Hybrid Porphyrin–Silicon Nanowire Field-Effect Transistor by Opto-Electrical Excitation

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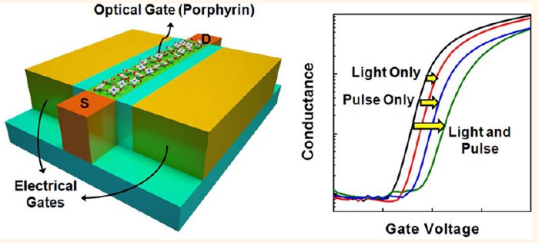
ABSTRACT



A porphyrin–silicon nanowire (Si-NW) hybrid field-effect transistor is introduced. The hybrid device has separate electrical and optical gates surrounding the Si-NW channel. Porphyrin, a component of chlorophyll, is employed as an optical gate to modulate the potential of the Si-NW channel. Due to the independently formed hybrid gates, both optical and electrical excitation can effectively modulate the device. The exposed porphyrin optical gate responds to the optical excitation, and independently formed electrical gates respond to the electrical excitation. Charge transfer characteristics between a semiconductor channel and the porphyrin optical gate are deeply investigated. Optical, electrical, and opto-electrical excitation methods are employed to analyze the charging and discharging behaviors. Of these methods, opto-electrical excitation enables the strongest charge transfer because the inversion electron formation by an electrical pulse and the photoinduced charge transfer by an optical stimulus are affected simultaneously. Discharging processes, such as rapid discharging, exponential detrapping, and the formation of metastable states are also analyzed.

KEYWORDS: organic silicon hybrid device · nanowire · field-effect transistor · porphyrin · photoinduced charge transfer · opto-electrical excitation

Organic silicon hybrid devices have been investigated recently to combine the advantages of silicon devices and organic materials. The silicon field-effect transistor (FET) is advantageous because of its small scale, high performance, and mature fabrication technology. Extremely small devices with high carrier mobility and small variability can be achieved through well-developed silicon fabrication technology.[1–4] Additionally, organic devices have advantageous optical properties and various functionalities. Organic light-emitting diodes (OLEDs) and organic photovoltaic cells (OPVCs) are examples that use their optical properties.[5,6] The advantages of both the silicon and organic devices can be merged in organic silicon hybrid devices.[7–14] The silicon nanostructure forms a framework for a hybrid device on which the organic materials are then functionalized. The organic molecules immobilized onto the stable frame device can be used for achieving optical sensitivity and diverse functionality. The organic silicon hybrid device is a convincing candidate that overcomes the limits that a conventional pure device faces. Various applications such as nonvolatile memory,[7–9] chemical sensors,[10] optical switches,[11] optical memory,[12] and optical inverters[13] have been introduced.

The basic operation principles of hybrid devices are based on the electron transfer between a silicon body and organic compounds. Both electrical excitation alone and optical excitation alone have been used to control electron transfer. Electrical excitation uses a strong instantaneous gate voltage pulse to confine inverted electrons inside the p-type body, which causes organics to trap the electrons.[7–9] Optical excitation uses external light to activate the organics to play a role in photoinduced charge transfer (PCT).[11–13] The photoactive organics can serve either as the electron donor or as the electron acceptor during illumination.

Previous reports have shown various interesting phenomena and important analyses; however, hybrid devices are still in an exploratory stage. Various unknown phenomena regarding charge transfer are not completely understood, and most of the structures proposed thus far are not ready for practical applications. In particular, the excitation method has not been thoroughly investigated, despite the fact that excitation has long been employed to characterize

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hybrid devices. Different excitation methods rely on different charging mechanisms even for the same type of organic material. Furthermore, the structural development of a hybrid device requires additional research. Most hybrid devices are based on a global back-gate, which cannot be applied in practical cases. A prototype of an individually gated hybrid device was introduced recently in which a partial nanogap is inserted into the gate insulator region; however, a few problems remain, such as a shadowing effect of the top-gate and poor structural instability.[12]

In this paper, the new design of a porphyrin–silicon nanowire (Si-NW) hybrid field-effect transistor is introduced. The proposed hybrid FET has a multi-gate structure composed of double electrical gates and a single optical gate so that the device can be modulated by both optical and electrical excitations. The optical gate is formed with porphyrins, which are a crucial photoactive component of chlorophyll. The proposed hybrid device is useful for analyzing charge transfer because a small variation in the charge state results in a large change in conductance. The charging and discharging processes for various excitation methods (*e.g.*, electrical, optical, and opto-electrical excitation) are evaluated.

Porphyrin is a special type of organic molecule. As an element of chlorophyll, porphyrin charges and transfers electrons during photosynthesis. Researchers have analyzed both porphyrin–silicon interfaces and porphyrin-related hybrid devices.[11–15] Porphyrins can act as either the electron donor or the electron acceptor, depending on the presence of a central metal atom.[11–13] Porphyrin is a a promising candidate for organic silicon hybrid devices because of their clear, reliable optical characteristics.

RESULTS AND DISCUSSION

Figure 1 shows a schematic and images of the fabricated device. A Si-NW is located between double electrical gates, and the source and drain are placed at the end of the NW (Figure 1A). The nominal width and height of the NW are 40 and 100 nm, respectively, and the thickness of the gate insulator is 30 nm (Figure 1B). The geometric parameters can be modified by controlling the fabrication conditions. A 3 nm layer of silicon dioxide (optical gate oxide) was placed between the Si-NW and the subsequently immobilized porphyrins. The optical gate oxide serves as a tunnel barrier between the porphyrin and the Si-NW, whereas the electrical gate oxide serves as a capacitive insulator to electrically modulate the potential of the Si-NW. The 3 nm thickness of the tunnel barrier effectively facilitates the electron charging and discharging processes.[12,17] Thereafter, the porphyrins were immobilized onto the top surface of the optical gate oxide, which straddled the Si-NW. The porphyrins were agglomerated in an amorphous state (Figure 1C). Electron

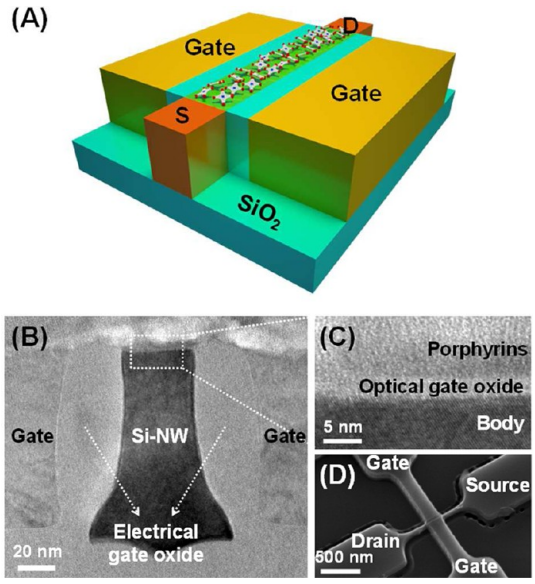

Figure 1. (A) Schematic of the hybrid nanowire FET. (B) Cross-sectional TEM image of the hybrid nanowire FET. (C) Magnified view of the top of panel B. (D) Bird's-eye SEM image of the hybrid nanowire FET.

dispersive spectrometry (EDS) analysis confirmed that the amorphous layer was porphyrin (see Figure S3 in the Supporting Information). The gate length was 200 nm with no undesirable short-channel effects because the channel potential of the Si-NW was tightly controlled by the double gates (Figure 1D).

The device was operated by modulating the channel potential in the p-type Si-NW (*i.e.*, n-channel). The charge states of the porphyrins affect the density of the inversion electrons, which determines the conductance of the Si-NW channel. The charge states of the porphyrins are controlled by external excitations. Optical excitation is a common method to control the charge state of the porphyrins. Porphyrins gradually absorb electrons during light illumination and become negatively charged.[12,13] The negatively charged porphyrins push electrons away from the surface of the Si-NW, thereby preventing the surface inversion electrons (*i.e.*, electrons in the p-type body) from being confined to the outer surface. In other words, the position of the conduction band is elevated as a result of the negatively charged porphyrins, which blocks the current paths between the source and drain. When the current paths become inhibited, a large gate voltage is required to make the same amount of current. As a result, the transfer curve shifts to the right after the electron charging process.

The operation principle was visualized by three-dimensional graphs using a commercial simulation tool (ATLAS/Silvaco).[18] Porphyrins were modeled as a floating gate placed on the Si-NW.[12] The graphs showed the energy level of the conduction band edge, which determines the amount of inversion electrons in the Si-NW. When there was no charge in the

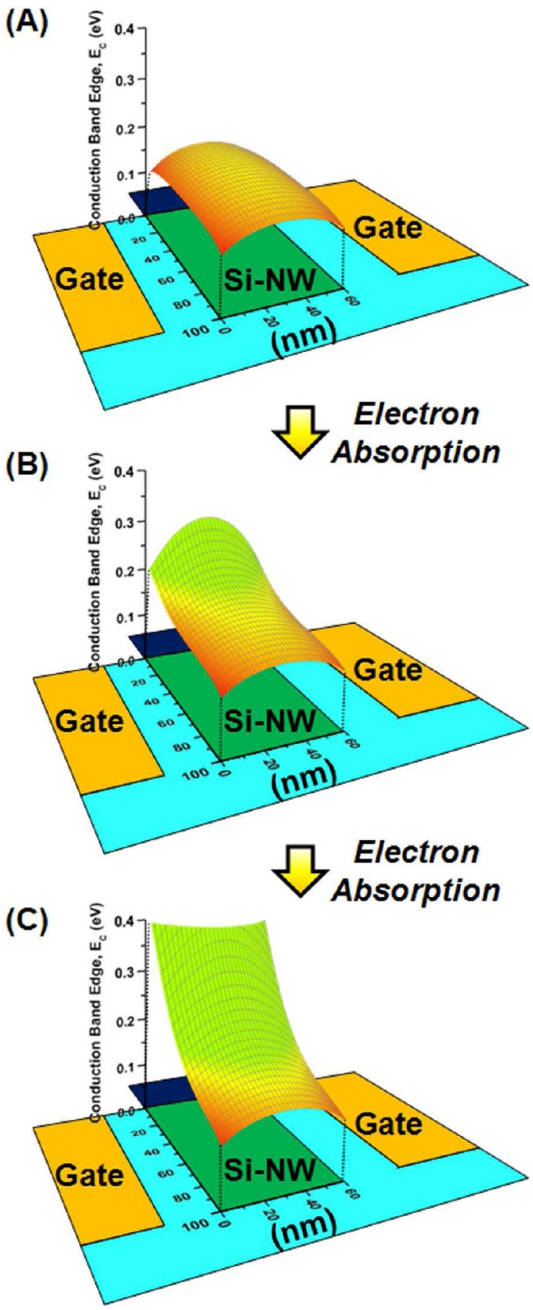

Figure 2. (A) Three-dimensional simulation graph of the conduction band energy when no charge is introduced into the porphyrins. (B) Conduction band energy when a small amount of charge is introduced into the porphyrins. (C) Conduction band energy when a large amount of charge is introduced into the porphyrins. The gate voltage is 0.2 V for all simulations.

porphyrins, the conduction band level was low throughout the Si-NW, which allows large numbers of inversion electrons to be formed (Figure 2A). Electrons from the source can be easily transported to the drain under this condition, resulting in high conductance. When porphyrins absorb electrons and become negatively charged, the conduction band level becomes elevated (Figure 2B,C). The elevation of the conduction band was stronger at the upper part of the Si-NW, which is close to the porphyrins. Electron channels are more inhibited by elevated conduction bands, which decrease the conductance in this case. These data suggest that the change of conductance and the shift of the transfer curve should be larger for more strongly charged porphyrins.

The electron absorbing behaviors during optical excitation can be described with a simple schematic (Figure 3A). For an n-channel FET, the source and drain are n-type, while the Si-NW is p-type. During illumination, porphyrins absorb electrons from the silicon body; however, there are not enough electrons to completely charge the porphyrins because the electrons are minor carriers in the p-type Si-NW. The light-induced electron−hole pair generation phenomenon is also negligible because the volume of the nanowire body is too small.[13] The charging effect that arises from the PCT was confirmed by the shift of the transfer curves (Figure 3C). The transfer curve was shifted to the right after optical excitation, as expected. A stronger gate voltage was required to create the same amount of inversion electrons because the current paths were blocked off because of the negatively charged porphyrins. The magnitude of the shift was saturated as the light's intensity increased. Thus, optical excitation alone cannot induce a large electron transfer.

Electrical excitation, which is an alternative method for charging porphyrins, is described by a simple schematic (Figure 3B). When a voltage higher than the threshold voltage (V_T) is applied to the double electrical gates in a dark ambient environment, a large inversion electron density (greater than 10^{12}/cm^2 or 10^{19}/cm^3) is induced at the outer surface of the Si-NW, which sufficiently countervails the background hole density ($\sim 10^{17}$/cm^3). Therefore, the Si-NW develops an electron-rich state, and some of the inversion electrons are dispersed toward the top surface of the Si-NW by the fringing field from the gates. This fringing field forces the electrons to move upward from the Si-NW and traps them within the porphyrins, which causes the porphyrins to become negatively charged and shifts the transfer curves to the right (Figure 3D). The curve shifts were larger with stronger gate voltages. However, the application of a voltage greater than 9 V causes the electrical gate oxide breakdown, which imposes a constraint on the maximum magnitude of the gate voltage by electrical excitation alone. Therefore, the magnitude of the shift saturated in a manner similar to that following optical excitation.

To overcome the aforementioned limitations, a new excitation method, namely, opto-electrical excitation, is proposed (Figure 4A). A strong gate pulse and optical illumination were simultaneously applied to synergistically strengthen the electron transfer behavior. Porphyrins can effectively absorb electrons through the PCT process because enough electrons were supplied

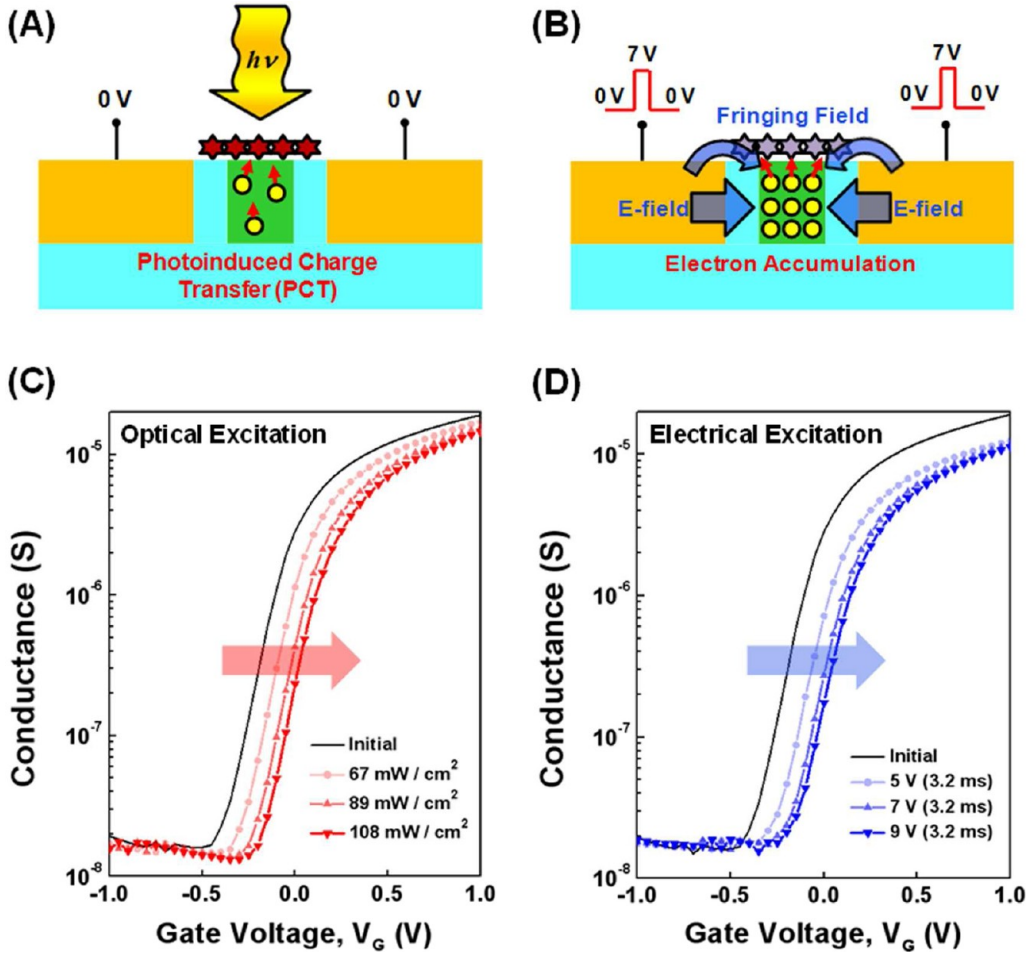

Figure 3. (A) Schematic that describes the principle of optical excitation. (B) Schematic that describes the principle of electrical excitation. (C) Transfer characteristics (*I*–*V* curve) before and after optical excitation. White light was used as the light source with an illumination time of 20 s. (D) Transfer characteristics before and after electrical excitation. The measurements were conducted in ambient darkness.

by the gate bias as inversion charges. The shift of the current–voltage (*I*–*V*) characteristics was clearly enlarged, even under a weak light intensity and small gate bias (Figure 4B). This amount of shift surpassed the saturated shift values that result from optical excitation or electrical excitation alone. A 4-fold larger curve shift was achieved under the same light intensity compared with optical excitation alone with a 67 mW/cm^2 intensity. An advantage of the proposed hybrid FET structure is its simultaneous opto-electrical excitation because the vertical double-gate channel can harness the electrical excitation, while the planar top channel favors the optical excitation because of the nature of the uncovered surface by the gate at the top of the Si-NW. The device parameters, such as an on–off current ratio and a subthreshold slope, were not degraded after excitation; that is, only parallel shifts occurred. Control experiments confirmed that the shifts of the transfer curves could be attributed to the charged electrons in the porphyrins rather than to other environmental effects (see Figure S4 in the Supporting Information).

The sustainability of the stored charges after excitation is an important concern. Absorbed electrons in the porphyrins were emitted back to the Si-NW after excitation. This discharging property was analyzed using a real-time transient curve (Figure 5A). The conductance sustained its initial value before the excitation. Right after the illumination, the conductance decreased because the current paths were blocked off. After the sudden decrease, the conductance rapidly returned to near its initial value as the electrons were emitted back to the Si-NW. This rapid discharging process occurred for approximately 2 min after the excitation. During a long illumination time, charging and discharging offset each other, thus creating an equilibrium state. After the light is turned off, discharging is still activated; however, charging by PCT is terminated. Therefore, weakly trapped electrons are rapidly emitted back to the Si-NW, allowing the decreased conductance to recover until it approaches its initial value.

Interestingly, the conductance did not completely return to its initial value. Some fraction of the electrons

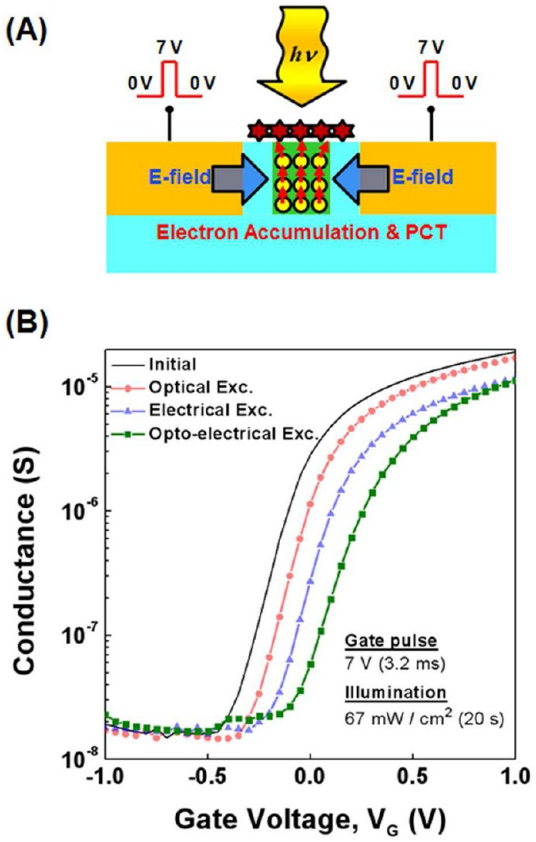

Figure 4. (A) Schematic describing the principle of opto-electrical excitation. (B) Transfer characteristics before and after optical, electrical, and opto-electrical excitations. The optical source was 67 mW/cm^2 white light applied for 20 s, and the gate voltage was 7 V applied for 3.2 ms.

in the porphyrins remained in the porphyrins even after several days. This semipermanent charged state is called a metastable state, which is caused by deeply trapped electrons in organic materials.[19,20] Unlike single-crystalline inorganic semiconductors, organic material usually exists in an amorphous state, which inherently creates a large number of trap sites. Some of the traps act as deep traps, which are located in a midgap energy level of the organic semiconductor. After the charges are trapped in the deep traps, it is difficult to detrap them unless a strong external electrical field is applied.[19] The density of the deep traps can be amplified due to the lattice distortion in an organic layer. Therefore, the metastable state is more clearly observed for an optical signal as compared to an electrical signal. The metastable state can be advantageous for the charge-retaining applications, such as optical memory. Conventional flash memory unavoidably uses a high gate bias (15−20 V) to enable carrier tunneling from a channel to a charge-trapping layer. This inevitably causes damage to the gate dielectric, and the large power consumption is problematic. However, the proposed optically assisted program method uses a program voltage of less than 7 V, and the damage to the gate dielectric is greatly reduced.

The same real-time optical charging experiment was conducted for a p-channel FET (PMOS). The source and drain are heavily doped by boron, and the Si-NW channel is lightly doped by arsenic. All other device parameters and experimental conditions are identical, except for the doping polarity. Opposite to the n-channel FET, the conductance increases upon illumination. This opposite shift of the conductance is caused by the different number of electrons in NMOS and PMOS samples. When porphyrins are attached to the p-type Si-NW in the NMOS sample, negatively charged porphyrins prevent the formation of inversion electrons. Electrons from the source cannot easily be transported to the drain, causing the conductance to be decreased. However, when porphyrins are attached to an n-type Si-NW in a PMOS, negatively charged porphyrins aid the formation of inversion holes. Holes from the source are readily transported to the drain, thus increasing the conductance.

The discharging process of the PMOS sample after illumination was similar to that of the NMOS. The increased conductance rapidly returned to a value close to the initial value in the first 2 min. The conductance did not completely return to its initial value, and the sample remained in a metastable state. The difference between the initial conductance and the metastable conductance was approximately twice that of the NMOS sample. As described in Figure 3A,C, the conductance shift by optical excitation is limited by the number of the electrons in the Si-NW body. Because an n-type Si-NW was used for the PMOS sample, there were more chances for electrons to transfer from the Si-NW to the porphyrin. Therefore, more electrons can fill deep traps in the porphyrin layer, which results in a large Δg, as shown in Figure 5B.

This discharging process was also characterized by curve shift analyses (Figure 5C). After the excitations, a rightward-shifted transfer curve tended to return back to the initial state, so the curve shift value decreased. In optical excitation, a rapid discharging phenomenon was seen during the first 2 min. The rapid discharging process was also observed after opto-electrical excitation but was not observed after pure electrical excitation. After electrical excitation, the curve shift value was maintained for approximately 20 min before it exponentially decayed, similar to the discharging process of inorganic-based flash memory, such as silicon-oxide-nitride-oxide-silicon (SONOS) memory.[21] The reason for this similarity is that the compulsive trapping method, which employs an instantaneous gate pulse, is similar to the program method of inorganic-based flash memory. Opto-electrical excitation also showed an exponential decay trend due to the effect of the trapped electrons.

The metastable state occurred in both the optical and opto-electrical excitation states; however, the amount of shift was notably larger for the opto-electrical excitation.

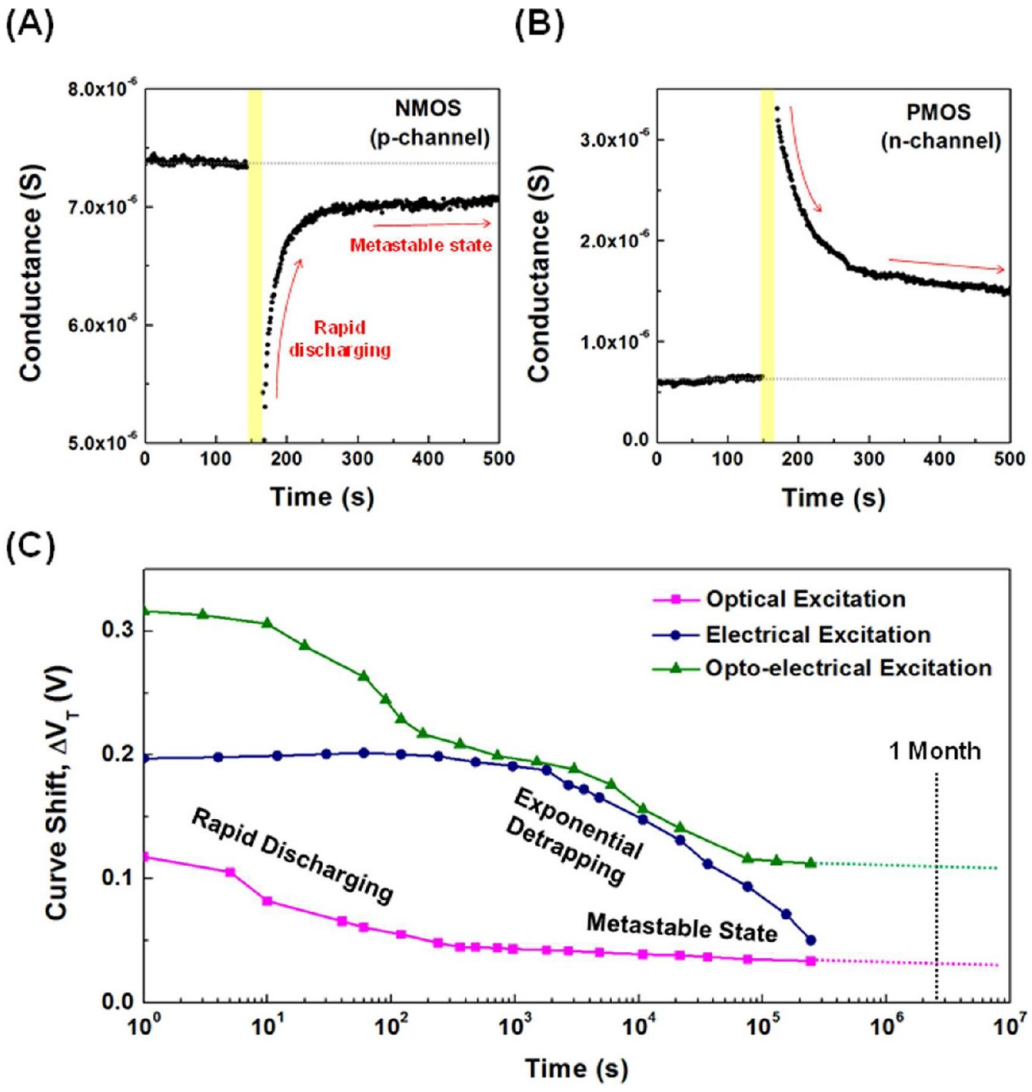

Figure 5. (A) Real-time discharging characteristic of n-channel MOSFET (NMOS) after optical excitation. Gate voltage was 0.5 V, and drain voltage was 0.05 V. Optical intensity was 67 mW/cm^2. (B) Real-time discharging characteristic of p-channel MOSFET (PMOS) after optical excitation. Gate voltage was −1.0 V, and drain voltage was −0.05 V. (C) Amounts of curve shift after optical, electrical, and opto-electrical excitations using 67 mW/cm^2 white light for 20 s as the optical source and 7 V for 3.2 ms as the gate pulse. The threshold voltage (V_T) was read at a drain current of 100 nA (*i.e.*, conductance of 2×10^{-6} S).

Although the density of the deep traps in the porphyrins was the same under both excitations, a larger amount of electrons can be inserted into the trap sites in the porphyrins because of inversion electrons. Even with similar amounts of curve shifting, electrical excitation alone cannot generate a metastable state. Because electrical excitation is based on compulsive charge movement using the short-term pulse duration of the external electric field, trap-level redistribution in the porphyrins does not occur. Therefore, deep traps cannot be sufficiently generated to cause a noticeable metastable state. For optical excitation, the metastable state occurred immediately after the rapid discharging process, while it occurred after the exponential decay process for the opto-electrical excitation. The opto-electrical excitation exhibited the combined charge sustainability of the optical and electrical excitations.

An interesting advantage of porphyrin is that a metal atom can be placed at the center of the porphyrin molecule. We prepared a Cu(II) meso-tetra(4-sulfonatophenyl)porphine molecule (Cu-porphyrin) to confirm the effect of the centered metal. Other chemical structures are identical to those used with the unmetalated porphyrin (H_2-porphyrin), except for the center metal atom. In previous research on porphyrin−In_2O_3 NW hybrid devices, improved redox behavior in a metalated porphyrin was demonstrated during pure electrical excitation, but the optical properties of the porphyrin were not considered.[8] The charge transfer characteristics with different excitation methods are compared in Figure 6A. For all three excitation methods,

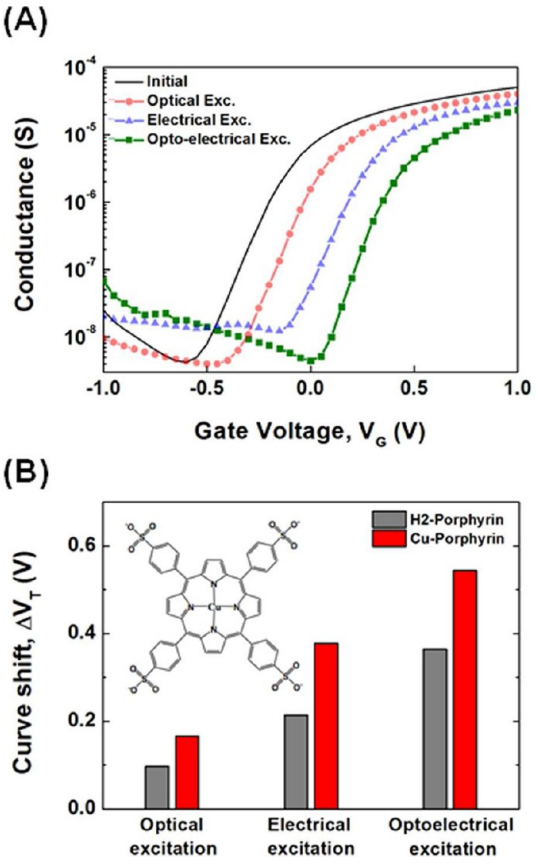

Figure 6. (A) Transfer characteristics before and after optical, electrical, and opto-electrical excitations when Cu-porphyrin was used for charged source. The optical source was 67 mW/cm2 white light applied for 20 s, and the gate voltage was 7 V applied for 3.2 ms. (B) Amount of curve shift when optical, electrical, and opto-electrical excitation was used. Gray bar presents the results when H_2-porphyrin was used, and the red bar presents the results when Cu-porphyrin was used. Inset schematic describes the chemical structure of Cu-porphyrin.

the shift of the threshold voltage showed an increase in the Cu-porphyrin (Figure 6B). This increment arises from the increased trap density in the porphyrin. The total trap density inside the porphyrin layer is increased due to the redox-active center metal atoms, allowing a large amount of charges to be trapped in the metalated porphyrin under the same excitation conditions.

CONCLUSIONS

In summary, a hybrid nanowire FET was proposed and analyzed in terms of its porphyrin–silicon interactions. The excitation methods could be thoroughly analyzed because of the hybrid gate structure, in which both the electrical and optical gates coexist in the nanowire body. The analysis of the optical, electrical, and opto-electrical excitations was performed based on the charging and discharging behaviors of the porphyrins. The optical excitation clearly caused the electron transfer through the PCT process; however, the amount of the curve shift was limited because of the lack of electrons in the p-type Si-NW. The electrical excitation can cause a larger curve shift by forming inversion electrons, but this shift was also limited because of the electrical breakdown by the high gate voltage. The largest curve shift that did not disrupt the structure was obtained using opto-electrical excitation. The porphyrins sufficiently absorbed electrons *via* the PCT process even under a weak light intensity and low gate voltage by forming inversion electrons at the Si-NW surface. The electron discharging process was also analyzed by the different excitation methods. The rapid discharging, exponential detrapping, and metastable state were observed in these analyses. The opto-electrical excitation showed combined discharging characteristics of the optical and electrical excitations. The experimental results and analyses of the proposed hybrid FET provide a clear understanding of the mechanisms associated with this new hybrid FET and provide new insight that can be used for further development of generic hybrid devices.

METHODS

The hybrid FET was fabricated using a top-down approach. A nanowire (NW) channel was made on a bulk silicon wafer using the Bosch process, which has advantages in terms of the fabrication cost and practicality compared with the silicon-on-insulator (SOI) wafer process.[16] The p-type nanowire channel was doped with boron at a concentration of 10^{17} /cm^3. A gate (G), source (S), and drain (D) were doped with arsenic at a concentration of 10^{20}/cm^3. The gate was made from polycrystalline Si (poly-Si), while the gate insulator was composed of thermally grown silicon dioxide (electrical gate oxide). A chemical mechanical polishing (CMP) process was employed to separate the electrical gates. This process removed the poly-Si completely and a hard mask nitride partially on the top surface of the NW. Here the hard mask nitride was used as an etching stopper during the CMP. Thereafter, the remaining hard mask nitride was etched out using hot phosphoric acid so that the top surface of the Si-NW was exposed. Finally, porphyrins were immobilized onto the exposed Si-NW by drop-evaporation for 24 h at room temperature. See the Supporting Information for further details regarding the fabrication processes.

Measurement of the device characteristics is conducted with a semiconductor parameter analyzer (4156C, Agilent Technology Inc.). The devices are kept in a dark environment in a measurement box during the experiments. An optical white light source (Megalight100, Schott) is placed inside the measurement box and turned on when optical excitation is applied. After contact of the probe tips onto the device pad (source, drain, and gate pads), a waiting time of 10 min passes for stabilization. To apply opto-electrical excitation, the light is initially turned on and the gate pulse using the parameter analyzer is applied at the midpoint of the illumination time. The *I*–*V* curve is then immediately plotted after the illumination. A delay of approximately 1 s is unavoidable owing to the manipulation of the buttons in the parameter analyzer.

Conflict of Interest: The authors declare no competing financial interest.

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Supporting Information Available: Detailed fabrication procedure, UV–visible and Raman spectra of porphyrin, energy-dispersive spectrometer (EDS) analyses, control experiment results, and body width dependency. This material is available free of charge *via* the Internet at http://pubs.acs.org.

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