

Proton Irradiation-Induced Electrostatic Modulation in ZnO Nanowire Field-Effect Transistors With Bilayer Gate Dielectric

Gunho Jo, Woong-Ki Hong, Minhyeok Choe, Woojin Park, Yung Ho Kahng, and Takhee Lee

Abstract—We report an efficient method to predictably control the conductance and operation voltage of ZnO nanowire field-effect transistors (FETs) with bilayer polyimide (PI)-SiO₂ gate dielectric by selectively generating oxide-trapped charges via proton beam irradiation. The bilayer gate dielectrics was made by polyimide and thermally grown SiO₂, which prevents negatively charged interface states between the gate dielectric and the ZnO nanowire after proton irradiation. The proton beam-induced charges trapped in the SiO₂ dielectric layer can effectively enhance the electric field toward the n-channel ZnO nanowire, which allows for more accumulation of electrons in the conduction channel of the ZnO nanowire. As a result, the conductance increased and the threshold voltages shifted toward the negative gate bias direction after irradiation. Furthermore, selective modulation of the electrostatic characteristics of the ZnO nanowire FETs was possible by varying the proton irradiation time, which is important for practical application of these devices.

Index Terms—Bilayer gate dielectric, field-effect transistor (FET), proton irradiation, ZnO nanowire.

I. INTRODUCTION

ONE-DIMENSIONAL nanoscale building blocks, such as nanowires and nanotubes, are used for a wide range of nanoelectronic devices because of their novel and fascinating properties [1]–[5]. In particular, semiconducting nanowires have been widely used as the active channel of field-effect transistors (FETs), which are the basic component of several types of nanodevices, including sensors [1], memory devices [6], and logic circuits [3], [7]. For successful application of these devices,

however, it is critical to realize predictable and reproducible control of the electronic properties of nanowire FETs to ensure and enhance the device performance. Therefore, there have been increasing demands for the development of effective methods to precisely control the electrical properties of nanowire FETs. Recently, several researchers have reported that modulation of the electrostatic properties of electronic devices was possible through charge association in the dielectric layers [3], [8]–[11]. For example, Vanheusden *et al.* have demonstrated a nonvolatile memory effect from mobile proton (H⁺) ions introduced into SiO₂ thin films in Si/SiO₂/Si structures [8]. Anghel *et al.* studied the trap-dynamics of photogenerated charges at the dielectric/polymer interface in carbon nanotube transistors [9]. We have also reported the ability to adjust the operating voltage of complementary logic circuits based on hybrid nanodevices composed of p-channel carbon nanotubes and n-channel ZnO nanowire transistors through proton irradiation-generated charges in the dielectric layers [3]. As such, it is important to explore the charge-assisted electrical tunability of nanowire devices to enhance the potential applicability of nanowire-based electronics. We have previously reported a study on the surface band engineering of ZnO nanowires FETs via the irradiation-generated charges in bulk SiO₂ and at the SiO₂/ZnO nanowire interface [11]. The suspended-type ZnO nanowire FETs were fabricated in order to provide substantial evidence of the observed threshold voltage shift in the regular (on-substrate-type) ZnO nanowire FETs, and to enhance the irradiation-induced local electric field by removing the interface states at the SiO₂/ZnO nanowire interface. However, the suspended-type FET devices showed relatively weaker gating effect. Furthermore, the fabrication of the suspended-type devices is relatively complicated and the device yield is also much low. Therefore, to achieve desired device functions for more practical applications, we have searched for an alternative way mimicking the suspended-type nanowire FETs by employing a bilayer gate dielectric device structure. This approach is not specific for nanowires; it should be useful for any other types of transistors based on nanotube, nanoribbon, and thin-film semiconducting channels.

In this study, we report proton irradiation-induced electrostatic modulation in ZnO nanowire FETs with bilayer organic-inorganic gate dielectrics based on spin-cast polyimide (PI) and thermally grown SiO₂, which allow only an additional field effect toward ZnO nanowire FET by preventing trapped charges at the SiO₂/ZnO nanowire interface after proton irradiation. PI dielectrics have attracted interest for merits including robustness to proton beam and chemical solvents (e.g., photoresist and

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G. Jo is with the Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ 08544 USA (e-mail: gunhojo@princeton.edu).

W.-K. Hong is with Korea Basic Science Institute Jeonju Center, Jeonju 561-756, Korea (e-mail: wkh27@kbsi.re.kr).

M. Choe and W. Park are with the Department of Materials and Science Engineering, Gwangju Institute of Science and Technology, Gwangju 500-712, Korea (e-mail: neo314@gist.ac.kr; skki2002@gist.ac.kr).

Y. H. Kahng is with the Research Institute for Solar and Sustainable Energies, Gwangju 500-712, Korea (e-mail: yhkahng@gist.ac.kr).

T. Lee is with the Department of Physics and Astronomy, Seoul National University, Seoul 151-747, Korea (e-mail: tlee@snu.ac.kr).

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acetone) [13]. Oxide-trapped charges induced by ionizing radiation in the bulk SiO₂ dielectric layer can effectively enhance the electric field in the conductive channel of n-type ZnO nanowire FETs, thereby increasing the conductance and shifting the threshold voltage toward the negative gate bias direction. Moreover, as the irradiation time was increased, larger amounts of oxide-trapped charges were induced. This reaction enhances the electric field and results in larger current flows and larger threshold voltage shifts. Therefore, varying the proton irradiation time was demonstrated to be useful for modulating the electrostatic characteristics of ZnO nanowire FETs.

II. EXPERIMENTAL DETAILS

A. Fabrication of ZnO Nanowire FETs With Bilayer Gate Dielectrics

ZnO nanowires were grown in a horizontal quartz tube furnace on Au-coated c-plane sapphire substrates through a vapor transport method. The diameter of the grown ZnO nanowires was found to be ~ 110 nm and a length extending to 5 μm or more, as determined by scanning electron microscopy (SEM) and transmission electron microscopy analyses. The method of synthesizing ZnO nanowires by vapor transport has been described in detail elsewhere [3]. ZnO nanowire FETs with bilayer organic–inorganic gate dielectrics were fabricated in the following way [12]. First, PI solution was spin-coated at 4000 r/min onto a 300-nm-thick SiO₂ layer on a heavily doped p-type (p⁺) Si wafer, followed by soft baking at 135 °C for 30 min to evaporate the solvent. The PI solution was prepared by dissolving a type of polyamic acid, p-phenylene biphenyl-tetracarboximide (BPDA-PDA, PI2610D, Dupont, Inc.), n N-methyl-2-pyrrolidone (1:5 ratio by weight). The thickness of the PI layer, an organic dielectric, was found to be ~ 15 nm. Then, the grown ZnO nanowires were dispersed in isopropyl alcohol by sonication for 2–3 min, and transferred onto the top surface of the PI/SiO₂/Si substrate by dropping and drying the nanowire suspension. Source and drain electrodes were patterned using photolithography and liftoff of a Ti (30 nm)/Au (40 nm) film. Finally, to improve the device's performance by enhancing the gate coupling effect, and to prevent the adsorption of water or gas molecules in ambient air [2], [3], the fabricated devices were coated with a poly(methyl methacrylate) layer by spin-coating at 4000 r/min, followed by soft-baking on a hot plate at 100–110 °C for 5 min.

B. Device Scheme and Characterization Methodology

The device layout of a ZnO nanowire FET with bilayer organic–inorganic gate dielectrics is illustrated in Fig. 1(a). The FET device was irradiated with a proton beam to create trapped hole charges within the SiO₂ dielectric layer. Fig. 1(b) presents a schematic illustrating the generation of electron–hole pairs and the charge build-up in an SiO₂ dielectric layer by proton irradiation [14]–[16]. The silicon dioxide in our ZnO nanowire FET structure is sensitive to ionizing radiation [16], [17]. It has been shown that ZnO is much more resistant to high-energy proton bombardment than other semiconductors, such as Si, GaAs, and

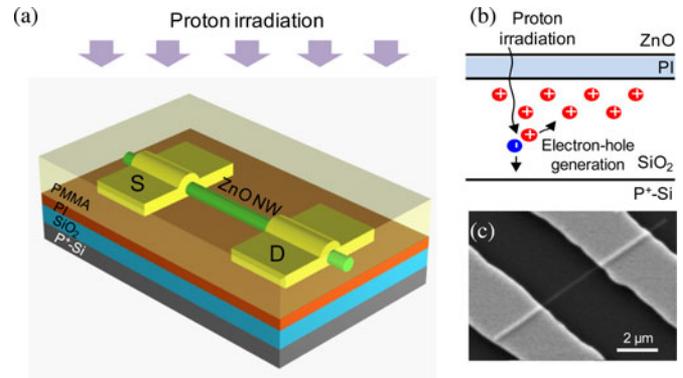


Fig. 1. (a) Schematic illustration of a ZnO nanowire FET device under proton irradiation. (b) Schematic of the charge build-up in the SiO₂ layer by proton irradiation. (c) SEM image of a ZnO nanowire FET device.

GaN [17]–[19]. PI is also durable against exposure to the proton beam [20]. Note that, in our experiment, the PI layer functions as a passivation layer to prevent negatively charged interface states, which can be produced by proton irradiation, between the SiO₂ and the ZnO nanowire [11], [14], [15]. Therefore, by using the bilayer organic (PI)-inorganic (SiO₂) gate dielectrics, only an additional field effect (by positive charges resulting from holes trapped in the SiO₂ dielectric layer) was allowed to occur in the n-type ZnO nanowire FETs. Fig. 1(c) shows an SEM image of a ZnO nanowire FET device with bilayer gate dielectric. Accelerated proton beams were generated using the MC-50 cyclotron (at the Korea Institute of Radiological and Medical Sciences). The proton beam used in our study had a uniformity of 90% or better, its average beam current was 10 nA, and its irradiated area was 5 × 5 cm². The proton beam energy was 10 MeV and the total fluences (Φ) during proton irradiation were 10¹⁰, 10¹¹, and 10¹² cm⁻², corresponding to proton beam irradiation times of 60, 600, and 6000 s, respectively. The current–voltage transfer characteristics of nanowire transistors were measured under dark environment before and after proton irradiation using a semiconductor parameter analyzer (Agilent B1500 A) in an ambient atmosphere at room temperature.

III. RESULTS AND DISCUSSION

A. Electrical Characteristics of Bilayer Dielectric ZnO Nanowire FETs After Proton Irradiation

The electrical characteristics of the ZnO nanowire FETs were measured before and after the devices were exposed to the proton beam. Fig. 2(a)–(c) shows representative electrical data of n-channel ZnO nanowire FETs, before and after proton irradiation with fluencies of 10¹⁰ cm⁻² (black, top), 10¹¹ cm⁻² (red, middle), and 10¹² cm⁻² (blue, bottom), respectively. The output characteristics (I_D – V_{DS} , drain current versus drain–source voltage) of the ZnO nanowire FETs exhibited well-defined linear regimes at low biases and saturation regimes at high biases. These features were still present after proton irradiation. However, it can be seen that the drain conductance ($G = dI_D/dV_{DS}$) of the ZnO nanowire FETs increased after the proton irradiation, which results from the shift in threshold voltage of the FETs.

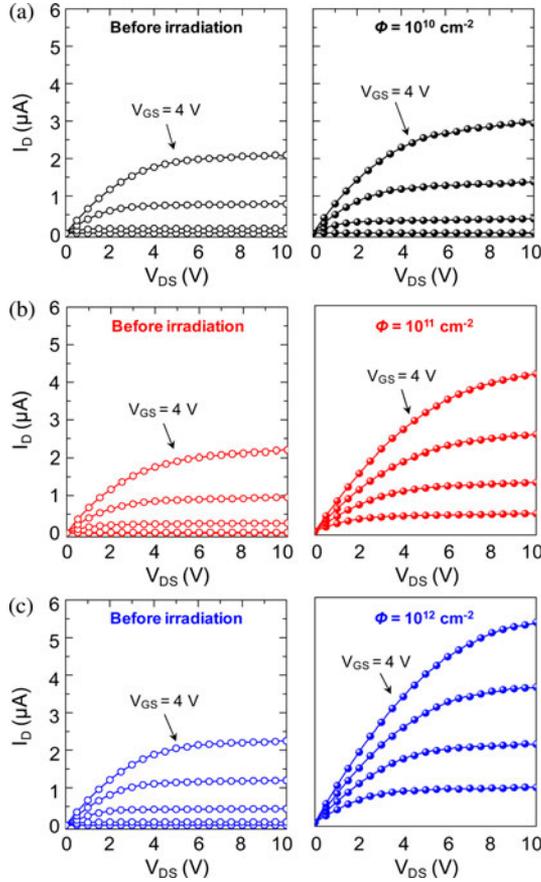


Fig. 2. Electrical characteristics of ZnO nanowire FETs before (open circles) and after (filled circles) proton irradiation: (a)–(c) I_D – V_{DS} output characteristics at various values of V_{GS} , ranging from 4 to -2 V (2-V interval), at proton fluences of 10^{10} (black), 10^{11} (red), and 10^{12} cm^{-2} (blue).

When ZnO nanowire FETs are bombarded with ionizing particles, electron–hole pairs are generated in the SiO_2 dielectric. The electrons in the SiO_2 are then immediately swept out of the oxide, because the electrons are much more mobile than the holes. As a result, some of the irradiation-induced holes are trapped at localized trap sites in the bulk SiO_2 layer, becoming positive oxide-trapped charges [14]–[16]. The proton-induced positive oxide charges in the SiO_2 dielectric layer effectively enhance the electric field in the direction of the ZnO nanowire, which induces more electron accumulation in the conduction channel and thus larger current flow [9], [12], [14]. In addition, as the irradiation fluence increased (Φ from 10^{10} to 10^{12} cm^{-2} , corresponding to irradiation times from 60 to 6000 s), the I_D – V_{DS} curves show that the drain conductances in the ZnO nanowire FETs increased under the same applied gate bias conditions [see Fig. 2(a)–(c)], indicating that more proton-induced trapped hole charges were generated in the SiO_2 layer.

Furthermore, the increased current at the same drain bias implies that the threshold voltage V_{th} of n-type ZnO nanowire transistors shifted toward the negative gate bias direction. The representative transfer characteristics (I_D – V_{GS} , drain current versus gate–source voltage) of ZnO nanowire FETs at V_{DS} of 1 V before (open circles) and after (filled circles) proton irradiation

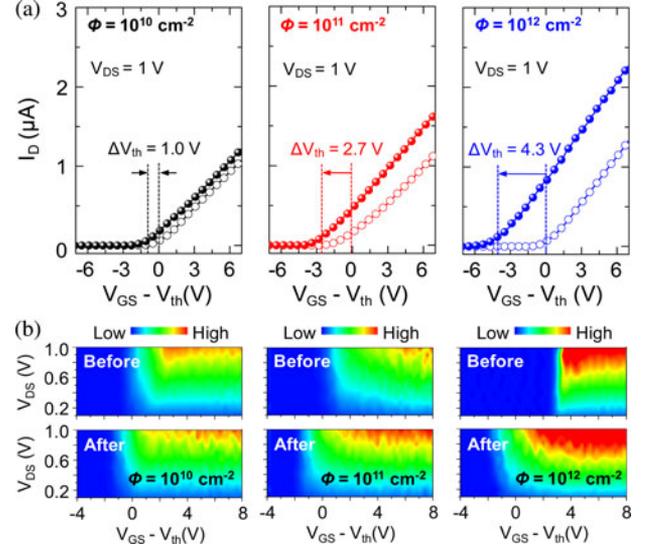


Fig. 3. (a) I_D – V_{GS} transfer characteristics of ZnO nanowire FETs measured at a V_{DS} of 1 V before (open circles) and after (filled circles) proton irradiation, for fluences of 10^{10} cm^{-2} (black), 10^{11} cm^{-2} (red), and 10^{12} cm^{-2} (blue). (b) Contour plots of transconductance g_m as a function of the gate electric field ($V_{GS} - V_{th}$) and the source–drain electric field V_{DS} , before and after irradiation at fluences of 10^{10} (left), 10^{11} (middle), and 10^{12} cm^{-2} (right). Note that V_{th} in $V_{GS} - V_{th}$ is the threshold voltage of the ZnO nanowire FET before proton irradiation and the shift in the threshold voltage (ΔV_{th}) is the difference between the threshold voltages before and after proton irradiation.

ation as a function of proton fluence clearly showed good ON and OFF switching behavior, which indicates that gate bias is effectively applied to the conduction channel through bilayer gate dielectrics [see Fig. 3(a)]. The shifts in the threshold voltage (ΔV_{th}) of the ZnO nanowire FETs were found to be -1.0 , -2.7 , and -4.3 V for fluences of 10^{10} , 10^{11} , and 10^{12} cm^{-2} , respectively. The threshold voltage shift in the negative gate bias direction is associated with an increase of the local electric field in the SiO_2 dielectric layer due to the trapped holes induced by the proton irradiation. It is important to note that this result demonstrates that the threshold voltage of nanowire transistors strongly depends on the proton irradiation fluence. Larger numbers of trapped holes from increased irradiation fluence lead to an enhanced vertical electric field, thus making it easier to create an accumulation channel for a given gate bias. Consequently, the threshold voltage shifts in the negative gate bias direction more with increasing irradiation fluence. This behavior can also be clearly observed in the contour plots of the transconductance ($g_m = \partial I_D / \partial V_{GS}$), which were obtained from I_D – V_{GS} curves measured for V_{DS} ranging from 0.1 to 1 V, as a function of the gate electric field ($V_{GS} - V_{th}$) and source–drain electric field (V_{DS}) before and after proton irradiation at fluences of 10^{10} , 10^{11} , and 10^{12} cm^{-2} [see Fig. 3(b)]. As the fluence increased, the conducting region shifted more toward negative gate voltage, with increased shifts in the threshold voltage, ΔV_{th} . Therefore, it can be concluded that electrostatic modulation in ZnO nanowire FETs depends highly on irradiation-induced charges in the dielectric layer.

Fig. 4 shows the change in the transconductance g_m and field-effect mobility μ_{FE} as a function of the irradiation time,

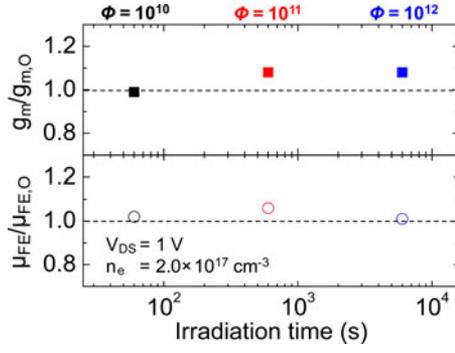


Fig. 4. Normalized transconductance ($g_m/g_{m,o}$) and field-effect mobility ($\mu_{FE}/\mu_{FE,o}$) as a function of the irradiation time.

normalized to the values ($g_{m,o}$ and $\mu_{FE,o}$) of the ZnO nanowire FETs before proton irradiation. The transconductance was found to be 168, 184, and 226 nS for ZnO nanowire FETs for fluences of 10^{10} , 10^{11} , and 10^{12} cm^{-2} , respectively. The field-effect mobility μ_{FE} of ZnO nanowire FETs can be calculated from the equation $\mu_{FE} = g_m L^2 / V_{DS} C_{NW}$. Here, L is the channel length (~ 4 μm) and $C_{NW} = 2\pi\epsilon_r\epsilon_0 L / \cosh^{-1}[(r+h_{bi})/r]$ is the approximate gate capacitance of the nanowires in the bilayer insulators (SiO_2 and PI), which can be calculated with a model of a cylinder on an infinite metal plate [7], h_{bi} is the thicknesses of the bilayer dielectric which is the sum of the thicknesses of the dielectric layer of SiO_2 ($h_{\text{SiO}_2} = 300$ nm) and PI ($h_{\text{PI}} = 15$ nm), r is the nanowire diameter (~ 110 nm), ϵ_{bi} is the dielectric constant of the bilayer gate insulator, and ϵ_0 is the permittivity constant of the vacuum. Note that we used ϵ_{bi} approximately as 3.9 which is the dielectric constant of the SiO_2 insulator, but not the PI dielectric constant (2.9), because the whole bilayer dielectric is much more dominated by a thick SiO_2 layer than a thin PI layer. The calculated mobility of ZnO nanowire FETs at an equal carrier concentration condition ($\sim 2.0 \times 10^{17}$ cm^{-3}) was found to be 115, 130, and 156 cm^2/Vs for ZnO nanowire FETs for fluences of 10^{10} , 10^{11} , and 10^{12} cm^{-2} , respectively. From Fig. 4, the transconductance and the field-effect mobility of the nanowire FETs were not significantly changed after proton irradiation. Therefore, we think that the observed changes in overall current–voltage characteristics can be described simply in terms of the shift in threshold voltage.

As explained already before, the bilayer dielectric ZnO nanowire FETs are similar case of the suspended-type ZnO nanowire FETs reported by Hong *et al.* [11]. However, the drawbacks in the suspended-type ZnO nanowire FETs are the poor gating effects, complicate fabrication process, and the low device yield. On the contrary, the bilayer dielectric ZnO nanowire FETs are simple in device fabrication, and more importantly, the device performance is better in terms of gating effects and ON/OFF switching behavior. Therefore, the technique demonstrated in this study can provide a viable alternative to achieve favorable device functions for practical applications.

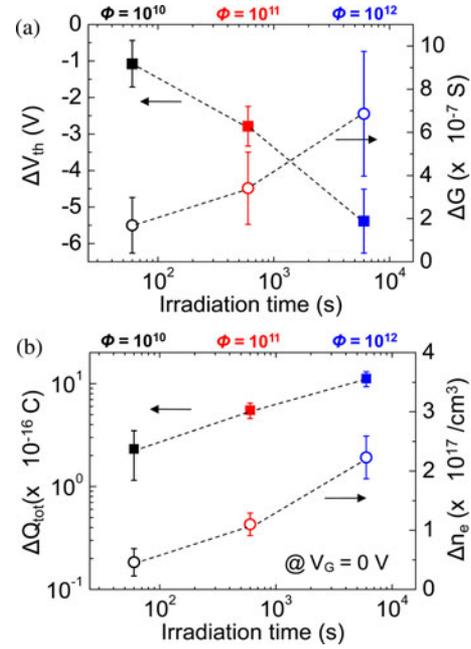


Fig. 5. Statistical distributions of (a) the threshold voltage shift ΔV_{th} and the conductance change ΔG , and (b) the total charge change ΔQ_{tot} and the carrier density change Δn_e of ZnO nanowires FETs after irradiation fluences of 10^{10} cm^{-2} (black, 11 FETs), 10^{11} cm^{-2} (red, 10 FETs), and 10^{12} cm^{-2} (blue, 15 FETs).

B. Statistical Analysis of Device Parameters

To statistically characterize the change in the threshold voltage ΔV_{th} , the change in drain conductance ΔG , the change in total charge ΔQ_{tot} , and the change in carrier density Δn_e of ZnO nanowire FETs after proton irradiation, a total of 36 nanowire FET devices were fabricated and systematically studied: 11 FETs, 10 FETs, and 15 FETs at fluences of 10^{10} cm^{-2} (60 s), 10^{11} cm^{-2} (600 s), and 10^{12} cm^{-2} (6000 s), respectively. Fig. 5(a) shows a plot of the threshold voltage shift and conductance change as a function of proton irradiation time. The threshold voltages were determined in the linear region by the extrapolation method at a drain voltage of 1 V [21]. The drain conductance G was measured at a drain voltage of 1 V and a gate voltage of 0 V. The values of the threshold voltage shifts and conductance enhancements of the ZnO FETs were found to be -1.07 ± 0.64 V and 169 ± 129 nS, -2.79 ± 0.54 V and 341 ± 167 nS, and 5.39 ± 0.88 V and 686 ± 289 nS for irradiation times of 60, 600, and 6000 s, respectively. Fig. 5(b) shows a plot of the changes in the total charge and carrier densities as functions of proton irradiation time. The total charge in the nanowire was estimated at zero gate bias using the relation $Q_{tot} = C_{NW} |V_{GS} - V_{th}|$ [22]. The carrier concentration, $n_e = Q_{tot} / e\pi r^2 L$, can also be determined at zero gate bias. The increased values of the total charge and carrier density of the ZnO FETs were found to be $(2.32 \pm 1.17) \times 10^{-16}$ C and $(0.46 \pm 0.23) \times 10^{17}$ cm^{-3} , $(5.53 \pm 0.94) \times 10^{-16}$ C and $(1.10 \pm 0.19) \times 10^{17}$ cm^{-3} , and $(11.2 \pm 1.83) \times 10^{-16}$ C and $(2.23 \pm 0.36) \times 10^{17}$ cm^{-3} for irradiation times of 60, 600, and 6000 s, respectively. Additionally, the subthreshold slope ($SS = dV_{GS}/d(\log I_D)$), repeated bias sweep and hysteresis characteristics of the bilayer

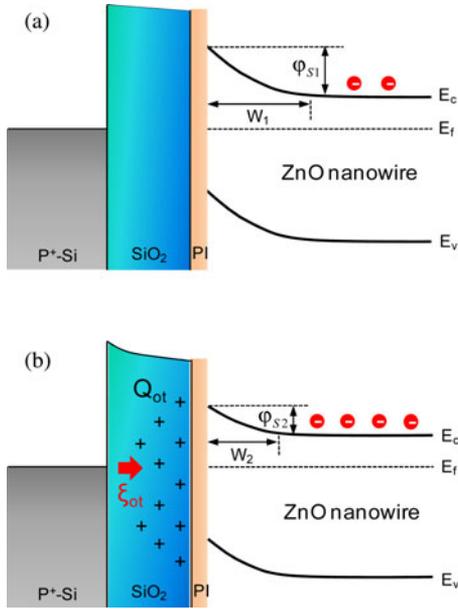


Fig. 6. Energy band diagrams of ZnO nanowire FETs (a) before and (b) after proton irradiation. φ_{s1} and φ_{s2} are surface barrier potentials before and after irradiation, and W_1 and W_2 are surface depletion widths before and after irradiation, respectively.

dielectric ZnO nanowire FETs were checked after proton irradiation (data not shown here). And, we did not observe any significant changes in these characteristics, indicating that our device structure is efficient to prevent the formation of additional interface states which can be created by proton irradiation. Consequently, it can be said that the electrical properties of our devices were well controlled by the proton irradiation fluence with reliable electrical characteristics.

C. Energy Band Diagram Before and After Proton Irradiation

To explicitly explain the experimental observations of the effects of proton irradiation on the electrical properties of the ZnO nanowire transistors, the equilibrium energy band diagrams across the ZnO nanowire FET before and after irradiation are shown in Fig. 6(a) and (b), respectively. After irradiation [see Fig. 6(b)], the electronic conduction band E_c and the valence band E_v of the ZnO nanowire exhibit a relatively smaller surface barrier potential φ_{s2} and a narrower surface depletion width W_2 than before irradiation [see Fig. 6(a)]. The difference is due to the smaller surface band bending, influenced by the local electric field from the positive oxide-trapped charges in the bulk SiO_2 [9], [12], [14]. The formation of such a constructive electric field decreases the surface barrier potential and the depletion region, and thus induces the threshold voltage to shift toward the negative gate bias direction, resulting in an increased electrical conductance under the same applied gate bias.

The threshold voltage shift in the ZnO nanowire FETs due to the oxide charges effect is similar to the case of the conventional metal–oxide–semiconductor (MOS) devices [23]. It is well known that oxide charges, such as interface trapped charges, fixed oxide charges, mobile ionic charges, and oxide

trapped charges usually contribute to threshold voltage shifts in MOS devices [21], [23]. For convenience, we assume in this discussion that all other charges remained unchanged as the oxide trapped charge Q_{ot} was introduced by proton irradiation, which produces the positively charged holes trapped in the SiO_2 dielectric layer [15], [23]. After irradiation, the voltage shifted across the oxide because of the oxide trapped charge ΔV_{ot} ; the shift be obtained from [23]

$$\Delta V_{ot} = -\frac{Q_{ot}}{C_{bi}} \quad (1)$$

$$Q_{ot} = \frac{1}{h} \int_0^h x \rho_{ot}(x) dx \quad (2)$$

where $\rho_{ot}(x)$ is the charge density per unit volume and h is the thickness of the dielectric layer. The effect on the voltage shift is weighted according to the location and the density of the charges. A larger density of trapped holes will cause a bigger shift. Proton-induced positive charges are equivalent to an additional positive bias on the semiconductor gate, so a larger negative gate bias is required to achieve the same semiconductor band bending. Therefore, in n-channel ZnO nanowire transistors, the trapped oxide charges will act as an additional electric field ξ_{ot} in the conductive channel, allowing less gate voltage to induce the same accumulation channel. Consequently, the threshold voltage shifts in the negative direction.

IV. CONCLUSION

In summary, we demonstrated a novel technique for the electrostatic modulation of ZnO nanowire FETs using oxide-trapped charges selectively generated in bilayer gate dielectric by proton irradiation. Proton irradiation-induced charges trapped in the SiO_2 dielectric layer significantly affected the electrical properties of n-channel ZnO nanowire FET devices. In particular, the conductance, total charge, and carrier density increased, and the threshold voltage shifted in the negative gate bias direction after proton irradiation. Furthermore, it was possible to selectively modulate the electrical characteristics of the ZnO nanowire FETs, which is important for practical applications, by varying the proton irradiation time.

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