Piezotronic and Piezo-phototronic Logic Computations Using Au Decorated ZnO Microwires

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Abstract

Using the coupling effect of localized Schottky barriers formed by Au nanoparticles decorated ZnO microwires (MWs) and piezoelectric potential created upon straining, the highly enhanced piezotronic and piezo-phototronic effect have been applied for logic computations, i.e. designing strain-gated transistors (SGTs) and light-strain-gated transistors (LSGTs) to process mechanical and optical stimuli on the devices into electronic controlling signals. Linking four (L) SGTs following the connection rules, two series logic devices, such as inverter, NAND, AND, NOR, OR and XOR gates with good rectifying behaviors have been fabricated. Moreover, a 2:1 multiplexer can be designed for two independent input square wave signals with different frequencies. The light-strain-gated piezo-phototronic logic device based on Au NPs decorated ZnO MWs may find applications in active flexible electronics, optical micro/nanoelectromechanical systems, tunable bio-optoelectronic probes, and adaptive optical computation.

Keywords: localized Schottky barriers, piezoelectric potential, strain-gated transistors, light-strain-gated transistors

Introduction

Recently, the coupling among piezoelectric potential, semiconductor properties, and optical excitation in wurtzite structured piezoelectric semiconductors, such as ZnO, CdS and GaN, has resulted in novel fundamental new phenomena, excellent device characteristics and various applications, leading to extensive attentions in the emerging field of piezo-phototronics.[1-6] Among the three materials, ZnO nanowire (NW) is of particular interest for investigating piezo-phototronic effect and implementing related applications due to its ultraviolet (UV) range optical response and intrinsic piezoelectric property. ZnO is an important material and its nanostructures has been widely studied for field effect transistors (FETs),[7] UV photodetectors,[8] light emitting diodes,[9] piezoelectric transducer,[10] and solar cells.[11] Especially, *via* coupling semiconducting and piezoelectric properties of ZnO,[12] nanogenerator,[13, 14] piezoelectric FETs,[15] piezoelectric diodes,[16] and piezoelectric humidity/chemical sensors[17] were demonstrated.

Up to now, most of the piezoelectric effect is using polarization charges created at metal/ZnO electrode contact under strains to modulate transport of local charge carriers across the SB, in which the strain stimuli applied on ZnO NW are converted into electronic controlling signals. For example, the NW piezoelectric strain-gated transistors (SGT) was introduced[2, 18] based on a two-terminal metal-semiconductor-metal (M-S-M) structure, using the piezoelectric potential in the NW to modulate the charge carrier transport at the contacts without applying a gate voltage as in traditional FET. However, the sensitivity (control of strain on current) is still not good enough for piezoelectric sensor due to weak couplings among the characteristics of piezoelectricity, photoexcitation and semiconductor. For example, an on/off ratio of 112 was observed for piezoelectric gated transistor,[19] where the working current was restricted to a small range and may limit its practical application.

Therefore, it must be necessary to develop an effective strategy to obtain high sensitivity by using strong couplings among the characteristics before the high performance principle devices of piezotronic and piezo-phototronic logic computations is realized. Our effective strategy is decorating Au nanoparticles (NPs) on ZnO surface to enhance the couplings and realizing the piezotronic and piezo-phototronic logic computations based on the enhanced coupling.

As stated above, piezoelectric potential and Schottky barrier (SB) at the ZnO/metal electrode contact is used as a “gate” that dictates the electrical transport properties of the piezoelectric device. However, the control level is limited. According to previous report, in the case of metal particles fabricated on a semiconductor surface, the Fermi levels in the two materials must be coincident, which results in charge transfer and band bending near the interface in analogy with that on the interface between the metal and semiconductor. Thus the Au NPs can be treated as a lot of SBs on the surfaces of ZnO,[20-22] which deplete more carriers near the ZnO surface, thicken the depletion region. In our previous study, we proved that the localized SBs (by Au NPs decoration) at the ZnO surface is much enhanced the sensitivity of the piezoelectric devices.[23] Therefore, we believe that the numberd SBs can be functioned as traditional SB at the electrode to improve performance of the device, and using the coupling effect of piezoelectric potential and localized SBs to fabricate piezoelectric two-terminal transistors must be important to enhance the current on/off ratio of the devices. Furthermore, the piezo-phototronic effect has been applied to design two-terminal transistors in our study, which process mechanical and optical stimuli on the devices into electronic controlling signals.

In this study, ZnO microwire (MW) (L)SGTs were fabricated by using the piezoelectric polarization charges created at the metal/ZnO interface under strains and UV light to modulate transport of local charge carriers across the SB. Linking four (L)SGTs following the connection rules, two series logic devices such as inverter, NAND, AND, NOR, OR, XOR

and 2:1 multiplexer (MUX) gates with good rectifying behaviors have been fabricated. Here, we got a high $I_{\max}/I_{\min}$ current ratio 9.3×10^4 , which is much higher than the former ratios in this field. The light strain gated piezo-phototronic logic devices based on Au NPs decorated ZnO MWs may find applications in active flexible electronics, optical micro/nanoelectromechanical systems, tunable bio-optoelectronic probes, and adaptive optical computing.

Experimental Section

ZnO MWs were obtained by one step chemical vapor deposition and discrete Au NPs were formed on ZnO surface by spraying deposition.[23] The Au NPs distribution on the upside of the surface (the bottom side surface is almost smooth) due to ZnO MW is lying down. The as-obtained ZnO MWs have lengths of several hundred micrometers, diameters of 0.5~10 μm . The piezoelectric SGTs were fabricated by transferring and bonding single ZnO MW with and without Au NPs laterally on the polyimide substrate (1 mm thick), respectively. Two ends of a MW fixed by silver paste served as source and drain electrodes, respectively, and an M-S-M structure device was formed. A thin layer of polydimethylsiloxane was used to package the device, which not only enhanced the mechanical robustness of the whole device but also prevented the degradation of the ZnO MW by gas or moisture in the environment.

The morphology and crystalline structure of the as-deposited ZnO MWs were characterized by a SEM (FEI Nova Nano-SEM 450) and TEM (FEI Titan G2 60-300). UV photo-response measurement was carried out between two electrodes at a fixed bias voltage of 8 V by switching the light from a portable UV lamp ($\lambda=365\text{ nm}$, 0.3 mW cm^{-2}) “on” and “off”.

Results and Discussion

Figures 1a-c show the working schematics of a traditional Junction field-effect-transistor (JFET). For an n-channel JFET, two P-type doped regions are used as grid electrode and both

ends of n-type semiconductor are used as drain and source; the current flowing from the drain to source under an applied external voltage U_{DS} is controlled by the gate voltage U_{GS} , resulting in the change of the channel width for transporting the carriers.

Analogously, Figure 1d shows the schematics of the piezoelectric transistor with metal-ZnO-metal structure, which formed SB at the electrode interface. When applying a strain, the created piezoelectric charges at the interface region control the carrier transport at the metal/ZnO interface, which forms a piezoelectric transistor. Thus, the externally applied gate voltage U_{GS} in a traditional JFET is replaced by an inner crystal potential in this piezoelectric transistor, which means that the piezoelectric transistor only needs external drain and source terminal. The controlling channel width in a traditional JFET is replaced by controlling the interface *via* inner crystal potential, and the current transported across the interface is the exponential of the local barrier height in the reversely biased case, the on/off ratio can be rather high due to the nonlinear effect. Thus, a voltage controlled device is replaced by a strain-controlled device *via* varying the local barrier height.[24] In Figure 1d and e, the external stimulus can be strain/light, which can increase/decrease the depletion region.

Figure 1e is the schematic of the Au NPs decorated piezoelectric transistor with metal-ZnO-metal structure. As we stated above, the Au NPs are treated as a lot of SBs on the surfaces of ZnO, which can improve performance of the device. Our previous studies proved that the coupling between piezoelectric potential and numberd SBs (by Au NPs decoration) at the ZnO surface can much enhance the sensitivity of the piezoelectric devices.^[23] For the Au NPs decorated piezoelectric transistor, the electrical transport properties of the device will be controlled more effectively by both the SB at the electrode contact and numberd SBs on the surface. The coupling among the characteristics of piezoelectricity, photoexcitation and semiconductor are enhanced because the numbered decorated Au NPs on the ZnO surface play the role of SBs, which supplies new and important control for field-effect-transistor (FET). Moreover, the input of the external stimulus can be also light which change the SB

height and the carriers' density. Figure 1f show the schematics of the logic units corresponding to our work, there are two kinds of stimulus (UV light and strain) corresponding to the (L)SGTs.

Figure 1. The working mechanism of the devices. (a), (b), (c) Schematics of a traditional JFET. (d) Schematics of a piezoelectric transistor with metal-ZnO-metal structure. (e) Schematics of the Au NPs decorated piezoelectric transistor with metal-ZnO-metal structure. In (d) and (e), the external stimulus can be strain/light, which can increase/decrease the depletion region. Then piezotronic and piezo-phototronic logic computations were realized, as shown in the schematic of the logic unit (L)SGTs in (f).

The single crystallinity of the ZnO MWs has been confirmed, and more details of the interface area between Au NPs and ZnO MW were shown by a high resolution transmission electron microscopy (HRTEM) image in Figure 2a. There are two distinct sets of lattice fringes at the interface, one of the lattice spacing of 0.52 nm is attributed to the [0001] planes

of the hexagonal wurtzite ZnO crystal[25] and the other of the 0.235 nm is to the Au (111) planes.[26] As shown in typical SEM image of Figure 2b, an M-S-M structure device was formed, two ends of a Au NPs decorated ZnO MW fixed by silver paste served as source and drain electrodes, respectively. The Au NPs are uniformly and discretely distributed on the surface of ZnO wire, as shown in the right part of the Figure 2b. Figure 2c is a picture of a flexible SGT which can be implemented periodic compressive and tensile strain.

Figure 2. The microstructure of the Au NPs decorated ZnO MW. (a) A high resolution TEM image of the Au NPs decorated ZnO MW. (b) SEM images of an Au NPs decorated ZnO MW based flexible SGT. The right part of (b) show more details of the uniform distribution of Au NPs on the surface of ZnO wire. (c) The picture of a flexible SGT on which strain can be implemented.

Once the substrate was bent, a compressive/tensile strain was created in the MW because the mechanical behavior of the entire structure was determined by the substrate. The magnitude and the sign of applied strains can be calculated following the method reported elsewhere.[27] Figures 3a (without Au NPs) and 3b-c (with Au NPs: b is tensile strain and c is the compressive strain) show the device's drain-source current in the dark condition as a function of voltage under various applied strain (the tensile/compressive strains are defined as positive/negative, respectively). **Generally, the nonlinear behavior shown in figure 3a, b and c should be related to the SB formed between the ZnO and the silver paste electrodes,[28, 29]**

and the shape of the I-V curve depends on the heights of the SB height, which functions as the controlling input signal to gate the electrical transport in the SGT.

It is obvious that the dark current experienced a weak decrease from the state of tensile strain to the state of compressive strain. The reason for the decreased dark current of the original ZnO device was analyzed as following. When a SGT is under strain, two typical effects may affect the carrier transport process. One is the piezoresistance effect because of the change in bandgap, charge carrier density, and possibly the density of states in the conduction band of the semiconductor crystal under strain. This effect is a symmetric effect on the two ends of the contacts without polarity and producing the function of a device.[1] The other reason for the decreased dark current of the original device was analyzed by piezoelectric effect: when compressive strain is introduced, both the band structure and piezoelectric potential would contribute to the rise in the SB height at the electrode contact, resulting in the further upward bent of conduction (E_c) and valence (E_v) bands at the surface of the ZnO MW.

For the device with Au NPs decoration (Figures 3b and c), the dark currents in figure 3a (without Au) is 4.58×10^{-4} A and that in figure 3b (with Au) is 1.04×10^{-4} A at the fixed bias of 20 V under the equivalent strain-free states. After covering with Au NPs, the negatively charged Au NPs depletes the carriers near the surface of ZnO NW, and numbered SBs are formed at the Au and ZnO interface.[23] Since the width of depletion layer is related to barrier height, depletion region near the Au NPs is larger than the depletion region far from the Au NPs. The formation of the large depletion region is the main reason for the decrease in the dark current after covering with Au NPs. The above working principle of the Au NPs decorated ZnO surface can be explained using band diagrams shown in supplementary information Figure S1. Once the substrate was bent, it is obvious that the dark current experienced a drastically decrease from the state of tensile strain to the state of compressive strain. The reason for the more decreased dark current is explained by using Au NPs

introduced SBs as stated above, after introducing strain, the SB height is increased much higher under equivalent compressive strain due to more SBs induced by Au NPs, and thereby the current has a sharp decrease as compared with the original ZnO MW as we described in Figure 3a. However, when a tensile strain is introduced, the SGT's band structure changes and opposite polarity piezoelectric potential generates, [6, 19] and at last the depletion region width and SB height decrease. Thus, the I-V curves shift upward with tensile strain and downward with compressive strain. **The above our experiments prove that the localized SBs at the ZnO surfaces can help improve the piezo-performance.**

As shown in Figure 3d, the photocurrent of the Au NPs decorated ZnO MW SGT also increases as the applied tensile strain increases and reduces as the applied compressive strain increases. The increase of photocurrent is explained as follows, when the device is illuminated by UV light, the stronger and expanding built in field enhances the spatial separation effect, and increases the photo-response gain.[30] The experiments demonstrated that optical illumination with appropriate energy and intensity can therefore modulate the conductivity of the ZnO MW device and turn it into electrically “on” state.

Furthermore, as shown in Figure 3e, the Au NPs decoration SGT was measured under various applied strain for several cycles. The current descend/rise by almost the same value in each cycle of straining and fully recover, being very similar to other literatures.[18, 19] For the ZnO MW SGT, the external strain (ϵ_g) acts as the gate input for controlling the on/off state of the SGT. Figure 3f, the drain/source current strain ($I_{DS}-\epsilon_g$) curves of the SGT under different bias voltage show that the current increases as the applied tensile strain increases and reduces as the applied compressive strain increases, where the threshold gate strain ϵ_T is around -0.368%. The on and off state for the SGT can be determined as the values obtained at $\epsilon_g(\text{off}) = \epsilon_T - 0.3\%$ and $\epsilon_g(\text{on}) = \epsilon_T + 0.7\%$. $I_{\max}$ (strain of 0.98%) = 0.3 mA and $I_{\min} = 3.23$ nA (strain of -0.81%) are hence obtained with the $I_{\max}/I_{\min}$ ratio of 9.3×10^4 at about 5 V fixed bias, which is much higher than the research. [19]

Figure 3. The I-V, I-T characteristics and of various devices. (a), (b) and (c), Typical I-V characteristics of the SGT at different strains without (a) and with (b, c) Au NPs. (b) is in tensile strain and (c) is in compressive strain. (d) Photocurrent of the Au NPs decorated ZnO MW SGT. (e) Current response of the Au NPs decorated SGT under alternative compressed/tensile strain at a fixed bias of 7.5 V. (f) I_{DS} - ϵ_g curves of the typical ZnO MW SGT measured at different fixed bias about 2 V, 5 V, 7.5 V and 10 V, the inset showing the detail of the compressive strain.

Two series of experiments were carried out to investigate the coupling of local SBs and piezoelectric effects on the performances of the piezoelectric strain gated inverters (SGIs) with and without Au NPs. We fabricated an inverter with two SGTs both on the top and bottom surfaces of the substrate, as shown in Figures 4a and c. The two SGTs have absolutely contrary on/off state, depending on whether the substrate is bent upward or downward. For the SGI logic unit, the strain input logic “1” is defined as: the upside #1 SGT is “off”-compressive and the downside #2 SGT is “on”-tensile, leading to the electrical output logic “0”-a lower voltage, while the strain input logic “0” is defined as: the upside #1 SGT is “on”-tensile and the downside #2 SGT is “off”-compressive, leading to the electrical output logic “1”-a higher voltage. These definitions are adopted in all demonstrations here and afterwards.

Figure 4. Schematics of two ZnO MW piezoelectric SGIs without (a) and with (c) Au NPs decoration performing logic operations in response to the strain inputs. Experimental output voltages with the strains of the ZnO MW piezoelectric SGIs without (b) and with (d) Au NPs.

The strain voltage transfer characteristic of the two SGIs were obtained by plotting the output voltages versus the applied external strains, as shown in Figures 4b and d. V_{OH} and V_{OL} represent the high and low output voltage of the SGI, with theoretical values of $V_{OH} = V_{bias} = 10$ V and $V_{OL} = 0$ V. Here, the logic swing of the two piezoelectric inverters are defined as $V_{OH} - V_{OL} = 8.25$ V - 1.79 V = 6.46 V (without Au) and 9.99 V - 0.002 V = 9.988 V (with Au NPs) according to the experimental values, respectively. The difference between experimental and theoretical values was probably caused by the voltage drop across the SGT which was “on” during the measurement for the two SGIs. Figure 4b shows that the original ZnO MW based SGI has a lower V_{OH} and a higher V_{OL} , and the output voltages were restricted to a small range, which blocks its practical application. For the Au NPs decorated SGI with a higher V_{OH} and a lower V_{OL} , when the applied strain is in the region of $\epsilon_g <$

0.48%, the electrical output is logic “1”; when the applied strain is in the region of $\varepsilon_g > 0.48\%$, the electrical output is logic “0”. It is obvious that the Au NPs decorated SGI is easier to implement logic transformation than the original ZnO SGI. The difference between the SGIs with and without Au NPs decoration were caused by controlling the electric transport process of the ZnO more effectively due to localized SBs introduced.

Figure 5. Strain controlled piezoelectric logic gates and some basic operations based on Au NPs decorated ZnO MWs. (a), (b) and (c): piezoelectric logic NAND gate. (d), (e) and (f): piezoelectric logic NOR gate. (a), (d): Schematic of the Au NPs decorated ZnO MW piezoelectric NAND and NOR gate. (b), (e) Shows the measured electrical output voltages of NAND and NOR gate versus the input strains (time). (c), (f) Experimental truth table of the output voltage with the two SGIs at various strain inputs.

More universal piezoelectric logic operations were obtained by systematically integrating the Au NPs decorated ZnO MW SGIs. According to corresponding connection rules (Figures 5a, d), the measured output voltages of the NAND and NOR gate are plotted in Figures 5b, e. Red lines are the electrical output voltages of logic gate; blue and green lines represent the strain input applied on SGIs A and B, respectively, where strain input “0” and “1” are defined

in Figure 4c. The output and input signals are listed in the truth table in Figures 5c, f. In a similar manner, piezoelectric strain controlling AND and OR gate have also been realized by integrating SGIs, as presented in Figure S2.

Strain-electricity coupling can also be used to design logic gates. Figure 6 shows an XOR gate and 2:1 MUX. For the XOR (Figure 6a), the drain electrode of #1 SGT is connected to the electrical input V_o while the drain electrode of #2 SGT is connected to $V_{\bar{o}}$, which is the logically compensate electrical input to V_o . The strain gated input logic for #2 SGT and #1 SGT also compensate each other. The change in the connections of the electrodes from those demonstrated in the SGI results in different logic functions. When the substrate is bent upward or downward, the electric output would be either “1” or “0”. For the XOR gate shown in Figure 6a, in the upside state, the electrical output is logic “0” when the strain input is logic “1” and the electrical input V_o applied on #1 SGT is logic “1”; while in the downside state, the electrical output is logic “1” when the strain input is logic “0” and the electrical input V_o applied on #1 SGT is logic “1”. As a supplementary description without the schematics here, the electrical output is logic “1” when the strain input is logic “1” and the electrical input V_o applied on #1 SGT is logic “0”; while the electrical output is logic “0” when strain input is logic “0” and the electrical input V_o applied on #1 SGT is logic “0”. All four output voltages of the XOR gate versus the input gate strains and electrical input are shown in Figure 6b.

The 2:1 MUX was realized according to corresponding connection rules in Figure 6c, where the drain electrodes of #1 SGT and #2 SGT are connected independently to two square wave signals with different frequency (input 1 with 0.5 Hz and input 2 with 1 Hz) rather than logically complements. For the upside state of Figure 6c, #1 SGT is “off” and #2 SGT is “on”, hence the electrical output is determined by #2 SGT and obtained frequency is 1 Hz. For the downside state of Figure 6c, #1 SGT is “on” and #2 SGT is “off”, hence the electrical output is determined by #1 SGT and the obtained frequency is 0.5 Hz. The measured electrical

output voltages and input signals with the two states are listed in the Figure 6d. Analogously, by expanding the 2:1 MUX, an n:1 MUX based on the same mechanism as above can be obtained, which enables us to pick one of the n inputs and direct it to the output.

Figure 6. Different from the above gates in Figures 4, 5 based on strain coupling, the gates are based on strain-electricity coupling. (a) and (b): Piezoelectric logic XOR gate. (c) and (d): 2:1 MUX. (a) Schematics of an Au NPs decorated ZnO MW XOR logic gate performing logic operations on strain and electrical inputs. (b) Logic operations and experimental truth table of the ZnO MW strain gated XOR logic gate. Red line is the electrical output of the XOR gate. Blue and green lines represent the electrical and strain inputs applied on #1 SGT and #2 SGT, respectively. (c) Schematics of an Au NPs decorated ZnO MW 2:1 MUX performing logic operations on two square wave signals with different frequency inputs. (d) The measured electrical output voltages in the two states.

We have further studied the piezo-phototronic effect in two terminal Au NPs decorated ZnO MW SGT by introducing both light and strain to the device. The “dual gate” programmable composed of four LSGTs. Each of these four LSGTs has two independently configurable gate controlling signals L_{in} and ϵ_{in} for light and strain inputs, respectively. The output from these logic units is electrical voltage. In the demonstration, the logic “0” state for L_{in} is defined as focusing the 365 nm UV laser with intensity of 0.3 mW cm^{-2} on #2 and #4 LSGTs; the logic “1” state is defined as focusing the UV laser on #1 and #3 LSGTs. The logic “0” state for ϵ_{in} is defined as applying -0.98% compressive strain on #1 and #2 LSGTs; the logic “1” state is defined as applying -0.98% compressive strains on #3 and #4 LSGTs. Figures 7a, d and g illustrate the configurations of piezo-phototronic AND, OR, and XOR logic gates comprising of four LSGTs, where both L_{in} and ϵ_{in} are logic “0”.

These piezo-phototronic logic operations are experimentally verified as shown in Figures 7b, e and h, with low voltage output (logic “0” output) at around 0 V and high voltage output (logic “1” output) at around 10 V, which is the bias voltage V_{bias} . Red lines are the experimentally measured electrical output voltages of the logic gates, while green and blue lines represent the logic levels of input strains and UV laser, respectively. The truth table of the input and output signals are presented in Figures 7c, f and i. Other piezo-phototronic universal logic units, including NAND and NOR logic gates, were also successfully demonstrated (Figure S3).

Figure 7. Different from the above gates in Figures 4-6, the gates are based on strain-light coupling, named piezo-phototronic logic computations. (a), (b) and (c) Piezo-phototronic logic AND gate. (d), (e) and (f) Piezo-phototronic logic OR gate. (g), (h) and (i) Piezo-phototronic logic XOR gate. (a), (d), (g) Schematic configurations of a piezo-phototronic logic AND, OR, XOR gates comprising of four LSGTs, taking optical and strain inputs combination (0,0) as a demonstration with piezo-potential distribution in the MWs highlighted using color code. (b), (e), (h) Electric output signals of an AND, OR, XOR gates under four input states as labeled, $V_{DD}=10$ V. (c), (f), (i) Experimental truth table of AND, OR and XOR gates.

Conclusions

In summary, by using numbered SBs formed at ZnO surface, the highly enhanced piezotronic and piezo-phototronic couplings have been applied for logic computations to design SGTs and LSGTs, which process mechanical and optical stimuli on the devices into electronic controlling signals. Two series logic devices such as inverter, AND, OR, NAND, NOR, XOR

gates with good rectifying behaviors and 2:1 MUX have been fabricated. The piezo-phototronic logic devices based on Au NPs decorated ZnO MWs may find applications in active flexible electronics, prosthetics, optical micro/nanoelectromechanical systems, tunable bio-optoelectronic probes, and adaptive optical computing.

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Graphical Abstract

Piezotronic and Piezo-phototronic Logic Computations Using Au Decorated ZnO Microwires