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Effect of High-Energy Proton Irradiation of ZnO-Nanowire Field-Effect Transistors

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The effects of proton irradiation on the electrical properties of ZnO nanowire field effect transistors (FETs) were investigated. The vertically-aligned ZnO nanowires were synthesized via a simple thermal evaporation method, and their structural and electrical properties were characterized. The FET devices were fabricated using the vertically-aligned ZnO nanowires and were then irradiated with the high-energy proton beams of 35 MeV with fluences of $4 \times 10^{11} - 4 \times 10^{12}$ protons/cm^2, comparable to an aerospace environment. The electrical properties of the ZnO nanowires FET devices exhibited significant changes in the electrical conductivity, the threshold voltage shift, and the surface punch-through after proton irradiation.

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I. INTRODUCTION

One-dimensional (1-D) nanostructures, such as nanowires and carbon nanotubes, are currently the subject of intensive research due to their potential use as building blocks for nanoelectronic applications. Of these, single-crystalline nanostructures of semiconducting metal oxides such as ZnO, In_2O_3 and SnO_2, have been synthesized and extensively studied [1–3]. In particular, ZnO nanostructures have attracted considerable attention for use in versatile applications, such as ultraviolet solid-state light emitters, photodetectors, sensors, and field effect transistors (FETs) [4–7], due to the unique properties of ZnO materials, which include the existence of highly stable excitons (60 meV), a high electron saturation velocity, the availability of large-area substrates, the amenability to wet chemical etching, the relatively low materials' costs, and the high radiation resistance [8,9]. Particularly, with regard to the radiation properties, it has been reported that ZnO is more tolerant than semiconductors such as Si, GaAs or GaN, to room-temperature bombardment by energetic particles [10–12]. This suggests that ZnO can be useful for applications on an aerospace radiation environment. The radiation environment in the Earth's magnetosphere consists of a nearly isotropic flux of energetic charged particles: 85 % protons, 14 % alpha particles and 1 % heavy ions covering the full range of elements [13]. The impingement of these high-energy particles on devices and circuits can cause undesirable effects, for example, latchup, electrical interference, charging, sputtering, erosion, and puncture of the target device. As a result, degradation of the device performance and lifetime or even a system failure of the underlying electronics may occur. Consequently, the effects of various types of radiation on 1-D nanostructure-based devices are needed to understand the device performance under an aerospace radiation environment and to develop radiation-robust devices and circuits.

However, despite many studies on the synthesis and the structural/electrical characterization of 1-D nanosturctures, including ZnO nanowires, in recent decade, only recently have the radiation effects on 1-D nanostructure-based devices been reported. For example, the structural characterization of GaN nanowire and the performance of GaN nanowire FETs during heavy-ion irradiation were presented by Ayres *et al.* [14]. Ju *et al.* also reported that ZnO nanowire FETs fabricated with organic gate nanodielectrics were more tolerant than those fabricated with SiO_2 gate dielectrics under a 10 MeV proton beam with fluences of 1×10^{10} and 5×10^{11} protons/cm^2, where all the ZnO nanowire FET devices were passivated with a layer of SiO_2 (∼300 nm) [15]. We also previously reported that single-walled carbon nanotube (SWNT) network FETs with metallic or semiconducting properties were very tolerant under high-energy proton beams at 10 − 35 MeV [16].

Here in this work, we report a systematic study of the electrical properties of ZnO nanowire FETs before and after proton irradiation. The ZnO nanowire FET devices were exposed to 35-MeV proton beams with flu-

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ences from 4×10^{11} to 4×10^{12} protons/cm^2, which are comparable to the aerospace environment. The proton-irradiated ZnO nanowire FET devices showed a significant positive shift of threshold voltage, *i.e.*, ΔV_{th} = 10.5 V and 9.7 V for fluences of 4×10^{11} and 4×10^{12} protons/cm^2, respectively. The FET devices exposed to proton beams also showed a decrease in the electrical conductivity. In particular, the current-voltage characteristics of the proton-irradiated ZnO nanowire FETs with a fluence of 4×10^{12} protons/cm^2 exhibited a power-law conduction behavior consistent with a space-charge-limited conduction mechanism in the presence of trap states.

II. EXPERIMENT

ZnO nanowires were synthesized by thermally vaporizing a mixed source of commercial ZnO powder (99.995 %) and graphite powder (99 %) with a ratio of 1:1 in a horizontal tube furnace. A Au thin film ($\sim$3 nm) layer as a catalyst for the growth of ZnO nanowires was deposited on a pre-cleaned c-plane sapphire substrate by using an electron beam evaporator. The source materials and the Au-catalyzed sapphire substrate were placed in an alumina boat; then, the boat was loaded at the center of a quartz tube. Then, the ZnO nanowires were grown on the Au-catalyzed sapphire substrate at $\sim$920 °C for 20 min under a flow of Ar and O_2 (0.2 % O_2 in Ar) at a flow rate of $\sim$50 sccm. The high quality of the ZnO nanowires synthesized is demonstrated using scanning electron microscopy (SEM), high resolution transmission electron microscopy (HRTEM), X-ray diffraction (XRD), and room temperature photoluminescence (PL). To fabricate the ZnO nanowire FET devices, we transferred ZnO nanowires in ethanol from the substrate to the silicon wafer with a 300 nm thick thermally grown oxide by dropping and drying a nanowire suspension. The nanowire suspension was made by briefly sonicating the substrate with ZnO nanowires grown on its surface in ethanol for 30 − 60 sec. The highly-doped p-type silicon wafer used in this study served as a back gate electrode. Metal electrodes consisting of Ti (100 nm)/Au (100 nm) were deposited by using an electron beam evaporator and were defined as the source and the drain electrodes by using a photolithography and lift-off process. The gap distance between the electrodes was typically 3 − 4 μm, as shown in Figure 2(b).

For the proton irradiation experiments, the accelerated proton beams were generated using a MC-50 cyclotron (at the Korea Institute of Radiological and Medical Sciences). The beam diameter was $\sim$6 cm, its uniformity was $\sim$90 %, and the average beam current was 10 nA. In our study, a 35-MeV proton beam energy was used, and the total fluence during the proton irradiation ranged from 4×10^{11} to 4×10^{12} protons/cm^2. In particular, for the 35-MeV proton irradiation, we used

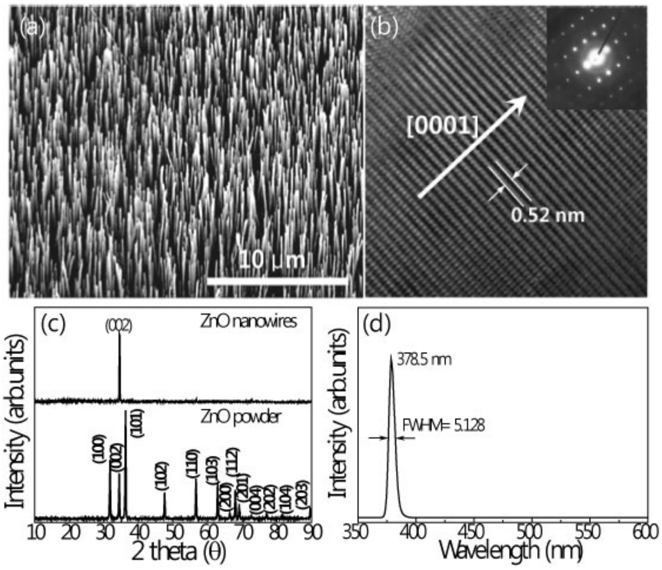

Fig. 1. (a) Scanning electron microscope (SEM) and (b) transmission electron microscope (TEM) images of vertically-aligned ZnO nanowires. The inset in (b) shows a selective area electron diffraction (SAED) pattern, indicating single-crystalline ZnO nanowires. (c) Photoluminescence (PL) at room temperature and (d) X-ray diffraction (XRD) peaks of ZnO nanowires.

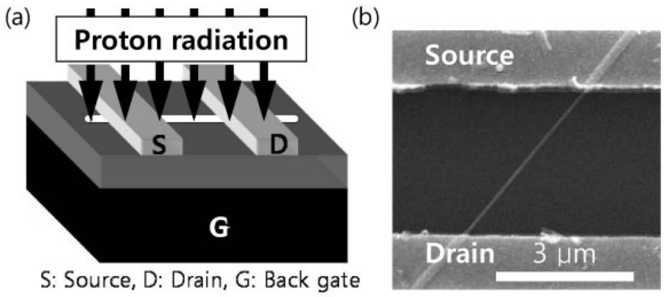

Fig. 2. (a) Schematic of a ZnO nanowire FET device under proton irradiation. (b) FESEM image of a ZnO nanowire FET.

the spread-out Bragg peak (SOBP) method, which is a range-modulating method to obtain a uniform longitudinal and transverse dose distribution in a target for a fixed proton energy [17]. Actually, the 30 − 60 MeV protons at a dose of $\sim 10^{12}$ protons/cm^2 are equivalent to the amount of proton radiation for a few hundred years in a low Earth orbit environment [18]. Note that the proton beam irradiated the whole area of the nanowire FET devices. In the actual aerospace radiation environment, protons or any other radiations irradiate the whole area of the device. The electrical measurements before and after proton irradiation were performed at room temperature by measuring the source-drain current-voltage characteristics as a function of the gate voltage applied through the highly-doped silicon back gate by using a semiconductor parameter analyzer (HP4155C).

III. RESULTS AND DISCUSSION

Figure 1(a) shows a SEM image (tilted view) of ZnO nanowires vertically and uniformly grown on the entire sapphire substrate. The structural characterization of the ZnO nanowires was performed by using HRTEM and XRD. Figure 1(b) shows the lattice fringes and the corresponding selective area electron diffraction (SAED) pattern (inset) of the ZnO nanowires. These results confirmed that the ZnO nanowire was a single crystal. The lattice spacing the of approximately 2.6 Å between adjacent lattice planes corresponds to the distance between two (002) crystal planes, confirming the [0001] direction as the preferred growth direction for ZnO nanowires. This is consistent with the XRD results. As shown in Figure 1(c), compared to the XRD profiles of the ZnO powder, those of the ZnO nanowires show a relatively much more intense (002) peak, indicating a preferred *c*-axis orientation. The optical properties of ZnO nanowires were investigated by using room-temperature PL spectroscopy. The PL spectrum of ZnO nanowires exhibits a strong, sharp, near-band-edge (NBE) excitonic-related ultraviolet (UV) emission at approximately 378 nm without deep level (DL)-related emission, as shown in Figure 1(d). These overall results confirm that the ZnO nanowires are single crystalline with a preferred growth direction of [0001].

Figure 2 shows the schematic structure and the representative field-emission scanning electron microscopy (FESEM) image of a single ZnO nanowire FET with the back-gate configuration. To avoid potential damage caused by electron beams during FESEM examination on the FET devices used in the proton irradiation experiments, we first examined and identified the FET devices of a single ZnO nanowire connecting the electrodes by using an optical microscope; then, we systematically measured the electrical properties of the nanowire FET devices before and after proton irradiation. Figures 3 and 4 present typical output characteristics (source-drain current versus source-drain voltage; $I_{DS}-V_{DS}$) and transfer characteristics (source-drain current versus gate voltage; $I_{DS}-V_G$) of the ZnO nanowire FETs without any surface passivation layer before and after 35-MeV proton irradiation. The irradiation times and the fluences during the proton irradiation for the devices shown in Figures 3 and 4 were 600 sec and 6000 sec, 4.1×10^{11} and 4.1×10^{12} protons/cm^2, respectively. We first measured the output and the transfer characteristics before proton irradiation, which were followed by exposure of the devices to the proton beam and systematic measurement of the electrical characteristics of the same devices after proton irradiation.

For the FET device exposed to a proton beam with a fluence of 4.1×10^{11} protons/cm^2 (irradiation time = 10 min), as shown in Figure 3(a), although the electrical conductivity decreased after proton irradiation, the source-drain current versus source-drain voltage

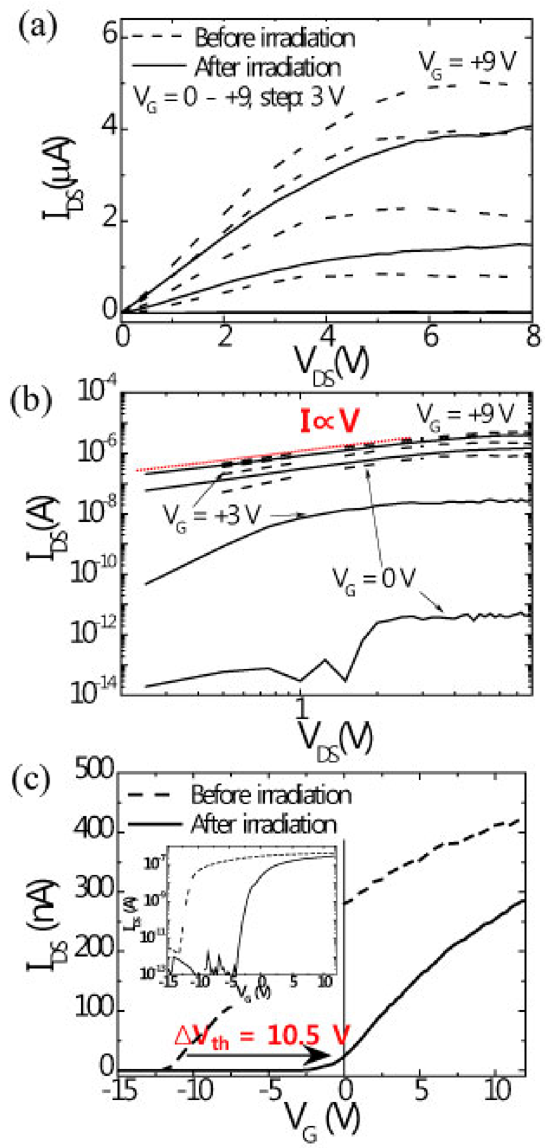

Fig. 3. Output characteristics (a) on a linear scale and (b) on a log-log scale, and (c) transfer characteristics of a ZnO nanowire FET before and after proton irradiation at 35 MeV with a fluence of 4×10^{11} protons/cm^2 (at $V_{DS} = 0.1$ V). The inset in (c) shows a semi-logarithmic plot of the source-drain current versus the gate voltage at $V_{DS} = 0.1$ V. The shift in the threshold voltage (ΔV_{th}) is 10.5 V.

($I_{DS}-V_{DS}$) curves before and after proton irradiation have well-defined linear regions at low bias and saturation regions at high bias. Figure 3(b) shows a logarithmic plot of the $I_{DS}-V_{DS}$ curves at different gate biases (from 0 V to +9 V in steps of 3 V). While the electrical conductivity of the proton-irradiated ZnO nanowire FET decreased, the FET device exhibited, qualitatively, a $I_{DS} \propto V_{DS}$ relationship in the overall gate bias as shown in Figure 3(b), indicating Ohmic conduction behavior [19]. This behavior was consistently observed for Ohmically contacted ZnO nanowires. As shown in the

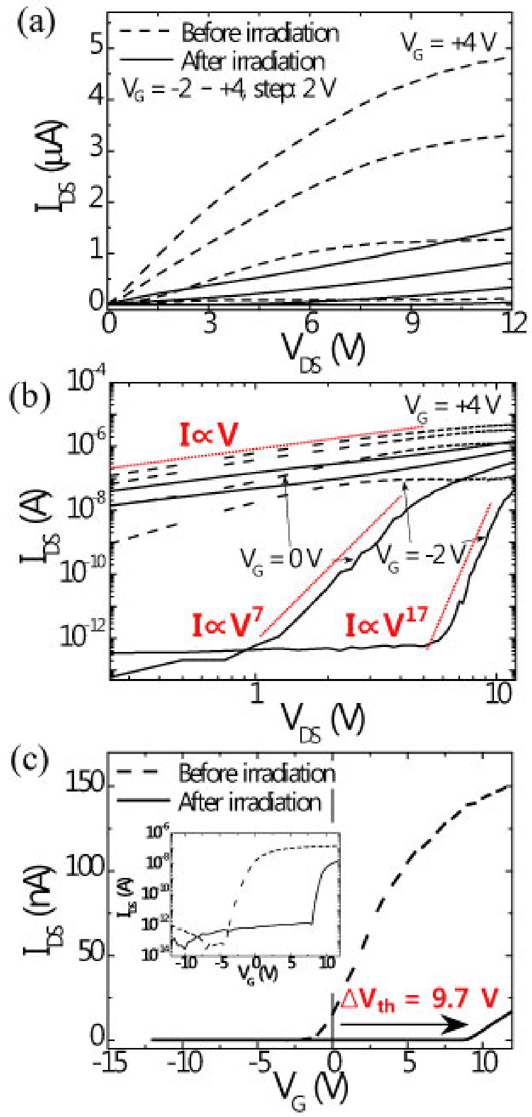

Fig. 4. Output characteristics (a) on a linear scale and (b) on a log-log scale, and (c) transfer characteristics of a ZnO nanowire FET before and after proton irradiation at 35 MeV with a fluence of 4×10^{12} protons/cm^2 (at $V_{DS} = 0.1$ V). The inset in (c) shows a semi-logarithmic plot of the source-drain current versus the gate voltage at $V_{DS} = 0.1$ V. The shift in the threshold voltage (ΔV_{th}) is 9.7 V.

inset of Figure 3(c), the current on/off ratio (Ion/Ioff) of the ZnO nanowire FET device both before and after proton irradiation is approximately $10^5 - 10^6$. Compared to source-drain current versus gate voltage ($I_{DS} - V_G$) measured before proton irradiation, a significant positive shift in the threshold voltage ($\Delta V_{th} = 10.5$ V) was observed in Figure 3(c), indicating that the Femi level shifted away from the conduction band after proton irradiation, consistent with the oxygen-induced depletion of free electrons [19].

The irradiation effects on the ZnO nanowire FET device caused by the proton beam with a higher fluence of 4.1×10^{12} protons/cm^2 are shown in Figure 4. The $I_{DS} - V_{DS}$ curves of the ZnO nanowire FET device before proton irradiation show well-defined linear regions at low bias and saturation regions at high bias. However, unlike the output characteristics of the device exposed to proton beam at a fluence of 4.1×10^{11} protons/cm^2 (Figures 3(a) and (b)), in the case of the higher fluence of 4.1×10^{12} protons/cm^2, in addition to a decrease in the conductance after proton irradiation, only linear regions existed in the $I_{DS} - V_{DS}$ characteristics, similar to punch-through phenomenon in short-channel effects [20]. The proton-irradiated ZnO nanowire FET exhibited qualitatively different output ($I_{DS} - V_{DS}$) characteristics, as evidenced by the power-law $I_{DS} \propto V_{DS}^n$ relationship shown in the logarithmic plot of the $I_{DS} - V_{DS}$ curves at different gate biases (from -2 V to $+4$ V in steps of 2 V) in Figure 4(b). Also, a transition from Ohmic conduction ($I_{DS} \propto V_{DS}$) at low bias to a different conduction ($I_{DS} \propto V_{DS}^n$) at high bias was clearly observed. These results are in good agreement with the space-charge-limited conduction behavior in the oxygen-depleted CdS nanowires reported by Gu and Lauhon [19]. This power law relationship, with $n > 2$, is a typical characteristic of a space-charge-limited conduction behavior in the presence of traps [19,21]. The current on/off ratio (I_{on}/I_{off}) of the ZnO nanowire FET both before and after proton irradiation is approximately $10^5 - 10^6$ as shown in the inset of Figure 4(c). Similar to the FET device exposed to the proton beam with a fluence of 4.1×10^{11} protons/cm^2 (Figure 3), one exposed to proton beam with a fluence of 4.1×10^{12} protons/cm^2 also showed a significant positive shift in the threshold voltage ($\Delta V_{th} = 9.7$ V), which was determined by comparing the $I_{DS} - V_G$ curves measured before and after proton irradiation (Figure 4(c)).

As shown in Figures 3 and 4, the significant shifts in the threshold voltage can be generally associated with the generation of positive trap states in the SiO_2 dielectric or at the ZnO NW-SiO_2 interface [16]. Although a complete understanding of the radiation effects on ZnO nanowire FETs is not currently available, a possible mechanism may be a radiation-induced trap-generation process in the gate insulator, SiO_2. It is well known that electron-hole pairs are generated during the radiation and that the electrons in SiO_2 are then immediately swept out of the oxide because the electrons are much more mobile than the holes [16, 22, 23]. Consequently, the holes remaining in the oxide reach the Si-SiO_2 interface via hopping transport and form positive trap states. Moreover, it is well known that the chemisorption of ambient gases, primarily oxygen, has a strong effect on the electrical properties of ZnO [24,25]. Therefore, the electrical properties of the proton-irradiated ZnO nanowire FETs seem to be influenced by the radiation-induced positive trap states and the oxygen-induced depletion of free electrons. However, further investigation is required to understand the detailed mechanism.

IV. CONCLUSION

In summary, we synthesized single-crystalline ZnO nanowires via a thermal evaporation method and fabricated ZnO nanowire FETs. To investigate the proton irradiation effects on the electrical properties of ZnO nanowire FETs, we irradiated the FET devices with the high-energy proton beams of 35 MeV at fluences of 4 $\times$ 10^{11} $-$ 4 $\times$ 10^{12} protons/cm^2, which are comparable to the aerospace environment. The electrical properties of ZnO nanowires FET devices exhibited significant changes, such as threshold voltage shifts, space-charge-limited conduction behavior, and surface punch-through, as well as a decrease in the electrical conductivity, after the high-energy proton irradiation. The radiation-induced changes in the electrical properties of ZnO nanowire FETs can be explained in terms of positive trap states within SiO_2 or near the SiO_2-ZnO nanowire interface and in terms of the oxygen-induced depletion of free electrons.

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REFERENCES

[1] Z. L. Wang, J. Phys.: Condens. Matter. **16**, R829 (2004).
[2] B. Lei, C. Li, D. Zhang, Q. F. Zhou, K. K. Shung and C. Zhou, Appl. Phys. Lett. **84**, 4553 (2004).
[3] S. V. Kalinin, J. Shin, S. Jesse, D. Geohegan, A. P. Baddorf, Y. Lilach, M. Moskovits and A. Kolmakov, J. Appl. Phys. **98**, 044503 (2005).
[4] M.-C. Jeong, B.-Y. Oh, M.-H. Ham and J.-M. Myoung, Appl. Phys. Lett. **88**, 202105 (2006).
[5] H. Kind, H. Yan, B. Messer, M. Law and P. Yang, Adv. Mater. **14**, 158 (2002).
[6] J. S. Kim, W. I. Park, C.-H. Lee and G.-C. Yi, J. Korean Phys. Soc. **49**, 1635 (2006).
[7] Z. Y. Fan, D. W. Wang, P. C. Chang, W. Y. Tseng and J. G. Lu, Appl. Phys. Lett. **85**, 5923 (2004).
[8] Y. Chen, D. Bagnall and T. Yao, Mater. Sci. Eng. B **75**, 190 (2000).
[9] D. C. Look, Mater. Sci. Eng. B **80**, 383 (2001).
[10] D. C. Look, J. W. Hemsky and J. R. Sizelove, Phys. Rev. Lett. **82**, 2552 (1999).
[11] F. D. Auret, S. A. Goodman, M. Hayes, M. J. Legodi, H. A. van Laarhoven and D. C. Look, J. Phys.: Condens. Matter **13**, 8989 (2001).
[12] S. O. Kucheyev, C. Jagadish, J. S. Williams, P. N. K. Deenapanray, M. Yano, K. Koike, S. Sasa, M. Inoue and K. Ogata, J. Appl. Phys. **93**, 2972 (2003).
[13] A. Holmes-Siedle and L. Adams, *Handbook of Radiation Effects* (Oxford University Press, Oxford, 1993).
[14] V. M. Ayres, B. W. Jacobs, M. E. Englund, E. H. Carey, M. A. Crimp, R. M. Ronningen, A. F. Zeller, J. B. Halpern, M.-Q. He, G. L. Harris, D. Liu, H. C. Shaw and M. P. Petkov, Diam. Relat. Mat. **15**, 1117 (2006).
[15] S. Ju, K. Lee, D. B. Janes, R. C. Dwivedi, H. Baffour-Awuah, R. Wilkins, M.-H. Yoon, A. Facchetti and T. J. Mark, Appl. Phys. Lett. **89**, 073510 (2006).
[16] W.-K. Hong, C. Lee, D. Nepal, K. E. Geckeler, K. Shin and T. Lee, Nanotechnology **17**, 5675 (2006).
[17] Y. K. Lim, B. S. Park, S. K. Lee and K. R. Kim, J. Korean Phys. Soc. **48**, 777 (2006).
[18] P. Le. Metayer, O. Gilard, R. Germanicus, D. Campillo, F. Ledu, J. Cazes, W. Falo and C. Chatry, J. Appl. Phys. **94**, 7757 (2003).
[19] Y. Gu and L. J. Lauhon, Appl. Phys. Lett. **89**, 143102 (2006).
[20] R. S. Muller and T. I. Kamins, *Device Electronics for Intergrated Circuits* (Wiley, New York, 2003).
[21] A. Rose, Phys. Rev. **97**, 1538 (1955).
[22] J. R. Srour and J. M. McGarrity, Proc. IEEE **76**, 1443 (1988).
[23] T. R. Oldham and F. B. McLean, IEEE Trans. Nucl. Sci. **50**, 483 (2003).
[24] L. Lagowski, E. S. Sproles, Jr. and H. C. Gatos, J. Appl. Phys. **48**, 3566 (1977).
[25] Z. Fan, D. Wang, P.-C. Chang, W.-Y. Tseng and J. G. Lu, Appl. Phys. Lett. **85**, 5923 (2004).