterization technique and noise spectroscopy. We demonstrated that applying a single-electron trapping-detraping process in the fabricated sensors allows the detection of ascorbate molecules with enhanced sensitivity. The results shed light on the development of highly sensitive tools for monitoring such important antioxidants molecules as ascorbate.

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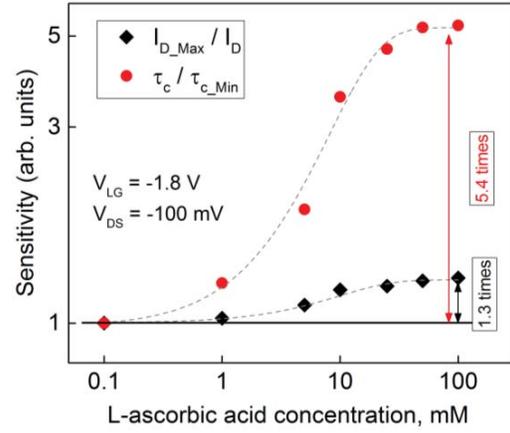


Fig. 5. The response of the Si TL NW FET-based sensor to different concentrations of ascorbate molecules (or L-ascorbic acid) in relation to the sensitivity calculated for both the standard approach (change in the drain current) and the single-trap approach (change in the capture time). The dashed lines represent a guide for the eye.

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