Multicolor Spectral-Specific Silicon Nanodetectors based on Molecularly Embedded Nanowires

Sharon Lefler,† Roi Vizel,† Ella Yeor,† Eran Granot,† Omri Heifler,† Moria Kwiat,† Vadim Krivitsky,† Miguel Weil,‡ Yuval E. Yaish*,∥ and Fernando Patolsky†,*‡

†School of Chemistry, Faculty of Exact Sciences, ‡Department of Materials Science and Engineering, Faculty of Engineering, and ∥The Department of Cell Research and Immunology, Faculty of Life Sciences, Tel Aviv University, Tel Aviv, 69978, Israel
‡Andrew and Erna Viterbi Faculty of Electrical Engineering, Technion, Haifa 3200003, Israel

Supporting Information

ABSTRACT: Silicon-based photodetectors cannot distinguish between different wavelengths. Therefore, these detectors relay on color-specific filters to achieve color separation. Color filters add complexity to color sensitive device fabrication, and hinder miniaturization of such devices. Here, we report an ultrasmall (as small as ~20 nm by 300 nm), red-green-blue-violet (RGBV) filter-free spectrally gated field effect transistor (SGFET) detectors. These photodetectors are based on organic-silicon nanowire hybrid FET devices, capable of detecting specific visible wavelength spectrum with full width at half-maxima (fwhm) under 100 nm. Each SGFET is controlled by a distinctive RGBV spectral range, according to its specific organic fluorophore functionalization. The spectral-specific RGBV detection is accomplished via covalent attachment of different fluorophores. The fluorophore molecules inject electrons into the nanowire structure as a result of light absorption at the appropriate RGBV spectral range. These photoinduced electrons modify the occupancies of the oxide’s surface states, shifting the device threshold voltage, thus changing its conductivity, and functioning as a negative stress bias in a p-type SiNW FETs. A positive biasing can be achieved via UV light-induced ionization, which leads to detrapping and translocation of electrons at the oxide layer. Furthermore, a novel theoretical model on the mechanism of action of these devices was developed. Also, we show that suspended SGFETs can function as nonvolatile memory elements, which unlike fast-relaxing on-surface SGFETs, can store discrete “on” (RGBV illumination) and “off” (UV illumination) states for several days at ambient conditions. We also demonstrate a unique single-nanowire multicolor photodetector, enabling in principle a broad spectral detection over a single silicon nanowire element. These highly compact, spectral-controlled nanodevices have the potential to serve in various future novel optoelectric applications.

KEYWORDS: Silicon, nanowires, optoelectronic, fluorophore, wavelength specific

Photodetectors are essential for imaging systems, environmental surveillance, communications, and biological sensing. Spectral discrimination by photodetectors is currently realized using broadband inorganic semiconductor photodiodes in combination with dichroic mirrors or a set of optical filters. Narrowband photodetection is usually realized by one of three approaches: (1) by combining broadband photodetectors with bandpass filters and nanocrystals in the photodiode active layer;1–3 (2) by splitting the light into its component colors;4 and (3) by intentionally enhancing absorption in a particular wavelength range via the plasmonic effect.5 These approaches are yet to demonstrate an appropriate set of performance metrics for any meaningful application, that is, spectrally tunable narrowband responses with full width at half-maxima (fwhm) of <100 nm. Potential candidates for spectral specific detection, such as quantum dots, were widely explored as color detectors, mainly in combination with graphene,6,7 however, these devices do not cover the entire visible spectrum. Only recently, the first color-specific nonsilicon-based detectors were reported, utilizing complex thin, as well as thick, film photodiodes based on perovskite crystals.8,9 Nevertheless, these perovskite-based photodetectors are at the millimeter scale and cannot be integrated into present silicon-based integrated circuits. Over the past decade, nanowire-based electronic devices have been demonstrated as a powerful sensing platform, displaying significant advantages such as rapid, direct,10 and ultrasensitive multiplexed detection of a wide-range of biological11,12 and chemical species.13–15 These devices overcome intrinsic limitations of planar field effect transistor (FET) devices, due to their one-dimensional (1D) nanoscale morphology.16,17 The exceptionally high surface-to-volume ratio of these nanostructures make their electrical properties extremely sensitive to charges adsorbed on their
surfaces, which alter the nanowire’s surface potential. This local change in the electrical field alters the carrier density within the nanowire structure, through electrostatic gating, thus leading to changes in the device measured current. Furthermore, nanowires can be synthesized with a fine control over their diameter, length, chemical composition, doping/electrical properties, and shape, and with the ability to create large-scale arrays of discrete nanowire elements. Recently, dye-sensitized (porphyrin) thin films were shown to induce electric current changes in silicon nanowires (SiNWs) by light-induced field effect gating. However, these devices are not spectrally selective, nor can they optically control the electrical “on”/“off” state of the device.

Following spectral-specific absorbance of fluorophores, an electron in the highest occupied molecular orbital (HOMO) is excited to the lowest unoccupied molecular orbital (LUMO), where it normally relaxes back to the ground state by emitting fluorescent light. Occasionally, once in 500–1000 excitations, the molecule visits the triplet state. At this triplet state, the fluorophore can transfer an electron to its surroundings. This phenomena is well observed in biological assays, as the formation of reactive oxygen species resulting in phototoxicity. Following electron transfer, the fluorophore enters a dark state, until regaining back an electron. This effect is known as redox blinking, and lasts for a few milliseconds in physiological solutions. In the present work, we study the
Figure 2. Spectral-specific response of SGFET devices. (a) Visible spectral response of unmodified as well as different R/G/B/V-modified SGFET devices, sequentially scanned from the red 700 nm to the 400 nm violet section (2.5 ± 1 mW cm⁻²) using monochromatic excitation with 10–12 nm steps (at 4 s intervals) at a total scan time of ∼300 s. The spectral current response $I_{sd}^{obs}/I_{sd}^{dark}$ of the different, top to bottom, violet (Alexa-430), blue (Alexa-488), green (Alexa-555), red (Alexa-610), and unmodified devices, is presented as a function of the wavelength. The spectral specificity is represented by an increase in the $I_{sd}^{obs}/I_{sd}^{dark}$ and the responsive wavelengths are indicated as dashed boxes as follows: violet (486–412 nm); blue (532–486 nm); green (554–590 nm); red (603–648 nm). All indicated wavelengths represent the center of the monochromatic excitation beam and have a ±6 nm shift due to the 10–12 nm bandwidth. (b) Absorbance spectrum of glass modified with the different fluorophores (Alexa 430, 488, 555, 610) as measured by a spectrophotometer (arbitrary units, dashed lines), and calculated spectral response of the different RGBV (solid lines). Note the close overlap between the absorbance spectrum and the SGFET responses. (c) Typical time-response of $I_{sd}$ (shown for blue-SGFET) by blue excitation (120 mW cm⁻², pulse duration is indicated in blue). Calculated rise time = 6.1 s, decay time = 35.6 s, $t_1 = 2.3$ s, $t_2 = 15.9$ s. Inset: sequential blue excitation cycles. (d) $I_{sd}$–$V_{sd}$ curves of p-type red-SGFET device without illumination and following 5 and 10 s illumination cycles of red excitation. Note the negative gate-like effect of the spectral-specific excitation. (e) Extracted $V_{th}$ (calculated from Supplementary Figure S5b) of red-SGFET device as a function of the accumulated red excitation time (2 mW cm⁻²). Inset: $V_{th}$ shift difference ($\Delta V_{th}$) as a function of natural logarithm of the accumulated excitation time ($t_e$). Black circles are experimental data and red line is a linear fit.

effect of photoinduced fluorophore-mediated electron transfer in conjugation with SiNWs in a dry environment to achieve spectral-specific nanodetectors. Nowadays, there is a growing need for nanoscale spectral-sensitive detectors for integration in miniaturized sensing devices in optoelectronic applications. Here, we present a novel paradigm of red-green-blue-violet (RGBV) spectrally sensitive, as well as UV/vis spectrally controlled, fluorophore-induced electrical gating of individual SiNW-based FET devices. These SGFETs detect different colors by exploiting the spectral-specific absorbance, and electron transfer tendency, of various fluorophores upon photoexcitation. By virtue of their relative simplicity and nanometric dimensions, these SGFETs could be used to form highly dense color sensors up to an effective pixel size of 20 nm × 300 nm, as well as forming 3D stacked sensors <500 nm in height.

Results and Discussion. Device Fabrication and Architecture. P-type boron-doped SiNWs (as well as n-type phosphorus-doped nanowires) were grown by the vapor–liquid–solid chemical vapor deposition method, followed by dry transferring to their destined locations on a silicon wafer, and the formation of electrical contacts by lithography and metal deposition steps. SiNWs of 80 and 20 nm diameter were used as the sensing elements in our spectrally gated FET devices (see Supporting Information). These SiNWs were used as part of a sensing multi-FETs array chip (Figure 1a), which consists of about 200 planar SiNWs FET devices, ~6 μm long (source-to-drain distance) (Figure 1b,c). Chips were fabricated on a 600 nm thermal-oxide silicon n-type wafer. The opto-sensing chip consist of four sensing areas, where each area can be chemically modified to detect a different wavelength, as indicated in Figure 1a,h for the different RGBV color detection (see Supplementary Figure S1a). In addition, FET devices of smaller dimensions, 300 nm source-to-drain distance, were fabricated by the transfer of SiNWs of 20 nm diameter onto a silicon wafer, followed by e-beam lithography to define the source and drain electrodes (Figure 1e,f). SiNWs were chemically functionalized using 3-aminopropyltrimethoxysilane (APDMES), followed by the cross-linking of amine-reactive Alexa-fluorophore NHS ester derivatives. The following commercially available Alexa fluorophores were used: Alexa 430, Alexa 488, Alexa 555, and Alexa 610 (see Supplementary Figure S1b). Chemical modification was performed postfabrication, effectively covering the whole SiNWs, as well as the wafer, surfaces. Whenever SiNWs were to be selectively modified (without modification of the whole wafer surface),
the silicon wafer was precoated with a thin ∼5 nm alumina layer, which serves as sacrificial layer, then removed after the APDMES functionalization step and just before the NWs modification with the different fluorophore molecules (Supplementary Figure S2a).

Spectral-Specific Response of Fluorophore-Embedded SiNWs-based Devices. SiNWs SGFET devices were functionalized using APDMES followed by the cross-linking of different amine-reactive Alexa fluoro-NHS esters (illustrated in Figure 1a,h), and as can be observed by their corresponding fluorescence emission (Figure 1d and Supplementary Figure S1a). A representative current-to-voltage ($I−V$) curve characteristic of a fluorophore-modified SiNW SGFET device is presented in Figure 1g. For comparison, a typical $I−V$ curve of a bare-SiNW FET device is presented in Supplementary Figure S1c. We first tested the spectral response of our Alexa 488-modified SiNWs devices (blue-SGFET), using a laser scanning microscope (Figure 1i). This set of experiments was performed using suspended SiNWs FET devices, in order to exclude any effect that may originate due to the concomitant chemical modification of the device’s surrounding surfaces. Suspended SiNW FET devices were fabricated using pre-etching of the silicon oxide layer between the inner-electrodes, for the formation of trench structures using reactive ion etching (RIE), leading to suspended SiNW elements attached to a metal electrodes array (see scanning electron microscope (SEM) image as inset in Figure 1i). The microscope was accommodated with 633 nm (10 mW), 546 nm (10 mW), and 488 nm (7.5 mW) lasers. All lasers were set to 1% of their power output, and only the area of the nanowire was excited for 10 s using all three lasers sequentially (Figure 1i, area A). As can be seen in Figure 1j (blue line), the blue detector nanodevice only responds to the blue 488 nm laser, as manifested by a $I_{sd}$ current elevation, up to 150% above its electrical baseline ($I_{sd}^{\text{light}}/I_{sd}^{\text{dark}}$). To be certain that the SiNW’s part of the device is responsible for the spectral-specific effect, we tested different parts of the SGFET device, including the SiNW section (Figure 1i, area A), the area of the electrodes (Figure 1i, area B), as well as the area where the SiNW is connected to the metal electrodes (Figure 1i, area C). The results clearly show that only the SiNW area is accountable for the observed spectral-specific effect, and also an illumination with a higher 20% laser power on the electrodes areas did not result in any SGFET $I_{sd}$ current elevation. This demonstrated the spectral-specific photoinduced $I_{sd}$ current elevation, similarly to the application of negative gate bias in p-type SiNW FETs. To extend the spectral detection of our devices, we tested different fluorophores with absorbance spanning the entire visible spectrum: Alexa-430 (violet-nanodetector), Alexa-488 (blue-nanodetector), Alexa-555 (green-nanodetector), and Alexa-610 (red-nanodetector)-functionalized planar devices (using nanowires of 80 nm in diameter for the fabrication of 6 μm channel-length SiNWs devices), as well as nonfunctionalized SiNWs devices as reference nonspecific nanodetectors. This time, we tested the spectral–electrical response of the different fluorophore-modified SiNWs detectors using a xenon monochromatic light source (Polychromator IV, Till Photonics) sequential scan of the visible spectrum from the red (700 nm) to the violet (400 nm) (Figure 2a). The total scan time was ∼300 s, at intervals of 10–12 nm spectral bandwidth (at 4 s intervals). The Alexa-modified p-type SiNW devices exhibit a clear elevation in their $I_{sd}$ current following spectral-specific response, and show no change in nonresponsive spectral regions. Examining the response curve of the Alexa-430 nanodetector (violet-nanodetector) demonstrates that light excitation ranging from 700 nm down to about 486 nm does not induce any change in the $I_{sd}$ current of the violet SGFET detector; however, starting at ∼486 nm down to about 412 nm there is a noticeable 2.2-fold elevation of the $I_{sd}$ current relative to the dark state level of the device ($I_{sd}/I_{sd}^{\text{dark}}$). We refer to this part as the spectral-specific effect, corresponding well to the expected absorbance spectrum of the fluorophore anchored to the SiNW devices (Figure 2b). Out-of-range illumination does not cause any elevation in the measured conductivity, and the $I_{sd}$ current gradually reverts back to its baseline level, as can be clearly seen in all our fluorophore-modified SiNW RGBV detectors (Figure 2b, solid lines). Calculation of the fwhm, and the relative responsiveness of each color nanodetector (Figure 2b) reveals fwhm values of under 100 nm for all RGBV detectors: 38, 45, 49, and 94 nm for the red, blue, green, and violet nanodetectors, respectively. This extracted $I_{sd}$ response entirely overlaps with the expected fluorophores absorbance spectra, as measured by a spectrophotometer (Cary 5000 Spectrophotometer, Figure 2b dashed lines). Altogether, our different RGBV SGFET nanodetectors show spectral-specific detection of the appropriate color, which is spectrally tuned by the absorbance spectra of their surface-anchored fluorophores. The measured rise and decay times using our blue-SGFET devices, following blue excitation of ∼120 mW × cm−2 (Figure 2c), are 6.1 s (with time constant $t_1 = 2.3$ s) and 35.6 s (with time constant $t_2 = 15.9$ s), respectively. We further tested the spectral specificity of an ultrashort e-beam lithography fabricated SGFET device (20 nm diameter SiNW with an inter-electrode gap of 300 nm), to be potentially incorporated in submicron color detection applications (Figure 1e,f). The results obtained from the ultrashort Alexa 488-modified blue-SGFET nanodevice (Supplementary Figure S3a–c) demonstrate high spectral specificity (Supplementary Figure S3a), as well as superior response and recovery times (Supplementary Figure S3b). The devices also display a typical trans-conductance response (Supplementary Figure S3c). Furthermore, SGFET devices display high optical as well as chemical stabilities, with a minimal response reduction of <5% after 140,000, 2 s switching cycles at 40 mW × cm−2 at ambient conditions (Supplementary Figure S4a). The performance and chemical stabilities of our devices were further enhanced by the formation of a conformal ALD-based alumina thin shell, 8–15 nm thickness (Supplementary Figure S4b), and shown to result in fluorophore-embedded nanowire hybrid structures of remarkably high chemical stabilities under the harshest tested conditions, while maintaining the spectroelectrical activity of the devices and the spectroscopic attributes of the embedded fluorophore molecules (see Supplementary Figure S4c–f). We believe that encapsulation of the fluorophore molecules by the alumina layer protects the fluorophores from degradation and undesired photobleaching under long illumination periods. Encapsulation of fluorophores was shown in the literature to serve as protecting means against fluorophores degradation.34,35

Spectral-Specific Response Attained via the Field Effect. To understand the effect underlining the observed increase in the electric current following spectral-specific fluorophore-mediated response, we tested the electric properties of SGFET devices following spectral-specific excitation. To this end, a red-SGFET (Alexa-610) nanodetector was excited with a 1.5 mW × cm−2 red light source for different time durations, and the effect on the resulting $I−V$ curves was measured (Figure 2d). The
shape of the $I_d/V$ curves following spectral-specific excitation exhibit similar response to that of applying an increasing negative $V_g$ biasing to a nonmodified p-type device (Supplementary Figure S1c), which results in an increased conductivity. The $V_{th}$ is defined as the minimum gate-to-source voltage differential needed to create a conducting path between the source and the drain terminals. We extracted the threshold voltage ($V_{th}$) change using sequential transconductance experiments, following red light excitation (2 mW cm$^{-2}$) of a red-SGFET (Figure 2e, calculated from Supplementary Figure S5b) to evaluate the spectral-specific field-effect gating. This gating effect is manifested by a shift in the $V_{th}$ as a function of excitation duration, ranging from $-0.1$ V without illumination to 7.5 V following 60 s of red light excitation, implying an accumulation of negative charges at the Si/SiO$_2$ interface trap states, serving as a local negative gate bias. Electrical characterization of the spectral response of our blue-SGFET nanodevices was repeated under different illumination intensities in the 40–220 mJ cm$^{-2}$ range, using a blue xenon light source, resulting in a correlative response, similar to the change seen in the $V_{th}$ shift (Figure S5a). We also tested the spectral-specific response of our nanodevices at various $V_g$ biasing potentials, ranging from $-9.5$ to 9.5 V (Supplementary Figure S5c) and found a direct correlation which clearly demonstrates a field-effect-based mechanism. At high negative bias, when the device is close to saturation, the effect of the spectral-specific effect is relatively small ($\Delta I_{ds} = 15$ nA), whereas at high positive bias, close to the device depletion region, the spectral-specific effect is substantially larger ($\Delta I_{ds} = 50$ nA). These experiments give good indication that the spectral specificity of the SGFET devices is attained via the field-effect mechanism.

To further investigate the source of the observed spectral-specific fluorophore-induced field effect, we performed a series of surface potential mapping experiments using Kelvin-probe microscopy (KPFM), in order to measure the surface potential change following spectral-specific excitation of our devices. Samples were prepared by transferring Alexa-modified SiNWs on top of native oxide silicon wafer substrates, resulting in a dense arrangement of SiNWs. Alexa 555-modified SiNWs were used, and a 25 μm$^2$ area was scanned, while the topography of the same area was simultaneously examined (Figure 3a). The atomic force microscopy (AFM) topography of the scanned area revealed several SiNWs, some lying flat in direct contact with the wafer surface (Figure 3a, indicated by a yellow arrow), while others stacked one on top of other forming partially suspended SiNWs (Figure 3a, indicated by a blue arrow). In parallel, KPFM surface potential was acquired, showing a uniform surface potential for the fluorophore-modified SiNWs (Figure 3b). Following 40 s of illumination at 546 nm, 200 mW cm$^{-2}$ (Figure 3c), it can be seen that the SiNWs have gained a significant rise in their surface potential, $\sim$60 mV (see Supplementary Figure S6a–d), while certain sections of the SiNWs, as well as the surrounding wafer areas, exhibit only a modest uniform potential rise. The sections of the SiNWs displaying increased surface potential upon wavelength-specific illumination are clearly identified in the topography scan as wires slightly elevated from the wafer surface. Note that each scan takes about 25 min, consequently all fast dissipating charges will not be detected during the slow KPFM measurements. One hour postillumination, an additional KPFM scan measurement shows that the accumulated charge was only detected at the most suspended SiNW sections, while it is completely lost from other nonsuspended parts of the nanowires (Figure 3d, indicated by a blue and yellow arrow, respectively).

Following spectral-specific excitation of fluorophore-modified p-type SiNWs devices, we observe an elevation in the conductivity of the nanowires, indicating a negative shift in electric potential at the Si/SiO$_2$ interface. KPFM measurements show that following spectral-specific excitation, there is an $\sim$60 mV shift in the electrical potential as detected at the outer surface of the fluorophore molecular layer decorating the SiNWs (Supplementary Figure S6d). This apparent contradiction can be resolved by the idea that photoinduced negative charges transfer from the fluorophore molecules at the outer molecular-layer of the chemically modified device toward the SiNW structure, resulting in a net positive electrical potential change as detected by the KPFM measurements. Our results clearly show the nature of the accumulated charge, which originates at the fluorophores decorating the SiNWs surface, and translocate to the SiNW structure following spectral-specific excitation.

Figure 3. Accumulation and dissipation of charges following spectral-specific excitation of SGFET devices. (a) AFM scanning results of 5 μm × 4 μm area of Alexa-555 modified p-type ~80 nm diameter SiNWs, which were dry-transferred onto a native oxide p-type silicon wafer. SiNWs bright suspended sections are marked by blue arrow, flat SiNWs sections are marked by yellow arrow. (b–d) KPFM scan displaying similar electrical potential distribution along all of the SiNWs, as well as the wafer surface. (c) Following 40 s of green light excitation (200 mW cm$^{-2}$), displaying bright positively charged areas in suspended sections (blue arrow). (d) One hour postexcitation, only suspended areas (blue arrow) of the SiNWs still retained their positive charges triggered by the light excitation. (e) SEM image of a suspended SGFET device. (f) Long-term measurements of the $I_{ds}/I_{ds}^\text{pre}$ of a suspended SGFET device under ambient conditions, following blue excitation for 20 s (2 mW cm$^{-2}$). The device retains $\sim$2 fold of the original value (before excitation) for about 14 h. Inset: First 2 min postexcitation. Note that planar SGFETs revert back to their original value about 1 min postexcitation (for comparison see Figure 2c).
**Suspended SGFET as Nonvolatile Memory Elements.** Our KPFM measurements suggest that fluorophore-modified suspended nanowire sections are capable to accumulate and hold negative electric charges following their spectral-specific excitation. To this end, we tested suspended nanowire SGFET devices (suspended devices in Figure 3e and Supplementary Figure S7a versus planar devices in Figures 1c and S1c). Electrical measurements of the current decay following spectral-specific excitation show a prolonged memory effect, which fully supports our KPFM observations (Figure 3f). While the $I_{sd}$ current of planar SGFETs reverts back to its original baseline value about 1 min postexcitation (Figure 2c), in suspended SGFET devices the light-induced elevated $I_{sd}$ current persists, and is found to be preserved at $\sim2$–$3$ folds above the baseline value for days postexcitation at ambient conditions (Figure 3f, Figure 4a for blue-SGFET, and Supplementary Figure S8 for green-SGFET). We attribute this prolonged memory gating effect to the isolation of the suspended SGFET nanowire devices from the wafer surface, leading to the extended retention of light-induced negative charges at the nanowire. In planar SGFETs, the close contact of the nanowire oxide with the wafer oxide promotes recombination events leading to charge equilibrium and redistribution of electrons along the wire, and reintroduction of electrons back into the fluorophore layer. These memory elements are nonvolatile in nature, depending only on the field-effect gating, which persists also in the absence of any applied $V_{gd}$ potential (Figures 4a and S8).

**Spectral Control of the “On” and “Off” States of the Suspended-Nanowire SGFET Nonvolatile Memory Devices.** Several optoelectronic applications require efficient control of the “on” (write) and “off” (erase) states of a device. In most cases, this is done using broad-band illumination to reach the “on” state and no illumination for the “off” state. However, this approach requires continuous illumination through the “on” state, which is highly power demanding, generating heat and noise sources. In other applications, the “on” state is generated via pulse illumination, whereas the “off” state is achieved via high-voltage gate bias. In this case, the device is only semioptically controlled. As described above, following spectral-specific excitation, negative charges translocate from the fluorophores to the SiNW and cause elevation of $I_{sd}$ for p-type SGFETs. The accumulation of charges, in particular in...
suspended devices, results in a prolonged "on" state that can persist for days at ambient conditions. We have found that UV illumination at <390 nm (≈3.1 eV), causes the reciprocal effect, causing a rapid reduction/elevation in \( I_{d} \) for p-type/n-type SiNW FETs, respectively (Figure S9a). Combining UV illumination with the SGFET spectral-specific excitation properties allows the control of the "on" (write)"/"off (erase)" states of our nonvolatile memory elements, without the use of an electrical gate bias. As presented for the suspended blue-SGFET devices (Figure 4b,c, and for green-SGFET Supplementary Figure S10a,b), following short UV illumination (365 nm, \( \sim 2 \) s), there is a rapid decrease in the \( I_{d} \) current of the device reaching the "off" state, while following spectral-specific blue excitation (488 nm, \( \sim 5 \) s) there is an elevation of the \( I_{d} \) current to the "on" state. The "on"/"off" switching is maintained for multiple sequential UV/blue excitation cycles, at intervals of 10 and 1 min (Figure 4b,c respectively) and for suspended green-SGFET (Supplementary Figures S10a,b, respectively). Similar experiments of spectral-specific/UV excitation cycles were done also using n-type blue-SGFET devices (Supplementary Figure S9b), displaying opposite behavior to the p-type SGFETs, where the blue excitation causes reduction of the electrical \( I_{d} \) current, while UV illumination results in an elevation of the \( I_{d} \) current. To the best of our knowledge, this represents the only device capable of performing color-coded spectral-specific writing/erasing switching.

**Suggested Model of Operation.** The measured SiNW SGFETs in this study belong to the subgroup of FETs named junctionless FETs (JLFETs), in which the doping level of the silicon channel and the source/drain regions are similar and high.\(^{38-40}\) In these devices a reasonable amount of current flows when they are turned on, and charge is fully depleted in the "off" state. Often, due to interface traps within the surrounding oxide, or in close vicinity of the Si/SiO\(_2\) interface, many SiNW FETs exhibit hysteretic behavior in their transfer characteristic (TC).\(^{41}\) The physical origin for this hysteresis relies on charging and discharging of interface states within the oxide which interact with mobile charges at the silicon wire depending on the position of the SiNW's Fermi energy, \( E_F \) and trap states. The charge of these interface traps can be negative, positive, or neutral, depending on the relative position of \( E_F \) and the intrinsic midgap energy level, \( E_F \) at the Si/SiO\(_2\) interface. Traps below \( E_F \) act as donors, that is, below \( E_F \) are neutral and above \( E_F \) are positively charged. Traps above \( E_F \) act as acceptors, that is, below \( E_F \) are negatively charged, and above \( E_F \) are neutral. Because interface traps can be located even 1 or 2 nm inside the oxide, they exhibit long capture time constants due to tunneling processes from the Si/SiO\(_2\) interface into the oxide (see Supplementary Figure S11a).\(^{42}\)

The relative position of \( E_F \) with respect \( E_F \) is determined by the external gate voltage, work functions difference between the channel and the gate, fixed charges at the oxide or its vicinity, \( Q_{\text{fixed}} \) and interface trap charges, \( Q_{\text{it}} \), according to the following relation\(^{43}\)

\[
V_g = V_g - V_{sc} + \varphi_i - \frac{Q_{sc}}{C_{ox}} - \frac{Q_{\text{fixed}}}{C_{ox}} - \frac{Q_{\text{it}}}{C_{ox}}
\]

where \( W_g/W_{sc} \) represents the gate/channel work function, \( \varphi_i \) is the electrostatic potential at the Si/SiO\(_2\) interface (\( r = R \)) measured with respect to \( E_F (r = 0) \), \( Q_{sc} \) is the space charge density, and \( C_{ox} \) is the gate-silicon mutual capacitance. All the

Q's and capacitance are defined per unit length. \( Q_{sc} \) can be expressed as \( Q_{sc} = 2\pi \frac{R_{\text{eff}}}{(E_F - E_{g} - \varphi_i)} = 2\pi \frac{R}{(E_F - \varphi_i)} \), where \( R \) is the wire radius, \( D_{\text{sc}} \) is the interface state density with units of \( \text{cm}^{-2} \cdot \text{eV}^{-1} \), \( e = \text{el} \) is the electron charge, \( E_F \) is the intrinsic midgap energy level at the wire origin (\( r = 0 \)), and \( \varphi_i = E_F - E_{g} = k_B T \ln(N_i/n_i) \) where \( N_i \) is the acceptors doping concentration of the studied SiNWs.

According to Shockley–Read–Hall\(^{44}\) model for nonradiative recombination of holes and electrons the density of negatively charged trap with energy \( \varphi_i \) located at a distance \( x \) from the interface into the oxide behaves as \( \exp(-x/\tau_i) \) with time constant, \( \tau_i \) which depends on both the surface density of electrons and holes in the silicon wire (\( n_i, p_i \)), as well as on the occupancy of the trapped state.\(^{45}\) At specific set of external parameters, the time constant of a trap for capturing an electron can be approximated by \( \tau_i^{-1} = n_i v_a \sigma_{ox} \), where \( v_a \) is the average thermal speed of free electron, and \( \sigma_{ox} = \sigma_{ox} e^{-2\delta x^2} \) is the reduced capture cross section for an electron located a distance \( x \) from the interface. \( \kappa_{ox} \) is the evanescent wavenumber of an electron within the oxide barrier which can be approximated to \( 1/(2\kappa_{ox}) \approx 1−2 \) Å under normal operation conditions.

Under applied stress biases from the back gate, the interface traps can be populated or depopulated according to their distance from the silicon interface, and densities of free charges at the Si/SiO\(_2\) interface which depend on \( \varphi_F \) that is, \( n_i = n_i \exp(\beta(e(\varphi_F - \varphi_i))) \), \( p_i = n_i \exp(\beta(e(\varphi_F - \varphi_i))) \) where \( n_i \) is the carrier density for intrinsic wire, and \( \beta = 1/k_B T \). These traps charges, \( \Delta Q_{\text{it}} \), will modify the threshold voltage of the device according to the following relation: \( \Delta V_{gs} \approx \Delta Q_{\text{it}}/C_{ox} \). Because of the long time constants of trapping and detrapping of interface states the system is far from thermodynamic equilibrium and a difference in the traps occupancies is common even for the same gate voltage when sweeping the voltage in the upward or downward direction. This phenomenon can be observed as hysteretic behavior in the TC of the device and will depend on the sweeping range and rate.\(^{41}\) Moreover, for p-type transistors, clockwise hysteresis reveals charge transfer between traps at the oxide, where counterclockwise hysteresis indicates charge trapping or detrapping between the silicon and the oxide.

As mentioned above, the time constant for trapping also depends on \( n_i \) which is typically modified by the gate voltage. However, it is well-known that under low charge injection, either by optical generation or other means, it is possible to significantly increase the minority charge carriers (in our case, electrons) above their thermal equilibrium value, that is, \( \delta n_i > \delta n_i = n_i^0 \), \( n_i^0 = \frac{\tau_i}{\tau_i} \) and \( \frac{\tau_i}{\tau_i} = \frac{n_i}{n_i} \) where \( n_i \) is the electron density at the wire interface under equilibrium conditions. In such circumstances, the charge trapping rate by the interface states will increase and a negative shift in the trap charge density will occur, that is, \( \delta Q_{\text{it}} < 0 \) (Figure 4d, illustration 3). As a consequence, the threshold voltage will increase (\( \Delta V_{gs} > 0 \)) causing current increase/decrease for p-type/n-type transistor. At steady state conditions, the excess electrons will recombine and the total charge density of the minority carrier will become \( n_i = \delta n_i = G_{\text{op}} \tau_{rec} \) where we assume high boron-doped wire (in our wires \( N_i = 10^{18} \text{cm}^{-3} \) hence \( \tau_i = 10^2 \text{cm}^{-3} \))\(^{44}\) \( G_{\text{op}} \) is the generation rate of excess electrons (\( G_{\text{op}} = \text{cm}^{-3} \text{ s}^{-1} \)) and \( \tau_{rec} \) is the indirect recombination time. Alternative explanation of \( \delta Q_{\text{it}} \) is the following: Under generation of extra electrons, above their equilibrium value two quasi Fermi energies form, one for the electrons and one for holes (\( E_{\text{fn}} \) and \( E_{\text{fh}} \)). Since we are interested in low charge injection, the holes concentration is...
barely changed, hence, $E_{\text{FB}}$ remains the same. However, $n_s$ is dramatically changed, therefore, $E_{\text{FN}}$ increases and more donor states at the interface captured an electron, reverting their charge state from positive to neutral, that is, making $Q_s$ more negative. If $E_{\text{FN}}$ increases above the midgap energy level $E_g$ at the Si/SiO$_2$ interface, acceptor states will capture an electron, shifting their charge state from neutral to negative, and again causing $\delta Q_s < 0$.

The duration of the charge injection ($G_{\text{op}}$) will not change $n_s$ since $\tau_{\text{rec}}$ is extremely short (sub ns$^{-1}$) and it reaches its steady state value very quickly. However, since $G_{\text{op}}$ is equivalent to gate bias stress, as the stress is longer more remote traps will be populated increasing $\delta Q_s$ and $V_{\text{th}}$ shift. Estimation for $\delta V_{\text{th}}$ can be done as follows: On the basis of our previous work$^{19}$ we assume an APDMES coverage of $\sim$1.2 $\times$ 10$^{14}$ molecules/cm$^2$, and a surface coverage of $\sigma_0 = 1 \times 10^{15}$ molecules/cm$^2$ of the Alexa fluorophores. Under strong light illumination, all the Alexa molecules are activated but only a small fraction ($\sim$0.1–0.2%) do not recombine spontaneously to their ground state. Few of these excited electrons can overcome the oxide barrier and be injected to the silicon with small probability, $T$, calculated by Wentzel, Kramers, Brillouin (WKB) approximation. Since the charge conservation must prevail, the following relation exists

$$2\pi(R + d_{\text{ox}})\sigma_t \times 10^{-3} \frac{1}{\tau_{\text{fi}}} T = \pi R^2 C_{\text{op}}$$

where $d_{\text{ox}}$ is the wire oxide shell ($3$–$4 \text{ nm}$), and $\tau_{\text{fi}}$ is the recombinative time of the Alexa fluorophores (approximately a few nanoseconds).$^{37}$ Putting together for $n_s \approx \delta n_s = G_{\text{op}}\tau_{\text{rec}}$ one finds

$$n_s = \frac{2}{R} \sigma_t \times 10^{-3} \frac{\tau_{\text{rec}}}{\tau_{\text{fi}}} T = 5 \times 10^{14} T \text{ cm}^{-3}$$

where $R = 40$ nm and $\tau_{\text{rec}}/\tau_{\text{fi}} \approx 0.1$ were used ($\tau_{\text{rec}} \approx$ few 100 ps and $\tau_{\text{fi}} \approx$ few ns). The maximum in the transmission probability calculated by WKB for an electron in the Alexa’s excited state and wire’s oxide barrier yields $T \sim 10^{-3}$, and the resulted electron density is $n_s = 5 \times 10^{14}$ cm$^{-3}$ or even $10^{15}$ (if 0.2% Alexa molecules undergo nonradiative recombination) which is much larger than $n_t$ but still much lower than $n_{\text{th}}$, in agreement with low generation rate. If all the traps in the additional window ($\delta(E_{\text{FN}} - E_{\text{FP}})$ will have enough time to be populated, the maximum shift in the threshold voltage will satisfy the following equation

$$\delta V_{\text{th}} \approx \frac{-\delta Q_s}{C_{\text{ox}}} = 2\pi \frac{\text{Re}\, D_q \delta(E_{\text{FN}} - E_{\text{FP}})}{C_{\text{ox}}}$$

The shift of the electron quasi Fermi energy is calculated from the injected carrier density using the following relation $\delta(E_{\text{FN}} - E_{\text{FP}}) = \epsilon_d T \ln(n_s/n_i)$, and can be as large as 0.11 eV for $n_s = 10^{12}$ cm$^{-3}$. For typical values of $D_q = 2 \times 10^2$ cm$^{-2}$ s$^{-1}$ eV$^{-1}$, nanowire radius of 40 nm, and nanowire–gate distance of 600 nm, $\delta V_{\text{th}}$ can be as large as 7.5 V, where we assume the gate–wire capacitance per unit length to be $C_{\text{ox}} = 2\pi \epsilon_0 \epsilon_\text{r} / \ln(2h/R)$,$^{38}$ $\epsilon_0$ ($\epsilon_\text{r}$) is the vacuum (oxide relative) dielectric constant and $h$ is the distance between the wire center and the highly doped silicon substrate. Because of the fact that $h$ is large, the capacitance is small and the threshold shift is much bigger in comparison with devices with smaller ratios of $2h/R$. This large expected shift is in good agreement with our experimental results and is much larger than the estimated threshold shift which is attributed to the electrostatic dipole moment formed between the positive Alexa molecules and their injected electrons.$^{50}$ This shift can be as large as 0.1 V in our setup, and certainly cannot explain the large measured voltage threshold shift found in our experiments.

As was mentioned before, population or depopulation of interface traps within the oxide can span several orders of magnitude in time, depending on the trap energy and distance from the Si/SiO$_2$ interface. As a consequence, the dependence of the voltage shift with respect to the duration of the applied stress (either by gate voltage or carrier injection) usually does not follow a single exponential decay or increase, but a logarithmic behavior$^{31}$ with respect to the stress duration ($t_0$) of the following form $\delta V_{\text{th}} \approx 2\pi \text{Re} \, D_q \delta E \ln(t_0/t_0)/C_{\text{ox}}$ where $\delta E$ is the relevant accessible energy window at the interface, and $t_0$ is the shortest time scale in the measurement.

In the next section, we quantitatively examine the agreement between our experimental results and the suggested theoretical model. First, we examine the TC of bare and modified wires and observe two important facts: (i) the APD MES modifications $V_{\text{th}}$ shifts almost 20 V toward negative gate voltages, implying accumulation of positive charge at the outer surface of the wires. After Alexa modifications the threshold voltage shift back toward less negative or even positive gate voltages (Supplementary Figure S11b). These two trends originate from the positively charged amine groups at the vicinity of the SGFETs after the APD MES modification, and the negatively charged groups of the Alexa fluorophores modification. The net charge that accumulates at the outer surface of the wire per unit length, denoted as $Q_{\text{fix}}$ will be extracted shortly. (ii) The TC for bare and modified SiNWs exhibit hysteretic behavior, depending on the sweeping rate and voltage range (Supplementary Figure S11c). For low sweeping rate (<10 mV/s) the hysteresis is counterclockwise (CCW) and for higher rates it is clock wise (CW). This hysteretic characteristic is a clear indication for interface states at the wire’s oxide, and their density ($D_q$) can be measured from the difference between the threshold voltages of the right and left sweeps using the following relations

$$V_{\text{FB}} \approx W_g - W_c - \frac{Q_{\text{fix}}}{C_{\text{ox}}} - \frac{2\pi \text{Re}^2 \, D_q \phi_B}{C_{\text{ox}}}$$

$$V_{\text{th}}(R, L) = W_c - W_{\text{fg}} + \phi(R, L) - \frac{Q_{\text{fix}}}{C_{\text{ox}}} - \frac{\pi R^2 N_e}{C_{\text{ox}}} - \frac{2\pi \text{Re}^2 \, D_q (\phi_B - \phi(R, L))}{C_{\text{ox}}}$$

where $R$ and $L$ denote values of the right or left curves, and $V_{\text{FB}}$ is the flat band gate voltage. For example, from the TC of the unmodified wire presented in Supplementary Figure S11d we could extract the surface density of fixed charges, $\sigma_{\text{fix}} = Q_{\text{fix}}/2\pi(R + d_{\text{ox}})e \approx 0.42 \times 10^{12}$ cm$^{-2}$, and $D_q = 1.67 \times 10^{12}$ cm$^{-2}$ eV$^{-1}$ which is in good agreement with interface states density measured in native oxide SiNWs.$^{37}$ Using the same $D_q$ in the TC depicted in Supplementary Figure S11b for the same wire before and after chemical modifications, resulted with $\sigma_{\text{fix}}$ (before) $= 0.51 \times 10^{12}$ cm$^{-2}$ and $\sigma_{\text{fix}}$ (after) $= 1.6 \times 10^{12}$ cm$^{-2}$.

As discussed before, upon spectral-specific light excitation the $V_{\text{th}}$ increases for p-type SGFETs. Taking $I_q \sim V_{\text{th}}$ sweep after each light excitation (Supplementary Figure S5b) enables us to consecutively extract the threshold voltage. Such analysis is presented in Figure 2e, where $V_{\text{th}}$ as a function of the

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accumulated time of illumination, which is equivalent for the stress duration in gated devices, is plotted. First, we see that for long stress duration the threshold shift almost saturates, implying traps population approaching thermodynamic equilibrium. Second, the saturation value is close to 8 V, in excellent proximity to the estimated value of 7.5 V. Third, we plot $\delta V_{th} = V_{th}(t) - V_{th}(t = 0)$ of the same data set versus $\ln(t)$ as predicted for a broad time constant dispersion of interface traps within the oxide$^{51}$ (Figure 2c, inset). The data nicely fit to the predicted model, and the slope ($a$) should be compared to $a = 2\pi R D_{ox} \delta E/C_{ox}$. This relation can be reverted for extracting the relevant energy window for traps population $\delta E$. Using $a = 1.6$ and $1.67 \times 10^{12} \leq D_{ox} \leq 2 \times 10^{12}$ cm$^{-2}$ eV$^{-1}$ one finds $0.12 \leq \delta E \leq 0.14$ eV which is in good agreement with our previous estimation of 0.11 eV.

When light is turned “off”, planar SGFET devices are relaxed back to their base state in tens of seconds (Figure 2c). The reason for this rapid relaxation relies on the fact that positively charged Alexa molecules can regain back their missing electron from the nearby oxide of the entire wafer. When these molecules return to their neutral states, no attraction between the negatively charged traps within the wire oxide to the positively Alexa molecules prevails, and the trapped electrons can relax back to the wire body in a similar rate for the trapping process. For suspended SGFET, as previously described (Figure 3f), current relaxation persists for days at ambient conditions. In this case, electrons from the wafer’s surface cannot diffuse toward the positively charged Alexa molecules, due to presence of the air gap, and as a result, only electrons which succeed to tunnel through the entire oxide shell of the wire can neutralize the Alexa molecules. This process lasts a long time because no light illumination is involved and causes slow relaxation of the current in suspended devices. UV illumination (Figure 4d, illustration 4) induces ionization and detrapping of electrons held in traps, which was shown to occur at an initial energy of $\sim 3.1$ eV in others experimental systems$^{52,53}$ as well as in commercial applications similarly to the effect seen in an erasable programmable read-only memory (EPROM) chip. UV-induced ionization consequently reintroduce electrons back into the fluorophore layer, reverting the wire conductivity back to its state before fluorophore excitation.$^{54}$ This repeated interplay between the fluorophores spectral-specific excited electrons and the UV oxide ionization allows control over the “on”/“off” state of our SGFETs.

**Single-Nanowire Multi-Color Detector.** Reducing the dimensions and complexity of color detection devices is of critical importance in future applications. For this purpose, we designed the fabrication of multicolor single-nanowire detectors. These devices consist of a single-nanowire element, electrically coupled in a manner that allows the formation of multiple SGFETs on a single nanowire unit through the use of lithography means. The segment-specific chemical modification with multiple fluorophores allows the formation of multicolor pixels along a single-nanowire, as illustrated in Figure 5a.

In order to demonstrate this concept, we embarked the design and fabrication of a dual-color detector device, consisting of a single nanowire dual-SGFT, chemically modified with Alexa-488 and Alexa-555 fluorophores. Shortly, single-nanowire dual-SGFTs were fabricated, displaying an active channel length of 2 $\mu$m each. The segment-specific fluorophores chemical modification was performed through the use of photolithography steps (please see Supplementary
modification of a red (Alexa 610 - red emission, left section)/violet (Alexa 430 - green emission, right section) on dual-color single-nanowire. (e) Normalized current response of a single-nanowire dual-color red-violet nanodetectors (excitation band-pass filters spectrum are indicated at the side of each excitation 10 s pulse) displaying highly spectral-specific response of each dual-color detector with a minimal crosstalk due to the fluorophore absorbance large separation.

Illustration 1 for details). The successful fabrication of the segment-specific fluorophore-modified devices is shown in Figure 5b. It can be clearly observed that the dual-SGFET single nanowire devices are successfully segment-specifically modified with two fluorophores, Alexa-488 (green fluorescence) and Alexa-555 (red fluorescence), each covering a different spectral range. This successful fabrication sequence allowed the creation of single-nanowire dual-color detector, as shown in Figure 5c (blue-green pixels) and Figure 5e (violet-red pixels). Important to note that Alexa-488 and Alexa-555 fluorophores display a considerably large spectral overlapping, (see Figure 2b for absorbance characteristics), thus, this fact is expected to be directly reflected on the spectroelectrical behavior of the resulting color detector devices.

The dual-color single-nanowire detectors exhibit a clear spectral-specific response upon excitation at wavelengths of blue (460–500 nm), green (520–550 nm), and red (590–620 nm). Examining the temporal response curves (Figure 5c) of the Alexa-555 segment (green pixel) versus the Alexa-488 segment (blue pixel) on the single-nanowire dual detector demonstrates that green light excitation induces a considerable change in the $I_d$ current of the respective green pixel detector, while only displaying a moderate current elevation for the blue pixel. Alternatively, blue excitation causes a fast and considerable change in the $I_d$ current displayed by the blue pixel segment, while leading to smaller current changes shown by the green pixel segment device. The smaller current changes observed in these cases are attributed to the spectral overlapping of the chosen fluorophores. Furthermore, red light excitation, outside the spectral range of the respective fluorophores, does not elicit a considerable change in the current of the blue-green single-nanowire dual-pixel device (Figure 5c). Figure 5e shows the spectrally selective electrical results of a similarly fabricated single-nanowire dual-pixel detector, modified with Alexa-430 and Alexa-610 fluorophores. These fluorophores display a considerable smaller spectral overlapping in comparison to the blue/green dual-pixel devices (Figure 5c). The effect of the reduced spectral overlap on the resulting electrical responses is evident, as expected. Notably, the large toolbox of available fluorophores allows the flexible design and fabrication of various spectral-specific detector combinations with controllable spectral width as well as improved spectral selectivity.

Conclusions. We demonstrated a novel concept of filter-free color detection at the nanometer scale. This new concept is based on the inherent high sensitivity of SiNW FETs, in conjunction with the spectral-specific absorbance and photo-induced charge transfer of various molecular fluorophores. We show that SGFET devices can be used to distinguish between different colors at the violet-red spectrum with a fwhm <100 nm bandwidth, which correlates to the absorbance spectra of the covalently attached fluorophores. In this work, we used commercially available Alexa-fluorophore derivatives, which are known to be highly stable; however, it is most possible that the use of other fluorophores, which absorb at different wavelengths and which have higher tendency to transfer electrons, could extend the spectral sensitivity and response time of the SGFETs. We demonstrate the nature of this spectral sensitivity by measuring the change in the surface potential of the excited fluorophore-modified devices, and show that the spectral-specific modulation of the SGFETs conductivity is due to the spectrally generated threshold voltage shift by repopulation of interface charged traps. The light illumination causes injection of minority charge carriers from the fluorophore to the SiNW body, and this excess charges enhance the capturing rate of electrons within the oxide traps, shifting the threshold voltage to positive values, and the conductivity increases. We present a way by which our SGFETs can be controlled spectrally without the need for a physical gate bias by using UV light, and be utilized to form spectrally controlled memory elements from suspended SGFET devices. Alternatively, the fabrication of such devices on a thinner oxide surface could be potentially used to enable fast erasing, using high voltage back-gate pulses in order to improve the decay time of the SGFETs. Furthermore, we developed the fabrication of ultrasmall multicolor single-nanowire detectors. Our highly compact nanometric SGFETs have the potential to be integrated into miniaturized optoelectronic applications, by virtue of their simple design. We have found that the chemical stability of our SGFETs can be greatly enhanced by the formation of a conformal ALD-based alumina thin shell, 8–15 nm thickness (Supplementary Figure S4b), forming fluorophore-embedded nanowire hybrid structures of remarkably high chemical stabilities under the harshest tested conditions, while maintaining the spectroelectrical activity of the devices and the spectroscopic attributes of the embedded fluorophore molecules (see Supplementary Figure S4c−f). This type of miniature optoelectronic devices would be of interest also in biological applications, to serve for example as in vivo fluorescence-signal detection, or as a color-sensitive artificial retina implants. After their alumina nanocapsulation, the SGFETs can also operate under wet physiological conditions. This allows the SGFETs to be incorporated into diverse bio-optoelectronic applications, where such compact sensors are highly required. All together, we believe that these compact SGFET devices would be highly useful in various future miniature optoelectronic applications.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications Web site. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.7b03873.

Materials and Methods describing all experimental procedures in detail. Alexa-fluorophores modification of RGBV sensing chips (Figure S1). SGFETs SiNW-restricted fluorophore modification scheme modification of RGBV sensing chips (Figure S2). Submicron blue-SGFET spectral-specific response (Figure S3). Durability of the fluorophore modified SiNWs in an alumina transparent shell (Figure S4). Field-effect of SGFETs following spectral-specific excitation (Figure S5). KPFP measurements of spectral specific charge accumulation of fluorophore modified SiNWs (Figure S6). Electrical
characteristics of suspended SGFETs (Figure S7). Long-term memory effect under ambient conditions of green-SGFETs (Figure S8). UV illumination of SGFETs (Figure S9). Spectral control of the “on” and “off” state of suspended green-SGFET nonvolatile memory elements (Figure S10). Electrical TC analysis of planar SGFETs (Figure S11) (PDF)

**AUTHOR INFORMATION**

**Corresponding Authors**
*E-mail: fernando@post.tau.ac.il.*

**E-mail:** yuval@ee.technion.ac.il.

**ORCID**

Fernando Patolsky: 0000-0002-1382-3357

**Notes**

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